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Biased continuous-time random walks for ordinary and equilibrium cases: facilitation of diffusion, ergodicity breaking and ageing

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We examine renewal processes with power-law waiting time distributions (WTDs) and non-zero drift *via* computing analytically and by computer simulations their ensemble and time averaged spreading characteristics. All possible values of the scaling exponent α are considered for the WTD $\psi(t) \sim 1/t^{1+\alpha}$. We treat continuous-time random walks (CTRWs) with $0 < \alpha < 1$ for which the mean waiting time diverges, and investigate the behaviour of the process for both ordinary and equilibrium CTRWs for $1 < \alpha < 2$ and $\alpha > 2$. We demonstrate that in the presence of a drift CTRWs with $\alpha < 1$ are ageing and non-ergodic in the sense of the non-equivalence of their ensemble and time averaged displacement characteristics in the limit of lag times much shorter than the trajectory length. In the sense of the equivalence of ensemble and time averages, CTRW processes with $1 < \alpha < 2$ are ergodic for the equilibrium and non-ergodic for the ordinary situation. Lastly, CTRW renewal processes with $\alpha > 2$—both for the equilibrium and ordinary situation—are always ergodic. For the situations $1 < \alpha < 2$ and $\alpha > 2$ the variance of the diffusion process, however, depends on the initial ensemble. For biased CTRWs with $\alpha > 1$ we also investigate the behaviour of the ergodicity breaking parameter. In addition, we demonstrate that for biased CTRWs the Einstein relation is valid on the level of the ensemble and time averaged displacements, in the entire range of the WTD exponent α.

I. Introduction

A. Anomalous diffusion

Anomalous diffusion processes feature a nonlinear scaling of the particle mean squared displacement (MSD) with the diffusion time,[1–13]

$$\mathrm{MSD}(t) = 2K_\beta t^\beta, \quad (1)$$

where K_β and β are the generalised diffusion coefficient and the MSD-based anomalous scaling exponent (in one dimension). Depending on the value of this exponent, one can distinguish subdiffusion ($0 < \beta < 1$), Brownian motion ($\beta = 1$), superdiffusion ($1 < \beta < 2$), ballistic motion ($\beta = 2$), and superballistic diffusion ($\beta > 2$), see ref. 3, 8 and 12. For modified processes of a Brownian-motion type—such as fractional Brownian motion and fractional Langevin equation motion[8,12]—the directions of particle displacements at consecutive time steps appear negatively and positively correlated for anti-persistent subdiffusion and persistent superdiffusion, respectively. Sub- and superdiffusive continuous-time random walks (CTRWs) are locally Markovian and thus do not contain correlations of increments of this type.[12,14]

Subdiffusive stochastic processes (for dispersive transport) are often used for the mathematical description of diffusion in crowded[9–11,15] and viscoelastic[14] environments of living biological cells. They have been applied, *e.g.*, to rationalise the properties of spreading of various macromolecules—such as proteins and nucleic acids—in the cell cytoplasm,[16–23] motions of DNA chromosomal loci,[25–31] diffusion of various ion channels along/in cell membranes,[32–36] heterogeneous subdiffusion[24] of short transmembrane proteins on the plasma membranes of T-cells,[37] diffusion of water molecules along[38] and of lipids within[39–42] lipid membranes, thermally-driven motion of lipids and insulin granules in cells,[43,44] diffusion of proteins involved in membrane crowding,[42,45–47] diffusion of polymeric mRNA molecules in viscoelastic cell cytoplasm,[20] and motion of colloidal tracers in entangled networks of actin filaments.[48] Some subdiffusive models with binding-unbinding kinetics also need to be mentioned.[49,50]

Superdiffusive processes (for enhanced or subballistic transport) are adapted to describe faster than Brownian or active motions. From the biological perspective, superdiffusive motions are detected for migrating bacteria, protozoa, and other microorganisms,[51,52]

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motor-driven[53,54] transport of virus particles along microtubuli inside living cells,[55] intracellular motor-driven motions of nano- and micro-particles,[56–59] persistent walks performed by motile cells and some microswimmers,[60–67] transient superdiffusion and ageing observed for amoeboid cells[62,68,69] and nematode worms,[70] active dynamics of neuronal messenger ribonucleoproteins,[71] as well as superdiffusion of self-propelled Brownian particles.[64,72,73] Generally, diffusion with accelerating diffusivity occurs in tilted/washboard potentials[74–77] and superdiffusion takes place in models of driven lattice Lorentz gas.[78] Other examples include geometry-induced superdiffusion,[79] tracer diffusion in plasma turbulence,[80] and field-induced superdiffusion[81] in actively-driven colloidal systems.[82,83]

Superballistic (or hyperdiffusive[84]) stochastic processes may occur at non-equilibrium conditions, *e.g.*, upon heat influx into the system, or for particle motion upon growing temperature.[85] Diffusion is accelerating also in tilted periodic potentials[86] and under non-equilibrium starting conditions. The classical Richardson–Batchelor relative spreading of particles in turbulent flows[87,88] should also be mentioned, as one of the first examples of superballistic diffusion, where MSD(t) $\sim t^3$ (see also ref. 1, 2, 67, 89 and 90 on superballistic diffusion, turbulence, and Lévy walks).

B. Continuous-time random walks

The paradigmatic CTRWs—recently completing a 50-year history[91]—is a class of renewal processes[92–97] featuring, *i.a.*, a well-studied case of power-law waiting time distributions (WTDs), $\psi(\tau) \sim \tau^{-1-\alpha}$. These processes with power-law WTDs are broadly and successfully used to describe the dynamics of transiently trapped particles,[2,67,98–102] for instance, in random energy landscapes, comb-like structures[2,5] and quenched trap models,[103] anomalous transport in disordered environments,[2,98] diffusion of tracers in ground water in porous and heterogeneous media,[104–110] transport properties in granular, fractal,[5,111] and glassy[112,113] media as well as in supercooled liquids.[114] For particle diffusion in random energy landscapes, *e.g.*, the power-law WTDs correspond to exponentially distributed energy barriers between neighbouring sites.[2,115] On a biological macroscale, CTRW-type processes with fat-tailed trapping times and of Lévy-type with broad jump-length distributions were discussed, *e.g.*, as possible mechanisms governing human mobility patters.[116–121] Applications of CTRWs for analysing the properties of financial time series is another important domain of research.[122]

Subdiffusive Montroll–Weiss CTRWs—with particle displacements MSD(t) $\sim t^\alpha$, with $0 < \alpha < 1$ and divergent mean waiting times[1,4,8,12,98,123–139]—form a widely used class of anomalous non-ergodic and ageing processes. Mathematically, subdiffusive CTRWs were shown to be non-ergodic in terms of non-equivalence of the ensemble and time averaged displacements even at long times[7,12,137] and to reveal strong deviations from predictions of the Boltzmann–Gibbs theory.[140] Ultraslow Sinai diffusion,[2,98] persistent Sinai-like diffusion in correlated Gaussian landscapes,[141] random walks with chaotically-driven bias,[142] as well as logarithmic diffusion in ageing jump processes[143] were also studied.

Ageing effects for the standard subdiffusive[12,137,139,144,145] and ultraslow[146] CTRWs were considered recently and identified, *i.a.*, in protein dynamics.[147] CTRWs with correlated trapping times were also investigated,[110,148,149] including effects of external constant and time-dependent force fields,[12,150,151] see also ref. 152–154. Noisy[155] and heterogeneous[156] walks, as well as CTRWs with coupled jump-lengths and waiting-time distributions,[3,149,157–159] walks in space- and time-dependent force fields,[160] CTRWs with periodicity and irreversible detachments[161] were studied too. On the level of ensemble averages, some scaling properties of CTRWs in the presence of bias and velocity fields were considered in ref. 130 and 162 (see also ref. 163 for the dynamics of Lévy walks in external fields, and the recent study of non-ergodicity for d-dimensional generalised Lévy walks[90]). Physically, the bias at each diffusion site can reflect inclined potential surfaces, existence of pressure gradients, *etc.*[98] For general analytical and numerical results for the time averaged MSD and ergodicity breaking of CTRWs we refer the reader to the studies.[7,8,12,134,150,164–173] Occupation times and weak ergodicity breaking (WEB) phenomena for biased CTRWs were considered also in ref. 174. As an important historical note, we mention that the superdiffusive behaviour of biased subdiffusive CTRW processes for $1/2 < \alpha < 1$ was already pointed out in the seminal studies of Shlesinger[125] and Scher and Montroll,[126] see also eqn (91) below.

C. Structure of the paper

The paper is organised as follows. In Section II we present the main equations of the model and their detailed derivations. We start in Section IIA with the Laplace transform expansions of the WTD, shortly list the details of simulations in Section IIB, and continue in Section IIC with presenting the general expressions for the ensemble and time averaged particle displacement characteristics. The general properties of renewal processes are used in Section IID to evaluate the two-point correlation functions for the walker positions in terms of the number of jumps taken. The main analytical results for the ensemble and time averaged particle displacements of biased CTRWs (see Fig. 1) are described in Section III, including a comparison to the findings from computer simulations. In Sections IIID, IIIB, and IIIA we derive the power-law scaling relations for particle spreading for biased CTRWs with $\alpha < 1$, $1 < \alpha < 2$, and $\alpha > 2$, correspondingly, both in leading and subleading orders in the (lag) time. For the limiting cases of $\alpha = 1, 2$ particle displacements acquire logarithmic corrections (not considered here). For all possible realisations of the WTD exponent α we check in Section IIIE the validity of the Einstein relation for the ensemble and time averaged displacements. We compute the degree of non-ergodicity for these

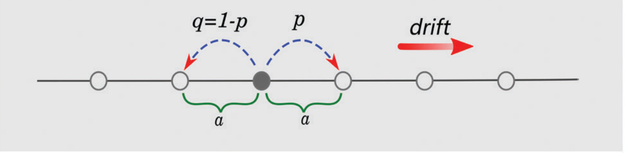

Fig. 1 Schematic representation of asymmetric particle jumps for biased CTRWs, with some parameters indicated.

biased walks, both in terms of the non-equivalence of the ensemble and time averaged observables as well as *via* assessing the ergodicity breaking parameter, EB, for the case $\alpha > 1$, Section IIIC. The results for the EB parameter are in agreement with simulations as well. In Section IV the main results and conclusions of the current study are summarised.

D. Summary of main results

Here, we overview the main results, see the phase diagram of Fig. 2 and Table 1. Case $0 < \alpha < 1$: for subdiffusive biased CTRWs we obtain, as expected, a non-ergodic and ageing diffusion stemming from the non-equivalence of the variance-based ensemble and time averaged displacements. These are denoted below as $\mathrm{Var}[x(\Delta)] = \langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2$ and $\overline{\delta\delta^2(\Delta)}$, respectively, and defined by eqn (13) and (15) below. In this case, $\mathrm{Var}[x(\Delta)]$ contains in the long-time limit the standard[125,126] subdiffusive term $\propto \Delta^\alpha$ and a field-induced contribution $\propto \Delta^{2\alpha}$. The time averaged displacement $\overline{\delta\delta^2(\Delta)}$ contains linear and superdiffusive terms, $\propto \Delta^1$ and $\propto \Delta^{1+\alpha}$, which both decrease in magnitude with the length of the trajectory T as $\propto 1/T^{1-\alpha}$. We mention here that the WTD exponent α differs from the anomalous diffusion exponents realised for the ensemble and time averaged particle–displacement characteristics.

Case $1 < \alpha < 2$: for superdiffusive exponents of WTDs we observe some marked differences for ordinary *versus* equilibrium processes. Namely, for ordinary processes (initiated simultaneously with the start of the measurement) $\mathrm{Var}[x(\Delta)]$ contains linear and subdiffusive terms, $\propto \Delta^1$ and $\propto \Delta^{2-\alpha}$, as well as a field-induced

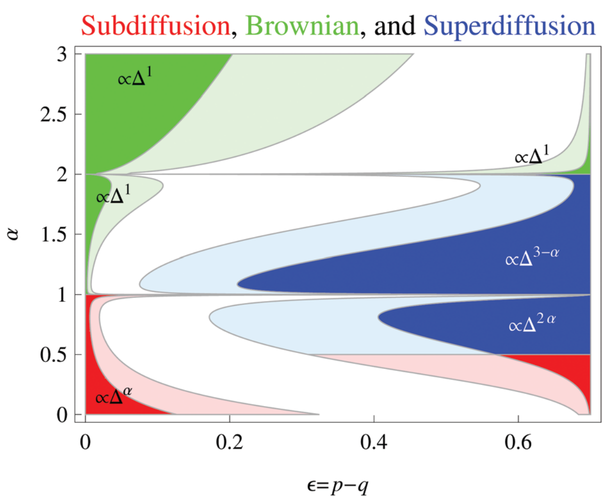

Fig. 2 Phase diagram of scaling regimes of particle spreading for equilibrium processes, shown on the example of ensemble averaged variance-based displacements. The leading scaling terms in $\langle x_{\mathrm{eq}}{}^2(\Delta)\rangle - \langle x_{\mathrm{eq}}(\Delta)\rangle^2$ are shown in the plane of the WTD exponent α and asymmetry parameter $\varepsilon = p - q$. Dark and light colours in each region of the phase space correspond to, respectively, at least 99% and 90% of particle displacements dominated by a given scaling term in respective $\langle x_{\mathrm{eq}}{}^2(\Delta)\rangle - \langle x_{\mathrm{eq}}(\Delta)\rangle^2$ expressions. The graph is based on the analytical results of eqn (54), (74) and (91) obtained in the main text, evaluated for $p = 0.7$ (as in other plots below) and for the lag time of $\Delta = 10^5$ (long-time limit). Note that sharp variations close to the boundary values $\alpha = 1$ and $\alpha = 2$ may get smoothened when respective logarithmic corrections are computed (not shown).

Table 1 Summary of the scaling results for biased CTRWs for all values of the WTD exponent α: from left to right, the mean and variance of the number of particle jumps, the variance-based ensemble and time averaged displacements, the respective ergodicity breaking parameters, weak ergodicity breaking in terms of the equivalence of the ensemble and time averaged particle displacements, the validity of the Einstein relation for the ensemble and time averaged moments, the dispersion-to-mean ratio, and, finally, the corresponding equations for these quantities/relations in the main text

Process	$\langle N(t)\rangle$	$\langle N(t)^2\rangle - \langle N(t)\rangle^2$	$\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2$	$\left\langle\overline{\delta\delta^2(\Delta,T)}\right\rangle$	$\delta\mathrm{EB}(T)$	WEB	Ensemble/time averaged Einstein relation	$\frac{\sqrt{\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2}}{\langle x(\Delta)\rangle}$	Equations
$\alpha > 2$, or.	$\sim \frac{t}{\mu_\alpha} + \mathrm{Const.}$	$\sim \frac{\sigma_\alpha^2}{\mu_\alpha^2}\frac{t}{\mu_\alpha}$	$\sim O(\Delta^1)$	$\sim O(\Delta^1)$	$\sim O(1/T)$	No	Holds/holds	$\sim O(\Delta^{-1/2})$	(43), (46), (48), (49), (83), (21), (97)/(99) and (100)
$\alpha > 2$, eq.	$\frac{t}{\mu_\alpha}$	$\sim \frac{\sigma_\alpha^2}{\mu_\alpha^2}\frac{t}{\mu_\alpha}$	$\sim O(\Delta^1)$	$\sim O(\Delta^1)$	$\sim O(1/T)$	No	Holds/holds	$\sim O(\Delta^{-1/2})$	(50), (52), (54), (55), (83), (21), (97)/(99) and (100)
$1 < \alpha < 2$, or.	$\sim \frac{t}{\mu_\alpha} + O(t^{2-\alpha})$	$\sim O(t^{3-\alpha})$	$\sim O(\Delta^1) + O(\Delta^{2-\alpha}) + (p-q)^2 O(\Delta^{3-\alpha})$	$\sim O(\Delta^1) + O\left(\frac{\Delta^1}{T^{\alpha-1}}\right) + (p-q)^2 O(\Delta^{3-\alpha})$	$\sim O\left(\frac{1}{T^{\alpha-1}}\right)$	Yes	Holds/holds	$\sim O(\Delta^{-(\alpha-1)/2})$	(60), (62), (65), (66), (83), (21), (97)/(99) and (100)
$1 < \alpha < 2$, eq.	$\frac{t}{\mu_\alpha}$	$\sim O(t^{3-\alpha})$	$\sim O(\Delta^1) + (p-q)^2 O(\Delta^{3-\alpha})$	$\sim O(\Delta^1) + (p-q)^2 O(\Delta^{3-\alpha})$	$\sim O\left(\frac{1}{T^{\alpha-1}}\right)$	No	Holds/holds	$\sim O(\Delta^{-(\alpha-1)/2})$	(69), (71), (74), (75), (83), (21), (97)/(99) and (100)
$0 < \alpha < 1$	$\sim \frac{t^\alpha}{k_\alpha \Gamma(1+\alpha)}$	$\sim O(t^{2\alpha})$	$\sim O(\Delta^\alpha) + (p-q)^2 O(\Delta^{2\alpha})$	$\sim O\left(\frac{\Delta}{T^{1-\alpha}}\right) + (p-q)^2 O\left(\frac{\Delta^{1+\alpha}}{T^{1-\alpha}}\right)$	—	Yes	Holds/holds	$\left(\frac{2[\Gamma(1+\alpha)]^2}{\Gamma(1+2\alpha)} - 1\right)^{1/2} = \mathrm{Const.}$	(87), (89), (91), (93), (83), (21), (97)/(99) and (100)

superdiffusive term $\propto \Delta^{3-\alpha}$. The time averaged displacement $\overline{\delta\delta^2(\Delta)}$ for ordinary processes is demonstrated to contain terms linear and superdiffusive in the lag time, namely $\propto \Delta^1$, $\propto \Delta^1/T^{\alpha-1}$, and $\propto \Delta^{3-\alpha}$. The last term is induced by the bias present in the system. For equilibrium CTRWs with WTD exponents in the range $1 < \alpha < 2$ we find that $\mathrm{Var}[x(\Delta)]$ contains linear and superdiffusive terms, $\propto \Delta^1$ and $\propto \Delta^{3-\alpha}$. Also, ensemble averaged displacements for equilibrium processes are identical to time averaged displacements, $\overline{\delta\delta^2(\Delta)}$, indicating ergodicity ($\overline{\delta\delta^2(\Delta)}$ contains no dependence on the trace length T in this case). Biased CTRWs with superdiffusive exponents α approach ergodicity anomalously slow, so that the variance-based ergodicity breaking parameter, see for definition eqn (77) below, decays with the trace length as $\delta\mathrm{EB} \sim 1/T^{\alpha-1}$.

Case $\alpha > 2$: for biased CTRWs with superballistic exponents both ensemble and time averaged displacements of the particles contain terms linear in time and lag time (bias-free and field-induced terms, respectively). In this case, ensemble and time averaged displacements are identical, indicating ergodicity. The ergodicity breaking parameter decays as $\delta\mathrm{EB} \sim 1/T$ with the trace length T, similarly as for a number of other anomalous diffusion processes.[12]

Table 1 also contains some scaling relations for the Scher–Montroll transport parameter—the ratio of dispersion to mean defined as $\eta(\Delta)$ in eqn (100), for an ensemble of particles spreading in external fields—for all realisations of WTD scaling exponents α as outlined above. Biased CTRWs for subdiffusive WTD exponents α feature a constant value of this coefficient.[125,126] For superdiffusive exponents $1 < \alpha < 2$ the scaling is shown below to be $\propto \Delta^{-(\alpha-1)/2}$. Finally, the relation $\eta(\Delta) \sim \Delta^{-1/2}$ is found in the long-time limit for biased CTRWs with WTD exponents $\alpha > 2$.

II. Main equations and their derivations

A. Waiting time distributions

Here, we consider a non-equilibrium CTRW stochastic process with a bias, *e.g.*, due to the presence of an external field. In such a system the probability for a Brownian particle to jump to the left,

$$q = 1 - p, \tag{2}$$

and to the right, p, are not equal, see the schematics in Fig. 1. We are interested below primarily in the effects of a bias on the ensemble and time averaged particle displacements. We consider the displacement by one lattice unit a at each particle jump, that is the step-size distribution is $\lambda(x) = 1/2\delta(|x| - a)$, where $\delta(x)$ is the Dirac delta-function. On each site random trapping times are drawn independently (renewal property) and distributed in the long-time limit identically on all sites according to the standard power-law (or Pareto-like) WTD,[7,8,12,175]

$$\psi(\tau) = \frac{\alpha\tau_0^\alpha}{\tau^{1+\alpha}}, \tag{3}$$

with a microscopic time-scale τ_0 (scaling factor) and WTD exponent α. For the normalised WTD given by eqn (3) with $\alpha > 1$, the mean waiting time $\langle\tau_{\mathrm{wait}}\rangle$ exists,

$$\mu_\alpha = \int_{\tau_0}^{\infty} \tau\psi(\tau)\mathrm{d}\tau = \frac{\alpha}{\alpha-1}\tau_0. \tag{4}$$

This is in contrast to the case $\alpha < 1$ with divergent mean waiting time, when the process is scale-free and the long-time dynamics is governed by rare but long trapping events, as, for instance, measured in ref. 32. For $\alpha > 2$ the variance of waiting times σ_α^2 with the WTD (3) also attains a finite value, namely

$$\sigma_\alpha^2 = \int_{\tau_0}^{\infty} (\tau - \mu_\alpha)^2\psi(\tau)\mathrm{d}\tau = \frac{\alpha}{\alpha-2}\tau_0^2 - \mu_\alpha^2. \tag{5}$$

The Laplace transform of the WTD (3) is given by

$$\hat{\psi}(s) \equiv \mathcal{L}_s\{\psi(\tau)\} = \int_{\tau_0}^{\infty} \mathrm{e}^{-\tau s}\psi(\tau)\mathrm{d}\tau. \tag{6}$$

This can be written to leading order for $s\tau_0 \ll 1$—that is, in the limit of long diffusion times, $t/\tau_0 \to \infty$—for different choices of the exponent α as follows

$$\hat{\psi}(s) \approx \begin{cases} 1 - k_\alpha s^\alpha, & 0 < \alpha < 1 \\ 1 - \mu_\alpha s + k_\alpha s^\alpha, & 1 < \alpha < 2 \\ 1 - \mu_\alpha s + \frac{1}{2}(\sigma_\alpha^2 + \mu_\alpha^2)s^2, & \alpha > 2 \end{cases}. \tag{7}$$

In this expansion, the constant k_α assumes the values

$$k_\alpha(0 < \alpha < 1) = \Gamma(1-\alpha)\tau_0^\alpha,$$
$$k_\alpha(1 < \alpha < 2) = |\Gamma(1-\alpha)|\tau_0^\alpha. \tag{8}$$

We mention here, for the sake of completeness, that the expansions for $\alpha < 1$ and $1 < \alpha < 2$ are obtained using, correspondingly, eqn (3.14) and (3.16) from ref. 175, see also ref. 178. The expansion for $\alpha > 2$ follows from the Taylor expansion of the exponent in eqn (6), see also eqn (1.7) and (1.8) in ref. 97. Without a bias, broad and fat-tailed distributions with $\alpha < 1$ give rise to a power-law subdiffusion.[8,12]

B. Details of numerical simulations

In Fig. 3 we exemplify typical trajectories generated in computer simulations of different biased CTRWs. To generate an ensemble of waiting times distributed according to the power law (3), first a random variable X with the uniform distribution on the interval [0,1] is seeded, from which a random variable Y is constructed in simulations as

$$Y(X) = X^{-1/\alpha}. \tag{9}$$

Therefore, the required WTD is given by $\psi(\tau) = \alpha\tau^{-1-\alpha}$ for $\tau > 1$ and it is 0 otherwise (we set $\tau_0 = 1$ in all the simulations).

C. Ensemble and time averaged displacements of continuous-time random walks with a drift: general expressions

For CTRWs with a bias, we define the analogues of the ensemble and time averaged MSDs of the particles *via* the respective variances.

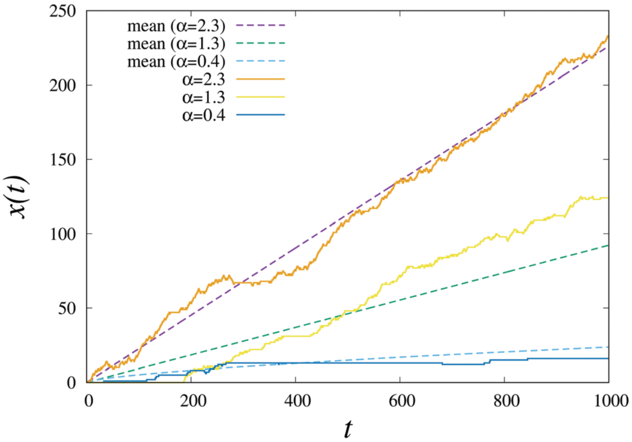

Fig. 3 Typical particle trajectories $x(t)$ obtained from computer simulations for different biased CTRWs. The values of the exponent α and analytical mean particle displacements (10) are given in the plot. Other parameters for this and other figures are: the lattice constant is unity (a = 1) and τ_0 = 1. The long stalling events for the subdiffusive case are to be noted, in contrast to rapid position changes of the walker for superdiffusive situations.

This procedure "removes" non-zero means from the observables and enables us to derive the scaling relations for the particle spreading properties with respect to the mean in the long-time limit. For the first moment of particle displacements after time Δ—with the initial condition $x(0) = 0$—one gets

$$\langle x(\Delta)\rangle = a(p-q)\langle N(\Delta)\rangle \neq 0. \quad (10)$$

Here $N(\Delta)$ is the number of jumps of the walker up to time Δ, see Fig. 1 and 3. Note that relation (10) follows from the fact that the total particle displacement after N jumps (or renewals) is the sum of identically distributed independent random variables. The mean displacement after one jump is

$$\langle \delta x\rangle = a(p-q), \quad (11)$$

so one has

$$\left\langle \sum_{j=1}^{N} \delta x_j \right\rangle = \langle N(\Delta)\rangle\langle \delta x\rangle. \quad (12)$$

Here and below, the angular brackets stand for ensemble averaging, while the overline denotes time averaging, consistent with our previous notations.[8,12] The variance-based displacement—or the second centred moment—is given by (using Wald's formula[96])

$$\begin{aligned}\mathrm{Var}[x(\Delta)] &= \mathrm{Dispersion}^2[x(\Delta)] \equiv \langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2 \\ &= (p-q)^2a^2(\langle N^2(\Delta)\rangle - \langle N(\Delta)\rangle^2) + 4pqa^2\langle N(\Delta)\rangle. \quad (13)\end{aligned}$$

Here, we used the relation (see ref. 95 and 96, and also eqn (9) in ref. 162)

$$\begin{aligned}\left\langle \left(\sum_{j=1}^{N} \delta x_j\right)^2 \right\rangle - \left\langle \sum_{j=1}^{N} \delta x_j \right\rangle^2 &= \langle N(\Delta)\rangle\left(\langle \delta x^2\rangle - \langle \delta x\rangle^2\right) \\ &+ \langle \delta x\rangle^2\left(\langle N^2(\Delta)\rangle - \langle N(\Delta)\rangle^2\right)\end{aligned} \quad (14)$$

and the fact that for displacements after one step $\langle \delta x^2\rangle = a^2$.

The ensemble averaged time averaged variance-based displacements can similarly be defined with respect to the mean of the increments as

$$\left\langle \overline{\delta\delta^2(\Delta)} \right\rangle = \frac{1}{T-\Delta}\int_0^{T-\Delta} \left\langle [(x(t+\Delta)-x(t)) - \langle x(t+\Delta)-x(t)\rangle]^2 \right\rangle \mathrm{d}t, \quad (15)$$

where Δ is the lag time along the trajectory of length T. Here, the new notation $\overline{\delta\delta^2(\Delta)}$ symbolises the respective deviations from the mean increments of particle positions in the integrand of (15), as compared to the standard definition of the time averaged MSD for drift-free processes,[8,12]

$$\overline{\delta^2(\Delta)} = \frac{1}{T-\Delta}\int_0^{T-\Delta} [x(t+\Delta)-x(t)]^2 \mathrm{d}t. \quad (16)$$

The latter is routinely used to analyse and interpret, *e.g.*, single-particle tracking data.[8,12,20,179,180] We consider the time averaged properties in the limit $\Delta \ll T$, the standard limit used for other anomalous diffusion processes.[8,12] The integrand of (15) equals the variance of particle increments with respect to the mean, that can be written as

$$\begin{aligned}&\langle (x(t+\Delta)-x(t))^2\rangle - \langle x(t+\Delta)-x(t)\rangle^2 \equiv \mathrm{Var}[\delta x(t,t+\Delta)] \\ &= \langle x(t+\Delta)^2\rangle - \langle x(t+\Delta)\rangle^2 + \langle x(t)^2\rangle - \langle x(t)\rangle^2 - 2\{\langle x(t+\Delta)x(t)\rangle \\ &\qquad - \langle x(t+\Delta)\rangle\langle x(t)\rangle\}. \quad (17)\end{aligned}$$

Using the fact that the particle position correlator has the form (see also ref. 96)

$$\langle x(t+\Delta)x(t)\rangle = a^2\langle N(t)\rangle + a^2(p-q)^2\langle N(t)(N(t+\Delta)-1)\rangle, \quad (18)$$

we get

$$\begin{aligned}&\langle (x(t+\Delta)-x(t))^2\rangle - \langle x(t+\Delta)-x(t)\rangle^2 = 4pqa^2[\langle N(t+\Delta)\rangle \\ &- \langle N(t)\rangle] - a^2(p-q)^2[\langle N(t+\Delta)\rangle - \langle N(t)\rangle]^2 \\ &+ a^2(p-q)^2[\langle N(t+\Delta)^2\rangle - \langle N(t)^2\rangle - 2\langle N(t)(N(t+\Delta)-N(t))\rangle]. \quad (19)\end{aligned}$$

In what follows we use the definition of ergodicity of a stochastic process in the Boltzmann–Khinchin sense[168] in the limit of $\Delta/T \ll 1$ as the equivalence of the ensemble and time averaged MSDs,[7,8,12,181]

$$\mathrm{MSD}(\Delta) = \left\langle [x(\Delta)-x(0)]^2 \right\rangle = \lim_{T/\Delta\to\infty} \overline{\delta^2(\Delta)}, \quad (20)$$

(and not any stricter definition such as mixing properties). Analogously, we call processes with non-zero mean displacements ergodic[7,8,12,181] if for short lag times and long trajectories the condition

$$\left\langle x^2(\Delta)\right\rangle - \left\langle x(\Delta)^2\right\rangle = \lim_{T/\Delta\to\infty} \overline{\delta\delta^2(\Delta)} \quad (21)$$

is satisfied.[8,12] We refer the reader to ref. 182 for the discussion of the effects of initial ensemble onto the ensemble and time averaged properties of some renewal processes. Note also here that comparison of higher-order time and ensemble averaged moments of particle displacements as a check of ergodicity breaking can also be employed, see ref. 144 and 152.

As we quantify below, for some choices of α we reach fully ergodic behaviour for long times, whereas, for instance, for $\alpha < 1$ the process is inherently non-ergodic and ageing[12,137,139] even in the long-time limit. Although some results for the ensemble averaged displacements of field-induced CTRWs are available,[98,130,139,150,162,177] the main focus of the current study is on the time averaged properties of particle spreading with respect to the mean, and also on quantifying the non-ergodicity of the process. This *per se* presents a considerable mathematical challenge.

D. Renewal properties and two-point correlation functions: ordinary and equilibrium processes

The diffusive properties of CTRWs can be understood from the theoretical concepts of renewal processes, see, *e.g.*, ref. 92–97, 177 and 178. Below, we use the renewal theory to compute the first and second moments of the number of particle jumps, as well as the correlator $\langle N(t)(N(t+\Delta)-N(t))\rangle$ which enter eqn (10), (13), (19), and (15) for the ensemble and time averaged displacements. We start with defining S_r as the sum of waiting times after r steps,

$$S_r = \sum_{j=1}^{r} \tau_j. \tag{22}$$

Then, using $\mathcal{K}_r$ for the cumulative distribution of S_r, for the probability of exactly r jumps occurred during the time interval $[0, t]$ one gets

$$\Pr[N(t) = r] = \mathcal{K}_r(t) - \mathcal{K}_{r+1}(t) > 0. \tag{23}$$

Then, the probability generating function—we here follow the derivations of Section 3.2 of Cox's classical book[92]—becomes

$$G(t,\zeta) = \sum_{r=0}^{\infty} \zeta^r \Pr[N(t) = r] = 1 + \sum_{r=1}^{\infty} \zeta^{r-1}(\zeta - 1)\mathcal{K}_r(t). \tag{24}$$

In Laplace space, using the relation between the cumulative probability distribution and the associated probability density $k_r(t)$

$$\hat{\mathcal{K}}_r(s) = \hat{k}_r(s)/s, \tag{25}$$

one obtains the general expression

$$\hat{G}(s,\zeta) = \frac{1}{s} + \frac{1}{s}\sum_{r=1}^{\infty} \zeta^{r-1}(\zeta - 1)\hat{k}_r(s). \tag{26}$$

In what follows, we split the consideration for the two typical renewal processes, namely for the ordinary (subscript "or" below) and equilibrium (subscript "eq") processes.[92,178] The fundamental difference between them is the fact that "an equilibrium renewal process can be regarded as an ordinary renewal process in which the system has been running a long-time before it is first observed".[92] Therefore, for the ordinary renewal process—which is physically initiated at the start of the observation, at $t = 0$—the probability density functions for the distribution of all waiting times are identical. This yields[92,95]

$$\hat{k}_{r,\mathrm{or}}(s) = \hat{k}_{r-1,\mathrm{or}}(s) \times \hat{\psi}(s) = [\hat{\psi}(s)]^r, \tag{27}$$

while for the equilibrium renewal process the first waiting time follows a different distribution, namely[92]

$$\hat{k}_{1,\mathrm{eq}}(s) = \frac{1 - \hat{\psi}(s)}{\mu s}, \tag{28}$$

that gives rise to (see also Section 2.5 in ref. 92)

$$\hat{k}_{r,\mathrm{eq}}(s) = \hat{k}_{1,\mathrm{eq}}(s) \times \left[\hat{\psi}(s)\right]^{r-1} = \frac{(1 - \hat{\psi}(s))\left[\hat{\psi}(s)\right]^{r-1}}{\mu s}. \tag{29}$$

Clearly, equilibrium renewal processes can only be considered if the mean waiting time μ exists. In the subdiffusive case ageing renewal theory has to be applied.[97,136,139]

Inserting relations (27) and (29) into (26) we get the probability generating function for the ordinary and equilibrium renewal processes as, respectively,

$$\hat{G}_{\mathrm{or}}(s,\zeta) = \frac{1 - \hat{\psi}(s)}{s[1 - \zeta\hat{\psi}(s)]} \tag{30}$$

and

$$\hat{G}_{\mathrm{eq}}(s,\zeta) = \frac{1}{s} + \frac{\zeta - 1}{\mu s}\hat{G}_{\mathrm{or}}(s,\zeta). \tag{31}$$

The first and second moments of the number of jumps of the walker can be obtained from the probability generating function[95,96,178] as

$$\begin{aligned} \langle N(t)\rangle &= \mathcal{L}_s^{-1}\left\{\left.\frac{\partial \hat{G}(s,\zeta)}{\partial \zeta}\right|_{\zeta=1}\right\}, \\ \langle N^2(t)\rangle &= \langle N(t)\rangle + \mathcal{L}_s^{-1}\left\{\left.\frac{\partial^2 \hat{G}(s,\zeta)}{\partial \zeta^2}\right|_{\zeta=1}\right\}. \end{aligned} \tag{32}$$

Here $\mathcal{L}_s^{-1}$ denotes the inverse Laplace transform over the respective variable. Using the generating functions (30) and (31), for the ordinary and equilibrium renewal processes we arrive at, respectively, (see ref. 92 for a detailed derivation; see also the supplement of ref. 118)

$$\begin{aligned} \langle N_{\mathrm{or}}(t)\rangle &= \mathcal{L}_s^{-1}\left\{\frac{\hat{\psi}(s)}{s\left[1 - \hat{\psi}(s)\right]}\right\}, \\ \langle N_{\mathrm{or}}{}^2(t)\rangle &= \langle N_{\mathrm{or}}(t)\rangle + \mathcal{L}_s^{-1}\left\{\frac{2\left[\hat{\psi}(s)\right]^2}{s\left[1 - \hat{\psi}(s)\right]^2}\right\} \end{aligned} \tag{33}$$

and

$$\begin{aligned} \langle N_{\mathrm{eq}}(t)\rangle &= \mathcal{L}_s^{-1}\left\{\frac{1}{\mu s^2}\right\}, \\ \langle N_{\mathrm{eq}}{}^2(t)\rangle &= \langle N_{\mathrm{eq}}(t)\rangle + \mathcal{L}_s^{-1}\left\{\frac{2\hat{\psi}(s)}{\mu s^2\left[1 - \hat{\psi}(s)\right]}\right\}. \end{aligned} \tag{34}$$

Inserting these expressions into eqn (13) we compute the variance-based ensemble averaged displacements.

To evaluate the time averaged variance-based displacements (19), we obtain the relation for $\langle N(t)(N(t+\Delta) - N(t))\rangle$ in terms of the double Laplace transform with respect to u_1 and s_1 performing the calculations according to the scheme developed in ref. 177 and 183. The variables u_1 and s_1 are related to the diffusion time t and lag time Δ, correspondingly, and the indices are used for the Laplace operator in order not to mix with the analysis above, where the Laplace variable s was related to t. Specifically, using eqn (18) and (19) of ref. 177 and eqn (5) and (6) in ref. 183, we get the joint two-point probability function for the walker to make $N(t)$ jumps during time $[0, t]$ and $N(t+\Delta) - N(t)$ jumps during time interval $[t, t+\Delta]$,

$$\hat{\hat{P}}_{N(t),N(t+\Delta)-N(t)}(u_1,s_1) = \frac{\left[\hat{\psi}(u_1)\right]^{N(t)}\left[\hat{\psi}(s_1)\right]^{N(t+\Delta)-N(t)-1}}{s_1(u_1-s_1)} \times \left[1-\hat{\psi}(s_1)\right]\left[\hat{\psi}(s_1)-\hat{\psi}(u_1)\right]. \tag{35}$$

Also, based on the renewal property of CTRWs, one can write the probability for the walker to perform $N(t)$ jumps up to time t and no jumps from time t to time $t+\Delta$ as

$$\hat{\hat{P}}_{N(t),0}(u_1,s_1) = \frac{\left[\hat{\psi}(u_1)\right]^{N(t)}}{s_1} \times \left[\frac{1-\hat{\psi}(u_1)}{u_1} - \frac{\hat{\psi}(s_1)-\hat{\psi}(u_1)}{u_1-s_1}\right]. \tag{36}$$

This expression is used to normalise the overall probability distribution, namely

$$\sum_{N(t)=0}^{\infty}\sum_{N(t+\Delta)-N(t)=1}^{\infty}\hat{\hat{P}}_{N(t),N(t+\Delta)-N(t)}(u_1,s_1) + \sum_{N(t)=0}^{\infty}\hat{\hat{P}}_{N(t),0}(u_1,s_1) = \frac{1}{s_1u_1}. \tag{37}$$

The correlator of particle jump numbers can then be expressed in terms of the joint probabilities (35) and (36) as the standard mean over independent random variables $N(t)$ and $N(t+\Delta) - N(t)$, namely

$$\langle N(t)(N(t+\Delta)-N(t))\rangle = \sum_{N(t)=0}^{\infty}\sum_{N(t+\Delta)-N(t)=0}^{\infty} N(t)[N(t+\Delta)-N(t)] \times \mathcal{L}_{u_1}^{-1}\mathcal{L}_{s_1}^{-1}\left\{\hat{\hat{P}}_{N(t),N(t+\Delta)-N(t)}(u_1,s_1)\right\}. \tag{38}$$

For the ordinary and equilibrium renewal processes we then obtain, respectively,

$$\langle N_{\text{or}}(t)(N_{\text{or}}(t+\Delta)-N_{\text{or}}(t))\rangle = \mathcal{L}_{u_1}^{-1}\mathcal{L}_{s_1}^{-1}\left\{\frac{\hat{\psi}(u_1)\left[\hat{\psi}(s_1)-\hat{\psi}(u_1)\right]}{s_1(u_1-s_1)\left[1-\hat{\psi}(u_1)\right]^2\left[1-\hat{\psi}(s_1)\right]}\right\} \tag{39}$$

and

$$\left\langle N_{\text{eq}}(t)(N_{\text{eq}}(t+\Delta)-N_{\text{eq}}(t))\right\rangle = \mathcal{L}_{u_1}^{-1}\mathcal{L}_{s_1}^{-1}\left\{\frac{\hat{\psi}(s_1)-\hat{\psi}(u_1)}{\mu u_1 s_1(u_1-s_1)\left[1-\hat{\psi}(u_1)\right]\left[1-\hat{\psi}(s_1)\right]}\right\}. \tag{40}$$

Note that to derive eqn (40) we took into account the different distribution of the waiting time for the first jump, eqn (28). The derivations of eqn (39) are given in eqn (7) of ref. 183; see also eqn (8.6) of ref. 97 (for the case $\alpha < 1$). Performing the long-time expansions of eqn (33) and (39) for the ordinary and of eqn (34) and (40) for the equilibrium renewal processes we have all the ingredients to evaluate the long-time variance-based ensemble and time averaged particle displacements for each choice of the WTD exponent α.

Note that for ensemble averaged quantities long diffusion times are assumed, which are compared in eqn (48) and similar equations below with the lag time Δ for the time averaged quantities. For the latter, in the displacement increments Δ is assumed to be much shorter than the running time t along the trajectory, see eqn (45) below. Therefore, the comparison of the variance-based expressions (13) and (15), as we perform below for each choice of α, is a mathematically valid procedure when the condition

$$\tau_0 \ll \Delta \ll \{t,T\} \tag{41}$$

is satisfied (for which τ_0 should be the shortest time scale in the problem).

Below, we use the long-time $\hat{\psi}(s)$ expansions (7) for different α values. To take the inverse Laplace transform, we use the (strong) Tauberian theorem (see, *e.g.*, Ch. XIII.5 in ref. 94, Ch. 5.1.5 in ref. 93, Ch. 2.2 in ref. 95, and ref. 184) to approximate the Laplace transform of the form $\hat{\phi}(s) \sim L(1/s)s^{-\rho}$ at $s \to 0$ *via* the long-time scaling

$$\phi(t) \sim t^{\rho-1}L(t)/\Gamma(\rho), \tag{42}$$

where $L(t)$ is a slowly varying function at $t \to \infty$, and $\Gamma(x)$ is the Gamma function.

III. Main results: particle spreading characteristics and non-ergodicity

We now connect the results of the renewal theory from Section II to the physically measurable quantities for the relevant ranges of exponent α.

A. Displacements for $\alpha > 2$: ordinary and equilibrium processes

For WTDs of the form (3) with $\alpha > 2$ both μ_α and σ_α^2 attain finite values and the consideration is fairly simple, so we start with this scenario. Below, the results are presented separately for the ordinary and equilibrium case. For the ordinary process, the leading order terms in the long-time limit ($t \to \infty$) for the average number of steps and the second moment follow from eqn (33) using (42),

$$\begin{aligned}\langle N_{\text{or}}(t)\rangle &= \frac{\langle x(t)_{\text{or}}\rangle}{a(p-q)} \sim \mathcal{L}_s^{-1}\left\{\frac{1+s(\sigma_\alpha^2-\mu_\alpha^2)/(2\mu_\alpha)}{\mu_\alpha s^2}\right\}\\ &\sim \frac{t}{\mu_\alpha}+\frac{\sigma_\alpha^2-\mu_\alpha^2}{2\mu_\alpha^2}\end{aligned} \tag{43}$$

and

$$\langle N_{\text{or}}^2(t)\rangle \sim \frac{t^2}{\mu_\alpha^2}+\frac{2\sigma_\alpha^2-\mu_\alpha^2}{\mu_\alpha^2}\frac{t}{\mu_\alpha}. \tag{44}$$

Similarly, the expressions for the variance and the correlator of the number of steps (39) in the limit

$$u_1 \ll s_1 \tag{45}$$

can be obtained as, respectively,

$$\langle N_{\text{or}}^2(t)\rangle - \langle N_{\text{or}}(t)\rangle^2 \sim \frac{\sigma_\alpha^2}{\mu_\alpha^2}\frac{t}{\mu_\alpha} \tag{46}$$

and

$$\langle N_{\text{or}}(t)(N_{\text{or}}(t+\Delta)-N_{\text{or}}(t))\rangle \sim \frac{t\Delta}{\mu_\alpha^2}+\frac{(\sigma_\alpha^2-\mu_\alpha^2)\Delta}{2\mu_\alpha^3}. \tag{47}$$

Then, the temporal evolution of the ensemble averaged particle displacements (13) and the trajectory-local displacement increments (19) in the long-time limit are

$$\begin{aligned}\langle x_{\text{or}}^2(\Delta)\rangle - \langle x_{\text{or}}(\Delta)\rangle^2 &\sim 4pqa^2\frac{\Delta}{\mu_\alpha}\\ &+ a^2(p-q)^2\frac{\sigma_\alpha^2}{\mu_\alpha^2}\frac{\Delta}{\mu_\alpha}\\ &\sim \langle (x_{\text{or}}(t+\Delta)-x_{\text{or}}(t))^2\rangle - \langle x_{\text{or}}(t+\Delta)-x_{\text{or}}(t)\rangle^2.\end{aligned} \tag{48}$$

Therefore, the spreading of particles with respect to the mean values is always linear in time Δ, indicative of Brownian transport properties. For symmetric walks with $p = q$ the second term in expression (48) disappears. Importantly, the particle displacements do not contain any t-dependence, indicating the stationarity of displacement increments. This gives rise to the equivalence of the time and ensemble averaged variance-based displacements,

$$\left\langle \overline{\delta\delta^2(\Delta)_{\text{or}}}\right\rangle = \langle x_{\text{or}}^2(\Delta)\rangle - \langle x_{\text{or}}(\Delta)\rangle^2, \tag{49}$$

rendering the underlying $\alpha > 2$ diffusion process ergodic.

For equilibrium[96] CTRW processes with power-law WTDs with exponent values $\alpha > 2$, performing analogously the inverse Laplace transform of eqn (34), we get

$$\langle N_{\text{eq}}(t)\rangle \sim \frac{t}{\mu_\alpha}, \tag{50}$$

$$\langle N_{\text{eq}}^2(t)\rangle \sim \frac{t^2}{\mu_\alpha^2}+\frac{\sigma_\alpha^2}{\mu_\alpha^2}\frac{t}{\mu_\alpha}, \tag{51}$$

$$\langle N_{\text{eq}}^2(t)\rangle - \langle N_{\text{eq}}(t)\rangle^2 \sim \frac{\sigma_\alpha^2}{\mu_\alpha^2}\frac{t}{\mu_\alpha}, \tag{52}$$

and

$$\langle N_{\text{eq}}(t)(N_{\text{eq}}(t+\Delta)-N_{\text{eq}}(t))\rangle \sim \frac{t\Delta}{\mu_\alpha^2}. \tag{53}$$

We refer the reader to Sections 4.1, 4.2, and 4.5 in ref. 92 for the derivation of the mean and variance of the number of renewals for a general ordinary and equilibrium process (when the variance exists). In the equilibrium situation, the mean and the variance of the number of particle jumps—see also Section 3.3 of ref. 92 for a derivation—are the same as for the bias-free CTRW process, see, *e.g.*, ref. 41. The scaling results for $\langle N(t)\rangle$ and $\langle N^2(t)\rangle - \langle N(t)\rangle^2$ were outlined for such α values in ref. 162, see also ref. 107.

Thus, eqn (50), (51) and (53) yield for the ensemble and time averaged variance-based displacements for the equilibrium process

$$\langle x_{\text{eq}}^2(\Delta)\rangle - \langle x_{\text{eq}}(\Delta)\rangle^2 = \langle x_{\text{or}}^2(\Delta)\rangle - \langle x_{\text{or}}(\Delta)\rangle^2 \tag{54}$$

and

$$\left\langle \overline{\delta\delta^2(\Delta)_{\text{eq}}}\right\rangle = \left\langle \overline{\delta\delta^2(\Delta)_{\text{or}}}\right\rangle, \tag{55}$$

i.e., the results which are identical to those for the ordinary process, see eqn (48) and (49). Namely, we find a linear-in-time spreading of the particles with respect to the mean in terms of the ensemble and time averaged characteristics, see also Fig. 2 and Table 1. We mention that for $\langle x(t)\rangle$ and $\langle x^2(t)\rangle - \langle x(t)\rangle^2$ some scaling results were derived previously.[162] Also, the reader can compare eqn (54) to the ensemble averaged displacements for stored energy-driven Lévy flights.[176,178]

From eqn (48) a critical exponent α_{crit} can be obtained: below this value the particle spreading properties get facilitated or enhanced by the bias, as compared to symmetric diffusive CTRWs with $\alpha > 2$ and $p = q = 1/2$. Namely, using the relation $4pq = 1 - (p - q)^2$, the condition for bias-enhanced spreading,

$$(\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2)|_{p,q} > (\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2)|_{p=q=1/2}, \tag{56}$$

reduces to

$$\sigma_\alpha^2 > \mu_\alpha^2. \tag{57}$$

This, using eqn (4) and (5), yields the condition for the WTD exponent

$$2 < \alpha < \alpha_{\text{crit}} = 1+\sqrt{2}. \tag{58}$$

The presence of drift for the WTD exponents outside of this range thus diminishes particle spreading.

The results of our computer simulations, performed as outlined in Section IIB, together with the long-time analytical scalings for the ensemble averaged variance, are presented in Fig. 4a and b. We observe that for $\alpha = 3$ the variance at a given time t gets reduced for more asymmetric walks, *i.e.*, for larger jump asymmetry parameters

$$\varepsilon = p - q. \tag{59}$$

In virtue of normalisation condition (2), a given value for ε unequivocally defines the probabilities p and q. The linear scale is chosen in Fig. 4 to demonstrate the linear growth of the

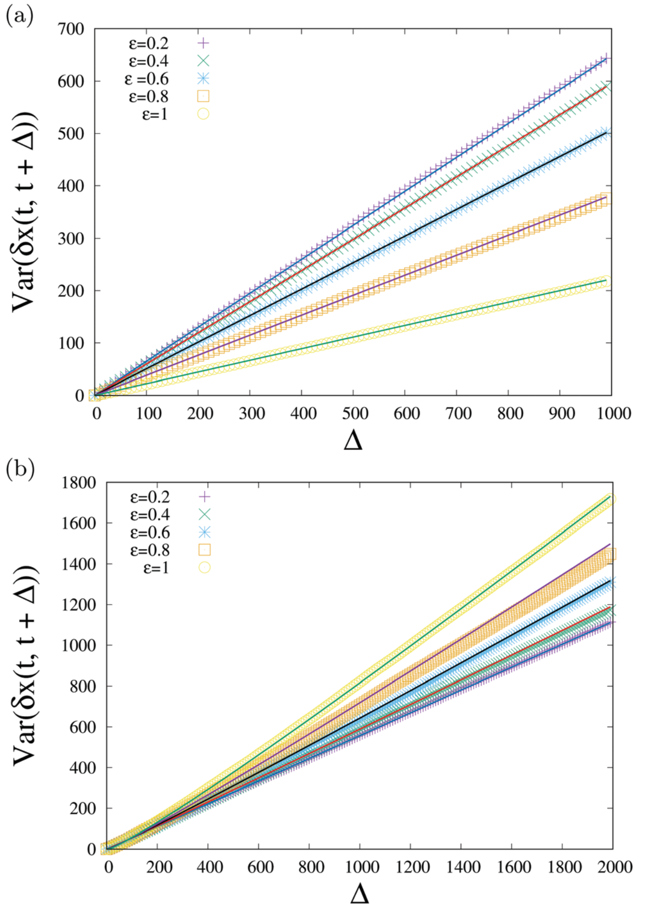

Fig. 4 Ensemble averaged variance for CTRWs (17) with exponents α = 3 and α = 2.2 (panels (a) and (b), respectively) *versus* lag time Δ for varying asymmetry parameter (59), as indicated in the plots. The results of computer simulations (symbols) follow the long-time asymptotes (48) (solid curves). In this and later plots of the particle displacements, ensemble averaging is performed over $M = 10^4$ trajectories.

ensemble averaged particle displacements. In contrast, for $\alpha = 2.2 < \alpha_{\text{crit}}$ the variance-based displacement increases for more asymmetric CTRWs, in agreement with relation (58).

B. Displacements for $1 < \alpha < 2$: ordinary and equilibrium processes

For CTRW processes with WTD exponent in the range $1 < \alpha < 2$, the Tauberian inversion (42) of the Laplace transforms (33) yields for the first two moments and variance of the number of particle jumps for the ordinary renewal processes that

$$\langle N_{\text{or}}(t)\rangle \sim \frac{t}{\mu_\alpha} + \frac{k_\alpha t^{2-\alpha}}{\mu_\alpha^2 \Gamma(3-\alpha)}, \tag{60}$$

$$\left\langle N_{\text{or}}^2(t)\right\rangle \sim \frac{t^2}{\mu_\alpha^2} + \frac{4k_\alpha t^{3-\alpha}}{\mu_\alpha^3 \Gamma(4-\alpha)}, \tag{61}$$

and

$$\left\langle N_{\text{or}}^2(t)\right\rangle - \langle N_{\text{or}}(t)\rangle^2 \sim \frac{2k_\alpha(\alpha-1)}{\mu_\alpha^3 \Gamma(4-\alpha)} t^{3-\alpha}. \tag{62}$$

We mention the two different powers in the long-time scaling for the first two moments, similar to the solutions of the bi-fractional diffusion equation.[138] The reader is also referred to eqn (3.9) and (3.10) of ref. 97 for the mean and variance of the number of renewal events in this α range. We note the existence of two distinct power exponents describing the fluctuations of $N(t)$, see eqn (60) and (61).

For the correlator of the number of steps we find

$$\begin{aligned} &\langle N_{\text{or}}(t)(N_{\text{or}}(t+\Delta) - N_{\text{or}}(t))\rangle \\ &\sim \frac{t\Delta}{\mu_\alpha^2} + \frac{2k_\alpha t^{2-\alpha}\Delta}{\mu_\alpha^3 \Gamma(3-\alpha)} \\ &- \frac{k_\alpha \Delta^{3-\alpha}}{\mu_\alpha^3 \Gamma(4-\alpha)} + \frac{k_\alpha t^{1-\alpha}\Delta^2}{2\mu_\alpha^3 \Gamma(2-\alpha)}, \end{aligned} \tag{63}$$

which is obtained *via* inverting the long-time expansion (39) in the double-Laplace domain for $u_1 \ll s_1$, namely

$$\begin{aligned} &\mathcal{L}_{s_1}\mathcal{L}_{u_1}\{\langle N_{\text{or}}(t)(N_{\text{or}}(t+\Delta) - N_{\text{or}}(t))\rangle\} \\ &\sim \frac{1}{\mu_\alpha^2 u_1^2 s_1^2} + \frac{2k_\alpha}{\mu_\alpha^3 u_1^{3-\alpha} s_1^2} \\ &- \frac{k_\alpha}{\mu_\alpha^3 u_1 s_1^{4-\alpha}} + \frac{k_\alpha}{\mu_\alpha^3 u_1^{2-\alpha} s_1^3}. \end{aligned} \tag{64}$$

We emphasise that the asymptotic behaviours of the moments and correlators of the jump numbers given above only contain the "correction terms" that are linear in k_α, as expected.

Inserting the long-time expansions (60)–(63) into eqn (13) and (19) we find that the ordinary CTRW process with WTD exponent in the range $1 < \alpha < 2$ is non-ergodic as the ensemble averaged displacement after time Δ given by

$$\begin{aligned} &\left\langle x_{\text{or}}^2(\Delta)\right\rangle - \langle x_{\text{or}}(\Delta)\rangle^2 \sim 4pqa^2\left(\frac{\Delta}{\mu_\alpha} + \frac{k_\alpha}{\mu_\alpha^2}\frac{\Delta^{2-\alpha}}{\Gamma(3-\alpha)}\right) \\ &+ a^2(p-q)^2 \frac{2k_\alpha(\alpha-1)}{\mu_\alpha^3 \Gamma(4-\alpha)}\Delta^{3-\alpha} \end{aligned} \tag{65}$$

for finite trace lengths T is, strictly speaking, not equal to the time averaged variance-based displacement,

$$\begin{aligned} \left\langle \overline{\delta\delta^2(\Delta)}_{\text{or}}\right\rangle &\sim 4pqa^2\left(\frac{\Delta}{\mu_\alpha} + \frac{k_\alpha \Delta}{\mu_\alpha^2 \Gamma(3-\alpha) T^{\alpha-1}}\right) \\ &+ a^2(p-q)^2 \frac{2k_\alpha \Delta^{3-\alpha}}{\mu_\alpha^3 \Gamma(4-\alpha)}. \end{aligned} \tag{66}$$

The latter is obtained *via* integrating

$$\begin{aligned} &\left\langle (x_{\text{or}}(t+\Delta) - x_{\text{or}}(t))^2\right\rangle - \langle x_{\text{or}}(t+\Delta) - x_{\text{or}}(t)\rangle^2 \\ &\sim 4pqa^2\left(\frac{\Delta}{\mu_\alpha} + \frac{k_\alpha \Delta}{\mu_\alpha^2 \Gamma(2-\alpha) t^{\alpha-1}}\right) \\ &+ a^2(p-q)^2 \frac{2k_\alpha \Delta^{3-\alpha}}{\mu_\alpha^3 \Gamma(4-\alpha)}. \end{aligned} \tag{67}$$

We find that both the ensemble and time averaged displacements (65) and (66) contain both linear and anomalous contributions in diffusion time Δ. We also mention in eqn (67) the explicit dependence on time t along the trajectory, that gives rise to a transient ageing effect. Using expressions (4) and (8) for μ_α and k_α, a more physical representation of (66) in terms of dimensionless time (Δ/τ_0) is

$$\left\langle\overline{\delta\delta^2(\Delta)_{\rm or}}\right\rangle \sim 4pqa^2\left[\frac{\alpha-1}{\alpha}\left(\frac{\Delta}{\tau_0}\right)+\frac{\alpha-1}{\alpha^2(2-\alpha)}\left(\frac{\Delta}{\tau_0}\right)\left(\frac{\tau_0}{T}\right)^{\alpha-1}\right] + \frac{2a^2(p-q)^2(\alpha-1)^2}{(3-\alpha)(2-\alpha)\alpha^3}\left(\frac{\Delta}{\tau_0}\right)^{3-\alpha}. \quad (68)$$

For the equilibrium renewal processes with $1 < \alpha < 2$ from eqn (34) we get

$$\langle N_{\rm eq}(t)\rangle = \frac{\langle x(t)_{\rm eq}\rangle}{a(p-q)} \sim \frac{t}{\mu_\alpha}, \quad (69)$$

$$\langle {N_{\rm eq}}^2(t)\rangle \sim \frac{t^2}{{\mu_\alpha}^2}+\frac{2k_\alpha t^{3-\alpha}}{{\mu_\alpha}^3\Gamma(4-\alpha)}, \quad (70)$$

and

$$\langle {N_{\rm eq}}^2(t)\rangle - \langle N_{\rm eq}(t)\rangle^2 \sim \frac{2k_\alpha t^{3-\alpha}}{{\mu_\alpha}^3\Gamma(4-\alpha)}. \quad (71)$$

The corresponding double Laplace space $\{u_1,s_1\}$-expansion of eqn (40) has the form

$$\mathcal{L}_{s_1}\mathcal{L}_{u_1}\{\langle N_{\rm eq}(t)(N_{\rm eq}(t+\Delta)-N_{\rm eq}(t))\rangle\} \sim \frac{1}{{\mu_\alpha}^2{u_1}^2{s_1}^2}+\frac{k_\alpha}{{\mu_\alpha}^3{u_1}^{3-\alpha}{s_1}^2} - \frac{k_\alpha}{{\mu_\alpha}^3u_1{s_1}^{4-\alpha}}+\frac{k_\alpha}{{\mu_\alpha}^3{u_1}^{2-\alpha}{s_1}^3}, \quad (72)$$

which differs from eqn (64) for the ordinary processes by the factor of 2 in one of the terms. This fact gets reflected in the particle displacement characteristics, namely, we find that

$$\langle N_{\rm eq}(t)(N_{\rm eq}(t+\Delta)-N_{\rm eq}(t))\rangle \sim \frac{t\Delta}{{\mu_\alpha}^2}+\frac{k_\alpha t^{2-\alpha}\Delta}{{\mu_\alpha}^3\Gamma(3-\alpha)} - \frac{k_\alpha\Delta^{3-\alpha}}{{\mu_\alpha}^3\Gamma(4-\alpha)}+\frac{k_\alpha t^{1-\alpha}\Delta^2}{2{\mu_\alpha}^3\Gamma(2-\alpha)}. \quad (73)$$

Therefore, the equilibrium process with WTD exponents in this range, contrary to the ordinary process, remains ergodic in the leading order in the sense of eqn (21). Namely, we get that the ensemble averaged variance-based displacement,

$$\langle {x_{\rm eq}}^2(\Delta)\rangle - \langle x_{\rm eq}(\Delta)\rangle^2 \sim 4pqa^2\frac{\Delta}{\mu_\alpha}+a^2(p-q)^2\frac{2k_\alpha\Delta^{3-\alpha}}{{\mu_\alpha}^3\Gamma(4-\alpha)}, \quad (74)$$

is equal to its time averaged partner,

$$\left\langle(x_{\rm eq}(t+\Delta)-x_{\rm eq}(t))^2\right\rangle - \langle x_{\rm eq}(t+\Delta)-x_{\rm eq}(t)\rangle^2 = \left\langle\overline{\delta\delta^2(\Delta)_{\rm eq}}\right\rangle. \quad (75)$$

Thus, the final results for the ensemble and time averaged transport properties are given by eqn (65), (66) and (74), (75) for the ordinary and equilibrium CTRW processes with $1 < \alpha < 2$, respectively. For the equilibrium situation, both averages contain terms linear in the lag time $\propto\Delta^1$ and superdiffusive contributions $\propto\Delta^{3-\alpha}$ that emerge due to the drift present in the system, see also Fig. 2 and Table 1. Also, the spreading of the particles with respect to the mean governed by the biased CTRW is linear-to-superdiffusive and it is always enhanced due to the bias, as compared to symmetric walks with $p = q = 1/2$, see also details in ref. 185. The reader is referred here to the detailed classification of dispersive and enhanced ensemble averaged properties of CTRWs with a constant velocity field.[130]

In this range of α exponents the factor $(\alpha - 1) < 1$ and the term $\propto\Delta^{2-\alpha}$ in eqn (65) is the difference of the ensemble averaged displacement for the ordinary *versus* the equilibrium processes that obey eqn (74). The time averaged particle displacements for these two situations—see eqn (66) and (75)—differ by the term depending on the trace length T for the ordinary case. Therefore, for biased equilibrium renewal processes in this α range the ensemble averaged time averaged displacements do not depend on the trace length T and such a process is ergodic (see Section IIIC for more details), in contrast to the ordinary process in this range of α exponent. More generally than the long-time equivalence of the ensemble and time averages in (21), ergodicity can be defined through the corresponding equilibrium ensemble average:[181] biased CTRWs with $1 < \alpha < 2$ appear ergodic in this sense.

In Fig. 5a we demonstrate the agreement of the analytical results and findings from computer simulations for the particle displacements, for both ordinary and equilibrium superdiffusive CTRW processes. For the chosen value of the WTD scaling exponent, $\alpha = 3/2$, the equilibrium processes have larger variance of particle displacements, as compared to the ordinary ones. Fig. 5a also supports the analytical trend that the variance of the number of particle jumps for the ordinary diffusion processes with $1 < \alpha < 2$ is $(\alpha - 1)$ times smaller than their equilibrium counterpart, compare eqn (62) and (71). This result is reminiscent to the observation for superdiffusive Lévy walks, see ref. 6 and 186–188.

In Fig. 5b we show the variation of the ensemble averaged variance-based displacements for systematically varying ageing time, setting $t = t_a$ in eqn (67) for $\langle(x_{\rm or}(t+\Delta) - x_{\rm or}(t))^2\rangle - \langle x_{\rm or}(t+\Delta) - x_{\rm or}(t)\rangle^2$, see ref. 12 for more details on ageing. Here, the ageing time is defined as the delay time between the initiation of the diffusion process with the trapping time distribution (3) and the start of displacement measurements, see ref. 8, 12 and 136. We find that for longer ageing times the magnitude of the variance-based particle displacements after a given diffusion time (denoted Δ in Fig. 5b) grows. This is in agreement with the theoretical prediction (67), compare the symbols and the asymptotes in Fig. 5b. Note that subdiffusive CTRWs considered in Section IIID reveal opposite ageing trends, as expected.

C. Ergodicity breaking parameter for $\alpha > 1$

The issue of irreproducibility or relative amplitude fluctuations of individual time averaged realisations can be quantified in terms of the relative standard deviation (RSD), which is the square root of the EB parameter. The reader is referred to, *e.g.*,

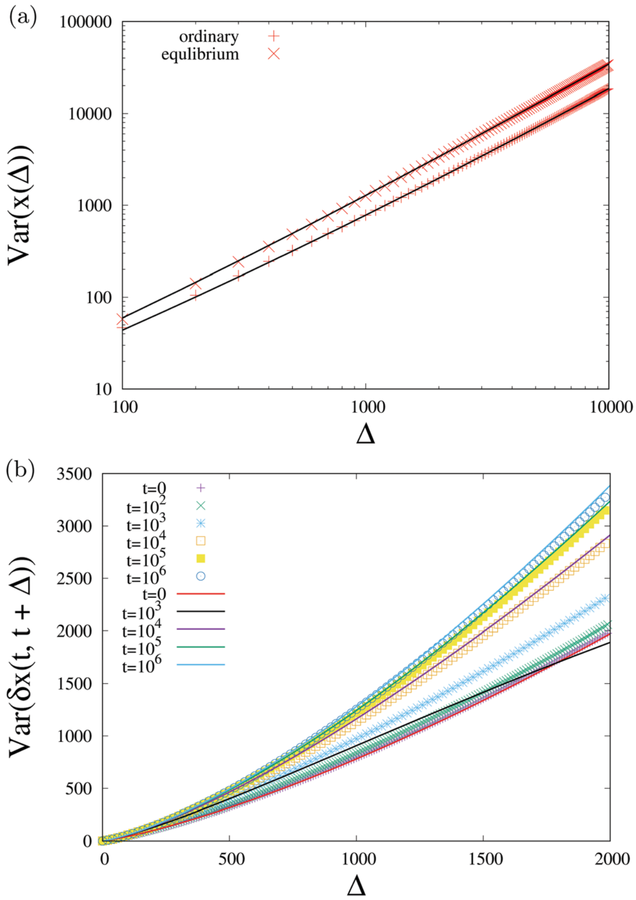

Fig. 5 (a) Ensemble averaged variance of particle displacements for CTRWs, $\langle x^2(\varDelta)\rangle - \langle x(\varDelta)\rangle^2$, with $\alpha = 3/2$ and asymmetry parameter $\varepsilon = 0.4$, plotted for both the ordinary and equilibrium processes in log–log scale. The results of computer simulations are the symbols and the asymptotic relations for the ordinary and equilibrium cases (eqn (65) and (74) in the text) are the solid curves. (b) Variance of particle displacements along the trajectory for the ordinary process, $\langle(x_{\rm or}(t+\varDelta) - x_{\rm or}(t))^2\rangle - \langle x_{\rm or}(t+\varDelta) - x_{\rm or}(t)\rangle^2$, for different ageing times as indicated in the plot. We set $t = t_a$ for ageing times in eqn (67) and compute the results for $\alpha = 3/2$.

ref. 172 and 189 for the RSD consideration for both equilibrium and ordinary renewal processes. The EB parameter itself is typically defined *via* the time averaged fourth and second moments of particle displacements *via*[7,8,12,102,190,191]

$$\mathrm{EB}(\varDelta) = \left\langle \left(\overline{\delta^2(\varDelta)}\right)^2 \right\rangle \Big/ \left\langle \overline{\delta^2(\varDelta)} \right\rangle^2 - 1. \quad (76)$$

For stochastic processes with non-zero mean displacements of the walker, such as our biased renewal processes, the natural generalisation of the EB parameter (76) is

$$\delta\mathrm{EB}(\varDelta) = \left\langle \left(\overline{\delta\delta^2(\varDelta)}\right)^2 \right\rangle \Big/ \left\langle \overline{\delta\delta^2(\varDelta)} \right\rangle^2 - 1. \quad (77)$$

The phenomenon of weak ergodicity breaking is the absence of convergence of individual time averaged MSDs to their corresponding ensemble averages at a given lag time $\varDelta$. Instead, the "distributional ergodicity"[7,8,170,178] or the convergence to a final distribution of time averaged trajectories is often realised. For some bias-free anomalous diffusion processes, such as drift-free subdiffusive CTRWs[7,8] and subdiffusive heterogeneous diffusion processes,[192–194] the EB parameter quantifies the intrinsic irreproducibility of time averaged MSD magnitudes. In other words, the generalised diffusion coefficient for each time averaged MSD realisation is itself a random quantity, following a given distribution.[7,49,165,169,173] Note here that the properties of particle diffusion in time-dependent and fluctuating diffusivity landscapes were also studied in ref. 167, 195 and 196.

Moreover, assessing the EB parameter can help quantifying extrinsic effects on particle diffusion, even if the underlying idealised mathematical process is perfectly reproducible. These effects may include, *e.g.*, medium heterogeneities (viscosity, friction, *etc.*),[9,197] size or mass polydispersity of the tracers, and possible population splitting based on mobility ranges of the walkers.[12,37,198]

Thus, in addition to the comparison of the second moments of the ensemble and time averaged variance-based particle displacements, for $\alpha > 1$ we evaluate the EB parameter as[7,12,144,164,190,192,194] the short lag time limit of eqn (77). Being based on the fourth moment of the time averaged displacement, compared to the second moment it is generally considerably harder to compute analytically for a number of processes.[144,164,190–192] However, for CTRWs with $\alpha > 1$ the particle increments along the trajectory, needed to compute the time averaged displacements (19), do not depend on time t along the trace.

For situations when the mean waiting time μ_α exists (that corresponds to $\alpha > 1$), to assess the change in fluctuations of $\overline{\delta\delta^2(\varDelta)}$ with overall trace length T we use the following approximation

$$\overline{\delta\delta^2(\varDelta)} \sim \frac{N(T)}{T} h_\alpha(\varDelta) + \frac{\int_0^{T-\varDelta} \langle x(t+\varDelta) - x(t)\rangle^2 \mathrm{d}t}{T-\varDelta}. \quad (78)$$

This works rather well when the mean number of trapping and jumping events grows linearly with the diffusion time. Note that for $\alpha > 1$ the integrand of the second term in eqn (78) does not depend on t for large t values. After ensemble averaging (15), one arrives at the identity

$$\left\langle \overline{\delta\delta^2(\varDelta)} \right\rangle \sim a^2 \frac{\langle N(T)\rangle}{T} h_\alpha(\varDelta) + \left(\frac{\varDelta}{\mu_\alpha}\right)^2 a^2 (p-q)^2, \quad (79)$$

where for $\alpha > 2$ the function $h_\alpha(\varDelta)$ is given by

$$h_\alpha(\varDelta) = \left(1 + \frac{{\sigma_\alpha}^2 - {\mu_\alpha}^2}{{\mu_\alpha}^2}(p-q)^2\right)\varDelta - \frac{\varDelta^2}{\mu_\alpha}(p-q)^2, \quad (80)$$

while for $1 < \alpha < 2$ one has

$$h_\alpha(\varDelta) = \frac{2(p-q)^2 k_\alpha}{{\mu_\alpha}^2 \Gamma(4-\alpha)} \varDelta^{3-\alpha} + 4pq\varDelta - \frac{\varDelta^2}{\mu_\alpha}(p-q)^2. \quad (81)$$

Inserting expression (78) into eqn (77) enables us to assess the dependencies of the δEB parameter in terms of the variance of the number of particle jumps as

$$\delta \mathrm{EB}(T,\Delta) \sim \frac{\dfrac{\langle N(T)^2\rangle}{\langle N(T)\rangle^2}-1}{1+\dfrac{2T\Delta^2(p-q)^2}{{\mu_\alpha}^2 h_\alpha(\Delta)\langle N(T)\rangle}}. \quad (82)$$

We now use the general expression (82) to derive the leading scaling of the ergodicity breaking parameter separately for $\alpha > 2$ and $1 < \alpha < 2$, both for ordinary and equilibrium situations. In the limit $\Delta/T \ll 1$, using the corresponding expression for δEB, $\langle N(T)\rangle$ and $\langle N^2(T)\rangle - \langle N(T)\rangle^2$, the final results for δEB (Δ,T) can be presented in the form

$$\delta \mathrm{EB} \sim \begin{cases} \dfrac{{\sigma_\alpha}^2/(\mu_\alpha T)}{1+\dfrac{2\Delta^2(p-q)^2}{\mu_\alpha h_\alpha(\Delta)}} \sim 1/T^1, & 2<\alpha,\ (\text{or., eq.}) \\ (\alpha-1)\dfrac{2k_\alpha/T^{\alpha-1}}{\left(1+\dfrac{2\Delta^2(p-q)^2}{\mu_\alpha h_\alpha(\Delta)}\right)\mu_\alpha \Gamma(4-\alpha)} \sim 1/T^{\alpha-1}, & 1<\alpha<2,\ (\text{or.}) \\ \dfrac{2k_\alpha/T^{\alpha-1}}{\left(1+\dfrac{2\Delta^2(p-q)^2}{\mu_\alpha h_\alpha(\Delta)}\right)\mu_\alpha \Gamma(4-\alpha)} \sim 1/T^{\alpha-1}, & 1<\alpha<2,\ (\text{eq.}) \end{cases}. \quad (83)$$

Thus, markedly different scaling relations for the ergodicity breaking parameter (77) *versus* the trace length are predicted by this approach. Namely, for $\alpha > 2$ eqn (83) yields a rather standard decay law

$$\delta \mathrm{EB}(T) \sim 1/T^1. \quad (84)$$

In contrast, a slower approach to ergodicity for WTD exponents in the range $1 < \alpha < 2$ is found, namely anomalously slow decay

$$\delta \mathrm{EB}(T) \sim 1/T^{\alpha-1}. \quad (85)$$

The δEB parameter for $\alpha > 2$ in this approach attains the same limiting behaviour for the ordinary and equilibrium processes. For $1 < \alpha < 2$ the δEB parameter for the ordinary situation is $(\alpha - 1)$ times smaller than that for the equilibrium case, see eqn (83).

Note that the evaluation of δEB for biased CTRWs with $0 < \alpha < 1$ is a more involved mathematical problem, that deserves a separate study (see ref. 7, 8, 41 and 168 for EB parameter evaluations for bias-free CTRWs). Also, note that in the limit $\Delta/T \to 0$ for unbiased subdiffusive walks the standard EB parameter approaches the limiting value[7,8,12]

$$\mathrm{EB}_{\mathrm{CTRW}} \approx \left(\frac{2[\Gamma(1+\alpha)]^2}{\Gamma(1+2\alpha)}-1\right). \quad (86)$$

Finally, the higher-order moments of the scatter distribution of time averaged particle displacements for biased CTRWs can also be interesting to analyse (albeit harder). For drift-free subdiffusive CTRWs the skewness and kurtosis were examined recently in ref. 144.

The results of numerical computer simulations together with analytical predictions for the δEB parameter are presented in Fig. 6. The quantitative agreement we observe for $1 < \alpha < 2$, supports the validity of eqn (83) and the underlying approximation (78). We refer the reader to Fig. 7 where relations (80) and (81) are checked for the cases $\alpha = 3$ and $\alpha = 3/2$, respectively in panels (a) and (b). Excellent agreement is observed for $\alpha = 3/2$, while some evident discrepancies between theory and simulations are found for $\alpha = 3$. This gets reflected later in somewhat inaccurate δEB values for biased CTRWs in the range $\alpha > 2$, as illustrated in Fig. 6b. Numerical computer simulations results agree well with the scaling (83) and support the power-law decay δEB(T) with T for biased CTRWs with $1 < \alpha < 2$, particularly for large Δ values. We should mention that the RSD evaluation is the most computationally demanding part of the current study. As an example, it takes about 6 hours on a standard workstation to compute the RSD for Fig. 6 for $M = 10^4$ trajectories (for $\Delta = 10^3$, $T = 10^6$, and $\alpha = 3$). For shorter lag times the relations of eqn (83) appears less applicable.

For comparatively large α values, as in Fig. 6b for $\alpha = 3$, relation (83) describes the scaling of δEB(T) correctly for different lag times. However, for the δEB magnitude at small Δ values the theory deviates from the results of computer simulations. Finally, further in-depth theoretical analysis *via* evaluation of the fourth time averaged moment of particle displacements for biased CTRWs with $\alpha > 2$ is needed. This, however, is beyond the scope of the present study (to be considered elsewhere).

D. Displacements for $0 < \alpha < 1$: non-equilibrium process

Bias-free subdiffusive CTRWs with diverging mean waiting times are known to be non-ergodic and ageing.[7,8,12,134,168] For biased CTRWs with $0 < \alpha < 1$ the derivations of ensemble and time averaged variance-based drift-corrected particle displacement characteristics are also somewhat more involved than for the ergodic and non-ageing case of $\alpha > 1$. Here, analogously, we perform the inverse Laplace transforms of $\alpha < 1$ expansion of $\hat{\psi}(s)$ in eqn (7). We find for the heavy-tailed WTDs in the leading order that (see also eqn (3.6) of ref. 97 and eqn (5.150) of ref. 93)

$$\langle N(t)\rangle = \frac{\langle x(t)\rangle}{a(p-q)} \sim \frac{t^\alpha}{k_\alpha \Gamma(1+\alpha)}, \quad (87)$$

$$\langle N^2(t)\rangle \sim \frac{2t^{2\alpha}}{{k_\alpha}^2 \Gamma(2\alpha+1)}, \quad (88)$$

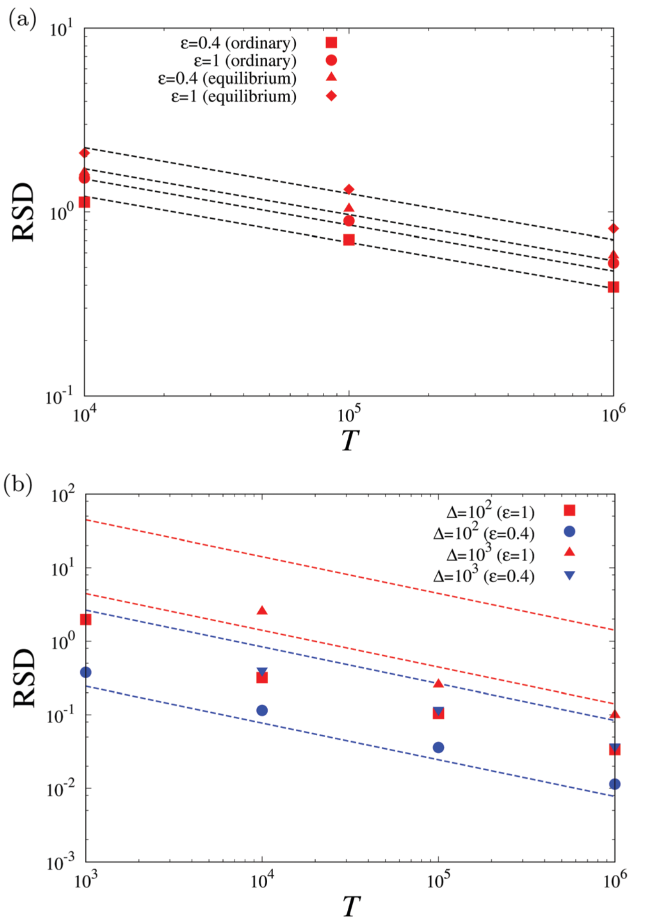

Fig. 6 Relative standard-deviation parameter for biased CTRWs, $\mathrm{RSD}(T) = \sqrt{\delta\mathrm{EB}(T)}$ as defined in eqn (77), with $\alpha = 3/2$ (panel (a)) plotted *versus* the trace length T for lag time $\Delta = 10^3$, both for ordinary and equilibrium processes. The values of the asymmetry parameter ε are indicated in the plots. Straight lines represent the theoretical scaling (83). Averaging over $M = 10^4$ traces was performed. When p value is not explicitly mentioned, we set $\varepsilon = 0.4$ and $p = 0.7$, coupled *via* (2). In panel (b), the RSD results *versus* the trace length T are presented, computed for $\alpha = 3$, for two asymmetry parameter values, and for lag times $\Delta = 10$ and 10^3. Simulation results are shown together with theoretical asymptotes (83). As the measurement time is larger than the lag time Δ by definition, for $T = 10^3$ the points for $\Delta = 10^3$ are missing in panel (b).

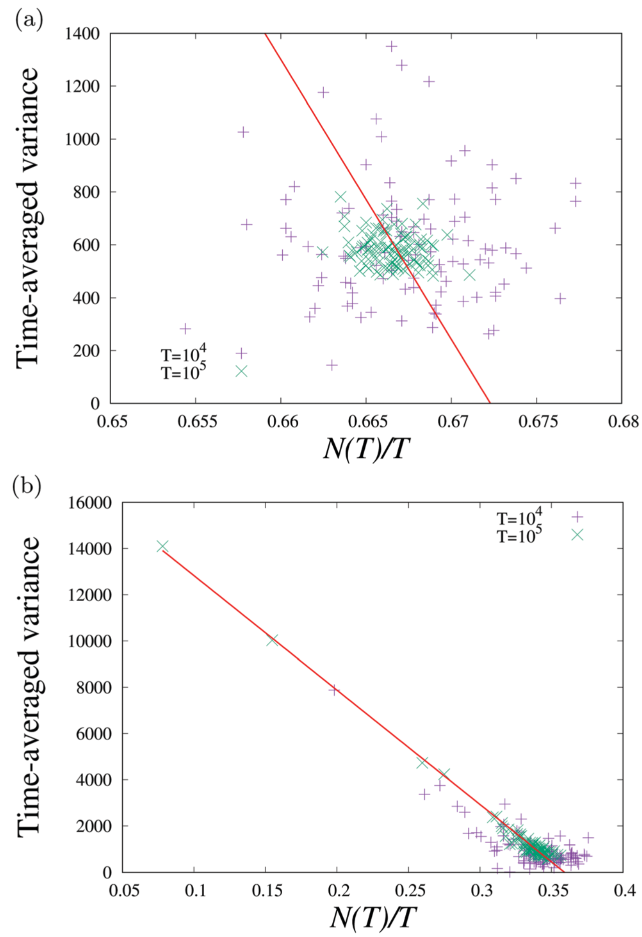

Fig. 7 Theoretical relations for the time averaged variance-based MSD (79)—with account for representations (80) and (81)—*versus* the results of computer simulations of $N = 10^2$ trajectories, computed for $\alpha = 3$ (panel (a)) and $\alpha = 3/2$ (panel (b)). Other parameters: $\varepsilon = 0.4$, $\Delta = 10^3$, $T = 10^4$ and 10^5. Note the extremely small range on the abscissa of panel (a).

and

$$\langle N^2(t)\rangle - \langle N(t)\rangle^2 \sim \left(\frac{2[\Gamma(1+\alpha)]^2}{\Gamma(1+2\alpha)} - 1\right)\frac{t^{2\alpha}}{k_\alpha{}^2[\Gamma(1+\alpha)]^2}. \quad (89)$$

We find for the correlator of the number of jumps (39) that

$$\begin{aligned}&\langle N(t)(N(t+\Delta) - N(t))\rangle\\ &\sim \mathcal{L}_{s_1}{}^{-1}\mathcal{L}_{u_1}{}^{-1}\left\{\frac{1}{k_\alpha{}^2 s_1{}^2 u_1^{2\alpha}} - \frac{1}{k_\alpha{}^2 s_1{}^{2+\alpha}u_1^{\alpha}}\right\}\\ &\sim \frac{t^{2\alpha-1}\Delta}{k_\alpha{}^2\Gamma(2\alpha)} - \frac{t^{\alpha-1}\Delta^{1+\alpha}}{k_\alpha{}^2\Gamma(2+\alpha)\Gamma(\alpha)},\end{aligned} \quad (90)$$

while the variance-based particle displacements are

$$\begin{aligned}\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2 &\sim \frac{4pqa^2\Delta^\alpha}{k_\alpha\Gamma(1+\alpha)}\\ &+ \left(\frac{2[\Gamma(1+\alpha)]^2}{\Gamma(1+2\alpha)} - 1\right)\frac{a^2(p-q)^2\Delta^{2\alpha}}{k_\alpha{}^2[\Gamma(1+\alpha)]^2}.\end{aligned} \quad (91)$$

The first moment and the variance expressions (87) and (91) are to be compared to eqn (41) of ref. 130 obtained from the generalised Galilei-invariant advection–diffusion equation approach to CTRWs in the presence of a velocity field (see also App. B of ref. 107). Also, for the increments of particle displacements along the trajectory we find

$$\begin{aligned}&\langle (x(t+\Delta) - x(t))^2\rangle - \langle x(t+\Delta) - x(t)\rangle^2\\ &\sim \frac{4pqa^2t^{\alpha-1}\Delta}{k_\alpha\Gamma(\alpha)} + \frac{2a^2(p-q)^2t^{\alpha-1}\Delta^{1+\alpha}}{k_\alpha{}^2\Gamma(2+\alpha)\Gamma(\alpha)}.\end{aligned} \quad (92)$$

Here, the explicit dependence on time t along the trajectory—that can also be considered in eqn (92) as ageing time, $t = t_a$—emphasises ageing in the system. In eqn (91) and (92) we also kept the terms of subleading order in the lag time Δ, to ensure the correct limit for bias-free subdiffusive CTRWs, with MSD(Δ) $\sim \Delta^{\alpha}$ at $p = q = 1/2$. For biased subdiffusive CTRWs, in eqn (91) additionally a second contribution $\sim\Delta^{2\alpha}$ appears, see also Fig. 2 and Table 1. It gives rise to a subdiffusive trend for particle spreading for $0 < \alpha < 1/2$ and to a superdiffusive spreading for the WTD exponent in the range $1/2 < \alpha < 1$, as already pointed out by Scher and Montroll[126] and Shlesinger.[125]

As we see from eqn (92)—see also eqn (59) in ref. 139 and eqn (12) in ref. 125 for the dispersion of subdiffusive biased CTRWs—the variance of particle displacement increments does exhibit an explicit t-dependence. Therefore, the ensemble averaged time averaged displacement $\left\langle \overline{\delta\delta^2(\Delta)} \right\rangle$ defined by eqn (15) reveals ageing effects.[8,12,136] Namely, its magnitude

$$\left\langle \overline{\delta\delta^2(\Delta,T)} \right\rangle \sim \frac{4pqa^2\Delta}{k_\alpha\Gamma(1+\alpha)T^{1-\alpha}} + \frac{2a^2(p-q)^2\Delta^{1+\alpha}}{{k_\alpha}^2\Gamma(2+\alpha)\Gamma(1+\alpha)T^{1-\alpha}} \qquad (93)$$

decreases as a power law with the trajectory length T, $\left\langle \overline{\delta\delta^2(\Delta,T)} \right\rangle \sim 1/T^{1-\alpha}$, similarly to the ageing properties of symmetric drift-free subdiffusive CTRWs.[8,12,137] Eqn (93) thus demonstrates that biased subdiffusive CTRWs are non-ergodic and ageing. Namely, the growth of the ensemble averaged variance-based displacements (91) with the lag time differs from that of time averaged ensemble averaged variance-based displacement (93), that is

$$\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2 \neq \left\langle \overline{\delta\delta^2(\Delta)} \right\rangle. \qquad (94)$$

In the limit of symmetric subdiffusive CTRWs (with $p = q$) the net displacement of the particles (10) vanishes. Then, the variance-based expressions (91) and (93) for the particle spreading turn into the results for the ensemble and time averaged MSDs of subdiffusive CTRWs. For the latter, in the limit $\Delta/T \ll 1$, one gets the standard ageing and non-ergodic scaling[7,8,12]

$$\langle x^2(\Delta)\rangle \sim a^2\left(\frac{\sin(\pi\alpha)}{\pi\alpha}\right)\left(\frac{\Delta}{\tau_0}\right)^{\alpha} \sim \left\langle \overline{\delta^2(\Delta)} \right\rangle \left(\frac{T}{\Delta}\right)^{1-\alpha}. \qquad (95)$$

We note here that some non-ergodic and ageing properties of drift-free subdiffusive CTRWs are similar to those observed for heterogeneous diffusion processes with power-law space-dependent diffusivities, $D(x)$, see ref. 192–194 (and also ref. 199 and 200).

The results for the ensemble averaged variance-based particle spreading of biased CTRWs with $\alpha < 1$ are illustrated in Fig. 8. We find that, in contrast to the findings for biased CTRWs with $1 < \alpha < 2$ shown in Fig. 5, for subdiffusive biased CTRWs the magnitude of particle displacements decreases for longer ageing times t_a. This is intuitively expected, as for a progressive ageing the probability to draw long trapping events from the distribution (3) increases, that in turn reduces the first and second moments of displacements. We find that for smaller values of the WTD exponent α—compare panels (b) and (c) in Fig. 8—the magnitude of variance-based particle displacements acquires stronger effects of growing ageing times t_a, in agreement with analytical predictions, eqn (92) and (93). Note that for shorter traces in panels (b) and (c) of Fig. 8 some discrepancy between the theoretical results and simulations findings appears for long lag times. In this limit, the lag time becomes comparable to the time along the

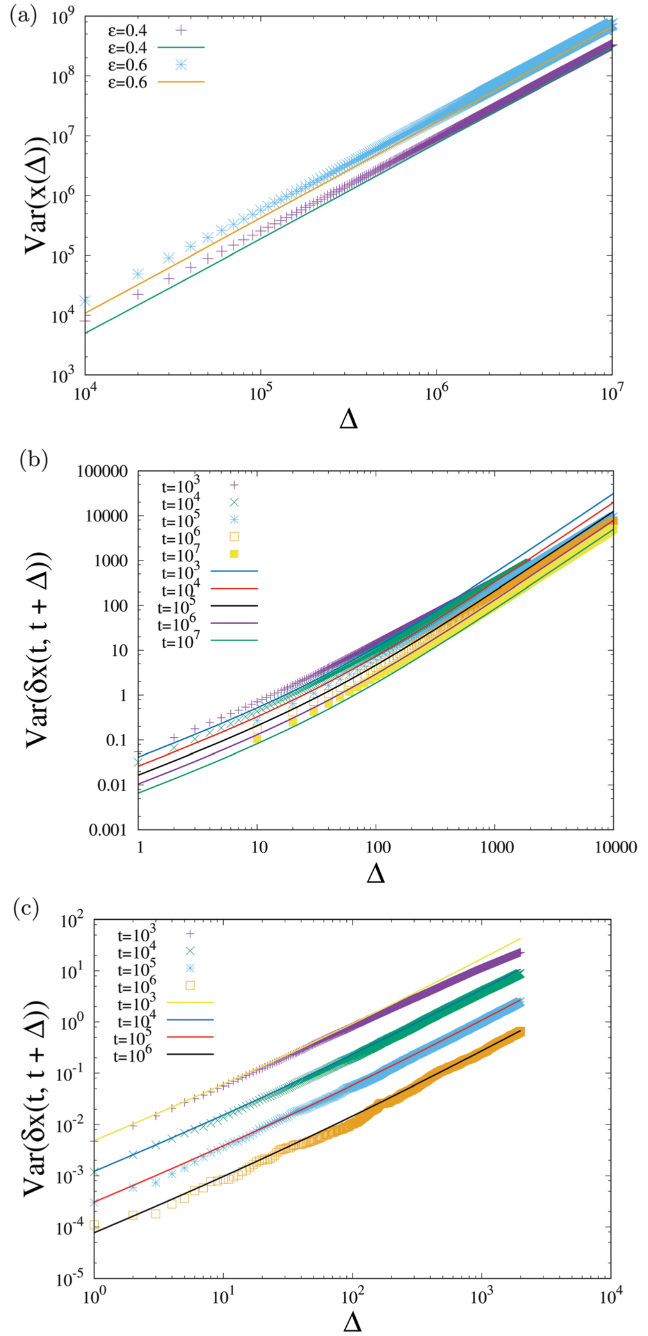

Fig. 8 (a) Variance-based ensemble averaged particle displacements for CTRWs, $\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2$, with $\alpha = 0.8$ and varying asymmetry parameter, plotted in log–log scale. Theoretical asymptotes of eqn (91) and (92) are the solid curves in both panels. The results for the variance of particle position increments along the trajectory, $\langle(x(t+\Delta) - x(t))^2\rangle - \langle x(t+\Delta) - x(t)\rangle^2$, according to eqn (92) plotted for varying ageing times $t = t_a$ are presented for $\alpha = 0.8$ (panel b) and $\alpha = 0.4$ (panel c).

trajectory, see Fig. 8b, c and eqn (41). Also, additional subleading terms, neglected in our analytical relations, can contribute.

The reader is referred here to ref. 12, 137 and 139 for the analytical results for ageing subdiffusive bias-free CTRWs. In this case, the MSD of the particles scales as $\mathrm{MSD_a}(t) = \langle[x(t_a + t) - x(t_a)]^2\rangle \sim t/t_a^{1-\alpha}$ in the limit of short times $t \ll t_a$ and as $\mathrm{MSD_a}(t) \sim t^\alpha$ in the limit of long diffusion times, $t \gg t_a$. The time averaged MSD of such CTRWs grows, in contrast to the ensemble average, always linearly with the lag time,[12,137]

$$\left\langle \overline{\delta^2(\Delta)}_a \right\rangle = \left\langle \frac{\int_{t_a}^{t_a+T-\Delta}[x(t+\Delta)-x(t)]^2 \mathrm{d}t}{T-\Delta} \right\rangle \sim \Delta^1. \qquad (96)$$

Thus, the scalings of the ensemble and time averaged MSDs are both linear only for long diffusion times and strong ageing conditions,[137] when a quasi-stationarity is achieved. On the other hand, for large Δ values the time averaged variance for biased subdiffusive CTRWs grows as $\propto \Delta^1$ and $\propto \Delta^{1+\alpha}$, see eqn (93) and also Table 1.

E. Einstein relation: ensemble and time averaged observables

Here, for biased CTRWs, we check the (second) generalised Einstein relation[2–4,6,7,56,57,131,139,182,188,201–207] that connects the first ensemble averaged moment of particle displacements in the presence of a (weak) constant force F, to the ensemble averaged MSD in the absence of force. Mathematically, the fluctuations of the force-free MSD are then connected to the MSD *via* the standard linear response relation

$$\langle x(t)\rangle_F = \frac{F\langle x^2(t)\rangle_0}{2k_\mathrm{B}T}, \qquad (97)$$

where $k_\mathrm{B}T$ denotes the thermal energy and the applied forces are weak enough so that $aF/(k_\mathrm{B}T) \ll 1$. Here and below, the subscript after the ensemble average brackets denotes a positive force applied, so that $p > q$ in Fig. 1. At equilibrium the jump probabilities between sites satisfy the detailed balance equation,

$$\frac{q}{p} = \exp\left[-\frac{aF}{k_\mathrm{B}T}\right] < 1. \qquad (98)$$

We thus find that the Einstein or the fluctuation–dissipation relation (100) holds for biased CTRWs with arbitrary positive exponents, $\alpha > 0$. Note that for the ordinary processes with $1 < \alpha < 2$ the term $\propto \Delta^{2-\alpha}$ in the force-free second moment (65) compensates the second term in eqn (60).

The generalised Einstein relation for the time averaged moments can be constructed similarly, see also ref. 7 for subdiffusive CTRWs,

$$\left\langle \overline{\delta^1(\Delta)} \right\rangle_F = \frac{F\left\langle \overline{\delta^2(\Delta)} \right\rangle_0}{2k_\mathrm{B}T}. \qquad (99)$$

We refer the reader to ref. 188 and 203 on violation of (99) and the effects of equilibrated starting conditions for superdiffusive Lévy walks.[67] Also note that for bias-free subdiffusive CTRWs ergodicity is violated, but the time averaged Einstein relation in form (99) holds.[7,203] For biased CTRWs we obtain that—similar to the ensemble averaged Einstein relation (97)—the time averaged relation (99) holds to leading order in the entire range of WTD exponents α.

IV. Discussion and conclusions

The main results of the current study, summarised in Table 1, reveal important differences in the particle-spreading dynamics of ordinary and equilibrium processes of biased CTRWs. We demonstrated, in particular, that for power-law WTDs with $\alpha > 2$ ensemble and time averaged variance-based particle displacements—denoted as $\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2$ and $\left\langle \overline{\delta\delta^2(\Delta)} \right\rangle$—are linear in time and lag time, respectively, and the diffusion is fully ergodic.

In the range $1 < \alpha < 2$, for equilibrium processes the spreading of the particles with respect to the mean contains both linear and superdiffusive contributions, but the diffusion remains ergodic. Depending on the time scale at which diffusion is monitored, these additional terms—proportional to the field strength (or, the asymmetry parameter ε)—can dramatically affect the magnitude and spreading characteristics of the particles. These features might become imperative for interpreting the experimental single-particle tracking data and proposing some CTRW-based physical mechanisms to rationalise them. For the ordinary situation in the same range of α exponents, the ensemble and time averaged spreading characteristics are not identical and the system exhibits ergodicity breaking and ageing.

Finally, for the mathematically richest case $0 < \alpha < 1$, the ensemble averaged displacement contains, in addition to the standard $\sim\Delta^\alpha$ time scaling, the term $\sim\Delta^{2\alpha}$. The latter can result in a superdiffusive spreading of the particles governed by biased CTRWs with the exponent $1/2 < \alpha < 1$, as known from previous ensemble modelling.[125,126] Biased CTRWs with the exponents in the range $0 < \alpha < 1$ are non-ergodic and ageing, similar to the drift-free CTRW processes.[7,8,12,136]

We also examined the behaviour of the ergodicity breaking parameter for superdiffusive and superballistic realisations of WTD exponents, α. We found the scaling behaviour $\delta\mathrm{EB}(T) \sim 1/T$ for $\alpha > 2$, while for $1 < \alpha < 2$ the approach to ergodicity is anomalously slow, namely $\delta\mathrm{EB}(T) \sim 1/T^{\alpha-1}$. The calculation of $\delta\mathrm{EB}(\Delta,T)$ for subdiffusive CTRWs $0 < \alpha < 1$ requires a special investigation (not presented here). Lastly, the ensemble and time averaged Einstein relations appear to hold for biased CTRW processes in the entire range of exponents α, see Table 1.

Our results can be of importance for quantifying various transport properties dictated by anomalous and biased diffusion processes, mathematically governed by the power-law distribution of trapping times at individual sites. The classical example is the Scher–Montroll transport[126] in disordered media based on the CTRW diffusion mechanism, recently also studied in the presence of ageing.[137,208] In Table 1—in addition to the scaling behaviour for the ensemble and time averaged quantities derived in Section III—we present the particle spreading parameter defined as (dimensionless) dispersion-to-mean ratio

$$\eta(\Delta) = \frac{\mathrm{Dispersion}(\Delta)}{\mathrm{Mean}(\Delta)} = \frac{\sqrt{\langle x^2(\Delta)\rangle - \langle x(\Delta)\rangle^2}}{\langle x(\Delta)\rangle}. \qquad (100)$$

This is the key parameter in the Scher–Montroll theory[126] describing the universal transport properties of hopping carriers in external fields biasing the motion. Considering a moving packet of particles spreading *via* a subdiffusive CTRW mechanism, $\eta(\Delta)$ is known to become independent on time, approaching the value $\eta(\Delta) \to \sqrt{\mathrm{EB}_{\mathrm{CTRW}}}$ for $0 < \alpha < 1$.[125,126] Evaluating expression (100) for superdiffusive and superballistic exponents, we find that for the biased CTRW processes with $1 < \alpha < 2$ the spreading parameter scales as $\eta(\Delta) \sim \Delta^{-(\alpha-1)/2}$, while for $\alpha > 2$ the scaling is $\eta(\Delta) \sim \Delta^{-1/2}$.

Here we also mention the non-equilibrium transport in multi-particle systems (as flux or current) described *via* the paradigmatic asymmetric simple exclusion process. This includes exact results obtained *via* Bethe's Ansatz for open-boundary systems for flux and its variance (as first and second ensemble-averaged moments).[212–214] Note, however, that the WTD in this situation is often exponential[215] and the system contains multiple particles on a finite interval which are exchanging with a reservoir. Our CTRW approach is based on power-law WTDs and delivers the results for infinite systems in the long-time limit both on the level of ensemble and time averaged MSD, as well as for the ergodicity breaking parameter. Combinations of scale-free waiting times and exclusion processes may become quite complex, but are definitely worth a deeper consideration.

Extending the current approach to CTRWs with tempered or truncated[188] power-law WTDs

$$\psi_{\mathrm{tr}}(\tau) \sim \frac{\exp[-\tau/\tau_{\mathrm{c}}]}{\tau^{1+\alpha}}, \tag{101}$$

as well as implications of memory[209,210] can be interesting to unveil. The problems of front propagation and first-passage time statistics[211] can be posed for this biased system too. Moreover, in the spirit of anomalous non-ergodic processes with space-dependent diffusivity,[192–194]

$$D(x) \sim |x|^{\frac{2(\beta-1)}{\beta}}, \tag{102}$$

which feature an MSD growth of the form (1), a dependence of the WTD exponent and short time scale on the particle position—namely, $\alpha \to \alpha(x)$ and $\tau_0 \to \tau_0(x)$—may also be considered. This generalisation mimics local random[98] or systematic heterogeneities[156] of the underlying physical medium (diffusion substrate). Note here that effects of equilibrium and ordinary ensembles on the displacement characteristics for biased deterministic superdiffusion were considered in ref. 182.

Abbreviations

WTD	Waiting time distribution
CTRW	Continuous-time random walk
MSD	Mean squared displacement
RSD	Relative standard deviation
WEB	Weak ergodicity breaking

Conflicts of interest

There are no conflicts to declare.

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