Analyzing anomalous diffusion processes: the distribution of generalized diffusivities

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Abstract: We present applications of a new tool for analyzing data from normal or anomalous diffusion processes: the distribution of generalized diffusivities $p_\alpha(D,\tau)$. It is defined as the probability density of finding in a time interval τ a naturally rescaled squared displacement, a generalized diffusivity, of size D. If the parameter α is chosen as the exponent of the asymptotic increase of the mean squared displacement, the first moment of this distribution becomes for large τ the generalized mean diffusivity $\langle D_\alpha \rangle$. Correspondingly $p_\alpha(D,\tau)$ describes also fluctuations of the diffusivity during the diffusion process. Since this distribution characterizes such processes in much more detail than only the mean squared displacement, and because it can be obtained easily from numerical or experimental data, we propose it as a standard tool for evaluating e.g. single particle tracking experiments. In this contribution we apply it to subdiffusive continuous time random walks and show how weak ergodicity breaking manifests itself in this distribution. Further, we investigate $p_\alpha(D,\tau)$ for anomalously enhanced diffusive transport in simple low dimensional Hamiltonian systems. We show that here $p_\alpha(D,\tau)$ itself does not become stationary, but can be decomposed into various components, each getting stationary for a different scaling parameter $\boldsymbol{\alpha}$, respectively. With the help of an analysis via the distribution of generalized diffusivities we can identify which structures in phase space contribute significantly to the anomalous chaotic transport in these systems.

Keywords: Anomalous transport, Brownian motion, continuous time random walk, chaotic behaviour, random walk, time series analysis.

1. INTRODUCTION

Many diffusion processes occurring in nature do not obey the well-known laws of normal diffusion. In particular, the mean squared displacement (MSD) deviates from the linear time dependence and follows asymptotically a power law.

$$\left\langle \Delta x^2(\tau) \right\rangle \sim \left\langle D_\alpha \right\rangle \tau^\alpha. \quad (1)$$

For $\alpha < 1$ we have subdiffusive behavior, which is observed in many physical, chemical, and biological systems. Concrete examples are charge-carrier transport in amorphous semiconductors (Scher & Montroll 1975), diffusion in living cells and membranes (Golding & Cox 2006), and also diffusion generated by intermittent nonlinear maps (Geisel & Thomae 1984). However, it should be noted that the origin of subdiffusion is not the same for all these systems. In the literature there exists different models which lead to subdiffusive behavior (Klages et al. 2008). One of the most prominent ones is the continuous time random walk (CTRW) (Metzler & Klafter 2000). On the other hand, superdiffusive behaviour, i.e. $\alpha > 1$, is found e.g. in turbulent transport (Shlesinger et al. 1987) and for chaotic diffusion in Hamiltonian systems (Geisel et al. 1987, 1988, Zaslavsky 2002). Often only the diffusion exponent α or the (generalized) diffusion constant $\left\langle D_\alpha \right\rangle$ is determined experimentally, but these numbers are very limited in distinguishing between the various physical mechanisms leading to normal or anomalous diffusion. Therefore in Bauer et al. (2011) we proposed to consider also the fluctuations of the diffusivity along a single trajectory or in an ensemble of normally diffusing particles. This was achieved by introducing the distribution of diffusivities $p_1(D,\tau)$, which can be considered as a histogram for the occurrence of the rescaled squared displacement $\Delta x^2(\tau)/\tau^\alpha$ with $\alpha = 1$. Here we generalize this idea to anomalous diffusion, i.e. $\alpha \neq 1$. The corresponding distribution of generalized diffusivities $p_\alpha(D,\tau)$ is introduced below and subsequently investigated for two cases of anomalous diffusion, the subdiffusive continuous time random walk and the superdiffusive case of chaotic transport in Hamiltonian systems.

2. THE DISTRIBUTION OF GENERALIZED DIFFUSIVITIES

The distribution of generalized diffusivities $p_\alpha(D,\tau)$ is defined as the probability density of finding within a time interval τ a rescaled squared displacement $\Delta x^2(\tau)/\tau^\alpha$ of size D. Formally it can therefore be expressed using the Dirac δ-function by

$$p_\alpha(D,\tau) = \langle \delta(D - \Delta x^2(\tau)/\tau^\alpha) \rangle, \qquad (2)$$

where $\Delta x(\tau) = x(t+\tau) - x(t)$ is the displacement of a random walker achieved in a time span τ. The angular brackets $\langle \ldots \rangle$ denote either a time average over a time span T, or an ensemble average, being identical for ergodic systems, whereas for non-ergodic systems we distinguish them, where necessary, by a subscript T or E, respectively. Often α is chosen as the exponent of the asymptotic increase of the mean squared displacement. In this case the first moment of this distribution is the, in general time-dependent, generalized diffusion coefficient i.e.

$$\int_0^\infty D\, p_\alpha(D,\tau)\, \mathrm{dD} \equiv \langle D_\alpha(\tau) \rangle \equiv \langle \Delta x^2(\tau) \rangle / \tau^\alpha. \qquad (3)$$

Obviously, for large time differences τ this expression becomes stationary and converges to the long-time generalized diffusion coefficient $\langle D_\alpha \rangle$ appearing in Eq.(1), i.e. $\lim_{\tau\to\infty} \langle D_\alpha(\tau) \rangle \equiv \langle D_\alpha \rangle$. It may happen that $\langle D_\alpha(\tau) \rangle$ does not become constant only asymptotically, but is totally independent of the time lag τ, i.e. $\langle D_\alpha(\tau) \rangle = \langle D_\alpha \rangle\ \forall\tau$, despite a still time-dependent distribution $p_\alpha(D,\tau)$. Such a case was observed for composite Markov processes in Bauer et al. (2011). It may happen, however, that also the full distribution $p_\alpha(D,\tau)$ does not at all depend on τ, $p_\alpha(D,\tau) = p_\alpha(D)$. This occurs e.g. for simple diffusion processes such as d-dimensional Brownian motion (Wiener process), where one easily verifies that

$$p_1(D,\tau) = p_1(D) \propto D^{-1+d/2} \mathrm{e}^{-D/(2\langle D_1 \rangle)}. \qquad (4)$$

This can be regarded as a consequence of the fact that the distribution $p_\alpha(D,\tau)$ from Eq.(2) can be viewed as a naturally rescaled version of the (symmetric) propagator $p(x,t)$ in dimension $d = 1$, $p_\alpha(D,\tau) = \sqrt{\tau^\alpha/D}\, p(\sqrt{D\tau^\alpha},\tau)$, or in higher dimensions, of the isotropic van Hove self-correlation function, Hansen & McDonald (2006). An obvious advantage of using for a data analysis the distribution of diffusivities $p_\alpha(D,\tau)$ instead of the propagator or the van Hove self-correlation function itself, lies in its simple relation to the conventionally investigated diffusion coefficients, Eq.(3), and its ability to discriminate between different process classes already via its stationarity properties. This will be explicitly demonstrated below for two concrete examples of anomalous diffusion.

3. SUBDIFFUSIVE CONTINUOUS TIME RANDOM WALKS

Continuous time random walks were established as models for anomalous diffusion several decades ago (Scher & Montroll 1975, Metzler & Klafter 2000). Subdiffusive cases found strong interest recently with the observation that they show the phenomenon of weak ergodicity breaking (Bel & Barkai, 2005). They are generated by a sequence of jumps (jump lengths x_i distributed according to a symmetric distribution $f(x)$ with finite second moment) interrupted by waiting events of duration t_i (distributed according to $\psi(t)$ with diverging first moment, in our simulations $\psi(t) = \alpha(t+1)^{-\alpha-1}$, $0 < \alpha < 1$). A very striking consequence is that the ensemble averaged mean squared displacement behaves anomalous, as expected in accordance with Eq.(1) with $\alpha < 1$, the time average, however, behaves normal, i.e. one has $\alpha = 1$, and the corresponding diffusion coefficient $\langle D_1 \rangle$ is an averaging-time T dependent random variable, meaning that it varies from trajectory to trajectory (Lubelski et al. 2008, He et al. 2008). Obviously it follows from Eq.(3) that also the distribution $p_1^{\mathrm{T}}(D,\tau) = \langle \delta(D - \Delta x^2(\tau)/\tau) \rangle_{\mathrm{T}}$ is a random variable (function), whereas $p_\alpha^{\mathrm{E}}(D,\tau) = \langle \delta(D - \Delta x^2(\tau)/\tau^\alpha) \rangle_{\mathrm{E}}$ is not. There exist additional striking differences. It turns out that the distribution of generalized diffusivities obtained from ensemble averages is τ-independent in the diffusion limit of the CTRW $p_\alpha^{\mathrm{E}}(D,\tau) = p_\alpha^{\mathrm{E}}(D)$, whereas the trajectory dependent function $p_1^{\mathrm{T}}(D,\tau)$ remains explicitly dependent on τ in the allowed τ-range. This is shown in Figs.1 and 2 for a case with $\alpha = 1/2$.

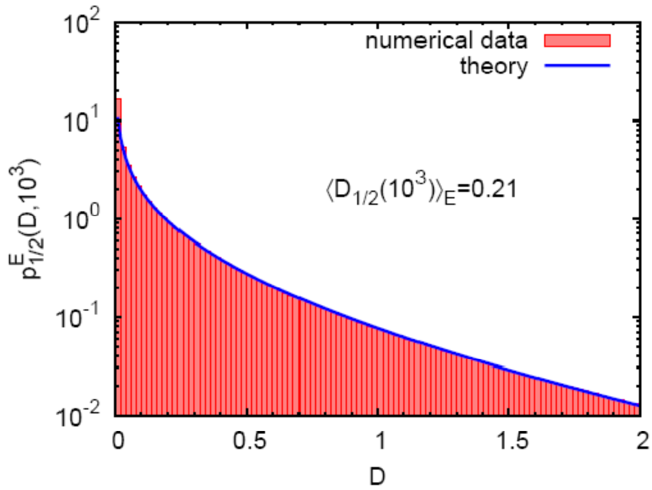

Fig.1: The distribution $p_{1/2}^{E}(D,\tau=10^3)$ of generalized diffusivities obtained from ensemble averaging for a CTRW with $\alpha = 1/2$, in comparison with the analytical result for $p_{1/2}^{E}(D)$.

Numerical results for $p_{1/2}^{E}(D,\tau=10^3)$ are compared in Fig.1 with an analytical result for $p_{1/2}^{\mathrm{E}}(D)$, which was obtained from a rescaling of the propagator $p(x,t)$:

$$p_{1/2}^{\mathrm{E}}(D) = \sqrt{\tau^{1/2}/D}\, p(\sqrt{D\tau^{1/2}},\tau) \qquad (5)$$

The propagator $p(x,t)$ is in the diffusion limit of the CTRW the solution of an associated fractional diffusion equation and can be expressed by a Fox H-function (Eq.(43) in Metzler & Klafter 2000). In contrast, the behavior and the appearance of $p_1^T(D,\tau)$ is quite different as is shown in Fig.2. It obviously consists of two components, a discrete, singular contribution at D = 0, and a continuous part, which becomes narrower with increasing values of τ.

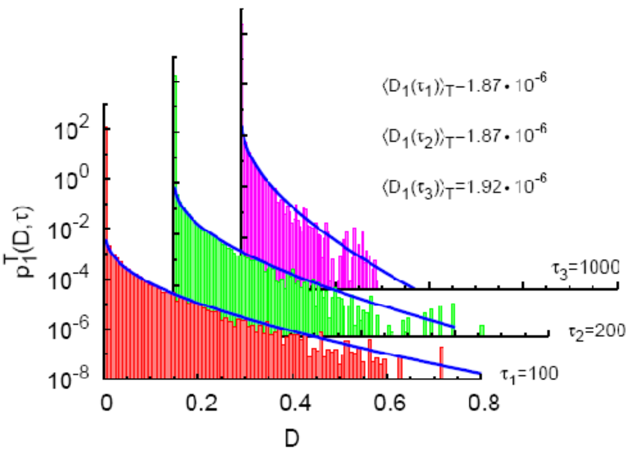

Fig.2: Numerically obtained histograms and the analytically obtained τ-dependence, Eq.(6), for the distribution $p_1^T(D,\tau)$ of the system considered in Fig.1. The first moment $\langle D_1(\tau)\rangle_T$ of the shown distributions is τ-independent ($\approx 1.9\ 10^{-6}$).

In addition the function is random, i.e. depends on the realization of the trajectory, but in a simple way: only the relative weight of the two components changes if the time-average is evaluated from another realization of the CTRW path. The shape of the continuous component is found numerically to be a properly rescaled version of the function $p_{1/2}^{\mathrm{E}}(D)$ displayed in Fig.1. By demanding that the first moment of $p_1^T(D,\tau)$ is equal to the random variable $\langle D_1\rangle_{\mathrm{T}}$, we find for general $\alpha < 1$ and for $\tau \ll T$ that

$$p_1^T(D,\tau) = \left(1-\tau^{1-\alpha}\frac{\langle D_1\rangle_{\mathrm{T}}}{\langle D_\alpha\rangle_{\mathrm{E}}}\right)\delta(D) + \tau^{1-\alpha}\frac{\langle D_1\rangle_{\mathrm{T}}}{\langle D_\alpha\rangle_{\mathrm{E}}}[\tau^{1-\alpha}p_\alpha^{\mathrm{E}}(D\tau^{1-\alpha})]. \qquad (6)$$

The weight of the singular part is the fraction of time (of the record of length T), for which between two time instances with distance τ the state does not change. By noting that according to Eq.(2) a distribution $p_\beta(D,\tau)$ with time scaling index β can be expressed equivalently by $p_\alpha(D,\tau)$ through rescaling

$$p_\beta(D,\tau) = \tau^{\beta-\alpha}p_\alpha(\tau^{\beta-\alpha}D,\tau), \qquad (7)$$

one sees that in the continuous component in Eq.(6), the term in square brackets is just the function $\mathrm{p}_{1/2}^{\mathrm{E}}(\mathrm{D})$ expressed with time scaling index $\beta = 1$. This means that the non-zero fluctuations in the (normal) diffusivity obtained from a single time series can be obtained from the fluctuations of the generalized diffusivity observed in an ensemble. Thus the breaking of ergodicity in a subdiffusive CTRW can be attributed to the additional appearance of the singular contribution in $p_1^T(D,\tau)$.

4. CHAOTIC SUPERDIFFUSION IN HAMILTONIAN SYSTEMS

It has long been recognized that chaotic diffusion in Hamiltonian systems is often anomalously enhanced due to the "stickiness" of transporting integrable islands in phase space (Geisel et al. 1987, 1988, Zaslavsky 2002). To see what information about the anomalous transport in such systems is revealed by applying our distribution of generalized diffusivities $p_\alpha(D,\tau)$, we treat as simple example diffusion in the two-dimensional standard map (Zaslavsky 2002) given by the following iteration $(x_{t+1},p_{t+1}) = (x_t + p_{t+1}, p_t + k/(2\pi)\sin(2\pi x_t))$ in discrete time t. For $k < k_c = 0.97\ldots$ one observes chaotic diffusion in the (extended) position coordinate x, whereas the motion in the momentum coordinate p is bounded. One finds numerically that the mean squared displacement in the position increases according to Eq.(1) with $\alpha = 1.74\ldots$, i.e. superdiffusively. We determine numerically for $k = 0.5$ the distribution $p_\alpha(D,\tau)$, and find the results displayed in Figs.3-5.

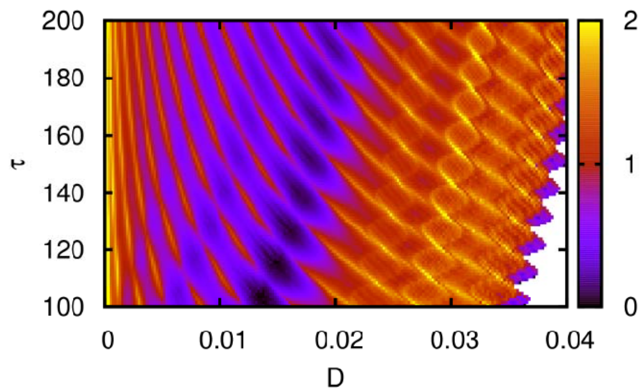

Fig.3: The distribution $p_\alpha(D,\tau)$ (logarithmic height scale, color coded) for the diffusion process in the position coordinate of the standard map.

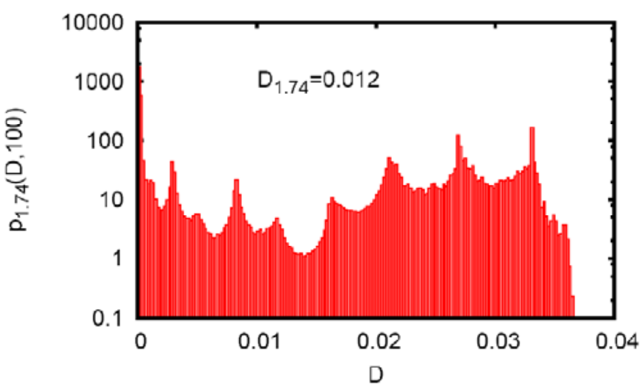

Fig.4: A cut through the distribution $p_\alpha(D,\tau)$ of Fig.3 at $\tau = 100$ exhibits a complicated multimodal structure.

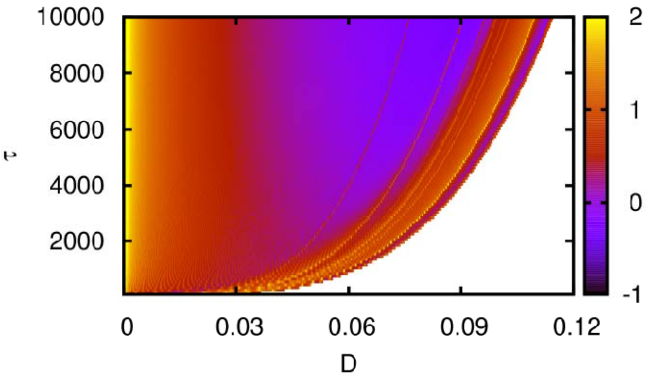

Fig.5: Same as Fig.3, but on a larger time scale.

One finds for small values of τ (Figs. 3, 4) a complicated multimodal structure, which vanishes for large τ (Fig. 5). In the latter region some peaks with a D-t dispersion very different from those in Fig.3 are found. To identify the origin of these structures it is helpful to transform the distribution $p_\alpha(D,\tau)$ with the aid of Eq.(7) to equivalent distributions $p_\beta(D,\tau)$ with scaling index $\beta = 0$ in Fig.6 and index $\beta = 2$ in Fig.7.

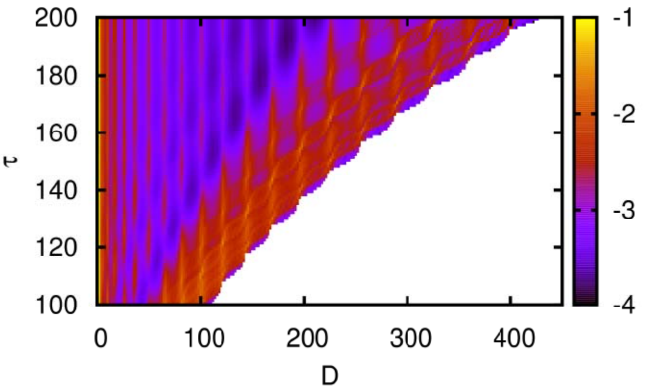

Fig.6: The shown distribution $p_0(D,\tau)$ obtained from $p_\alpha(D,\tau)$ via Eq.(7) reveals localized behavior.

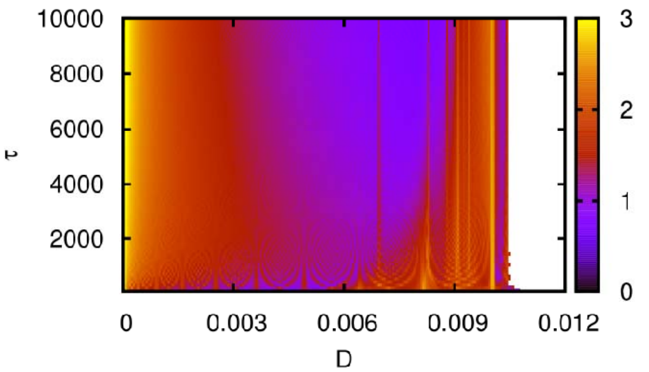

Fig.7: The displayed distribution $p_2(D,\tau)$ obtained from $p_\alpha(D,\tau)$ via Eq.(7) reveals ballistic transport.

The appearance of vertical structures in Fig.6 and Fig.7 imply localized motion or ballistic transport, respectively. The vertical ridges in Fig.6 are at positions $D = n^2, n = 1, 2, 3, \ldots$, which indicate the positions of the main elliptic islands in the extended standard map(at the centers of the unit cells of length one in the x-direction), thereby explaining also the location of the maxima in Fig.4. The vertical ridges in Fig.7, on the other hand, represent ballistic motion induced by island chains of certain rational winding numbers, which can also be identified in the systems phase space. We see that analyzing tracks of anomalously diffusing states with the help of the distribution of diffusivities $p_\alpha(D,\tau)$ and its rescaled equivalents helps to disentangle the various mechanisms responsible for the anomalous transport.

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