

## Lévy statistics in Taylor dispersion

Albert Compte\* and Juan Camacho

*Departament de Física, Física Estadística, Universitat Autònoma de Barcelona, E-08193 Bellaterra, Catalonia, Spain*

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The longitudinal dispersion for a fractal time random walker being dragged by a solvent flowing through a tube is studied by means of the Langevin and Fokker-Planck formalisms. One observes that for asymptotic long times the dispersion is superdiffusive despite the fact that in a resting background the characteristic diffusion regime is subdiffusive. The resulting behavior is also at variance with the standard diffusive behavior obtained in Taylor dispersion for a Brownian walker. [S1063-651X(97)02911-5]

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### I. INTRODUCTION

Lévy statistics appear in two generalizations of Brownian diffusion: fractal time random walks (FTRW's) and Lévy flights. These methods have proved useful to model a variety of physical systems, from transport in amorphous materials [1] or transport of magnetic holes in rotating magnetic fields [2] to diffusion in rotating fluids [3] or turbulent diffusion in plasmas [4], among many other examples [5].

In a previous article [6] we made some connection between FTRW's and Taylor dispersion. There, though, the diffusive mechanism was modeled by means of a generalized continuous time random walk (CTRW) formalism [7], where the particles remained fixed between successive jumps (and were not driven by the underlying flow during these periods). It was seen there that Taylor dispersion appeared only as a second-order phenomenon for long times.

A more realistic picture for Lévy diffusion in a flowing fluid, with the particles being continuously dragged by the stream, therefore remains to be proposed. An important question would then be whether, when such a system is restrained to flow through a pipe, Taylor dispersion turns out to be the relevant dispersion mechanism for long times, i.e., whether the coupling of convection and transverse diffusion brings about the leading term for the long time mean square displacement.

A possible physical realization of such a system could be diffusion of macromolecules in flowing polymeric solutions. In these systems, the entanglement of the diffusing macromolecules with the complex structures in the matrix has been modeled in terms of a trapping time distribution with infinite moments [8] and has been checked experimentally for gel electrophoresis [9].

We intend here to approach this question by using the Langevin equation combined with the diffusion equation associated with FTRW's. The use of the Langevin formalism for diffusion in nonhomogeneous flows is not new [10–12]. Here we shall adapt the methods that were presented in [12] for the case of Brownian diffusion in Taylor dispersion in order to allow for Lévy statistics in the waiting time distribution of the walker.

The structure of the paper is as follows. In Sec. II we deal

with the Langevin equation for fractal time random walkers in the absence of flow. In Sec. III we generalize it to include the drag due to a solvent flowing through a tube and we compute the mean square displacement in the flow direction with the help of a Fokker-Planck-like equation describing the transverse diffusion of the Lévy walker. Section IV contains the conclusions.

### II. LANGEVIN EQUATION FOR FTRW'S

Our first objective here is to establish the form of a Langevin equation associated with a FTRW. To this aim we recall the usual description of these movements within the CTRW formalism, where the probability density of the walker who started at  $t=0$  from  $x=0$  is, in the Fourier-Laplace domain,

$$\rho(k, u) = \frac{1}{u} \frac{1 - \varphi(u)}{1 - \psi(k, u)},$$

$\psi(k, u)$  being the Fourier-Laplace transform of the distribution of step lengths and waiting times of the walker and  $\varphi(u) = \psi(k=0, u)$  the Laplace transform of the distribution of waiting times at a site. From this result it is straightforward to obtain the mean squared displacement of the walker in terms of the distribution of step lengths and waiting times as

$$\langle \Delta x^2 \rangle = - \left. \frac{\partial^2 \rho(k, u)}{\partial k^2} \right|_{k=0} = - \frac{1}{u} \left. \frac{1}{1 - \varphi(u)} \frac{\partial^2 \psi(k, u)}{\partial k^2} \right|_{k=0},$$

where we assumed that  $\psi(x, t)$  is analytic and symmetric in  $x$ . If we now choose a Gaussian form for the step length distribution  $\psi(k, u) = \varphi(u) \exp(-\sigma^2 k^2)$ , the mean square displacement of the random walker in continuous time is

$$\langle \Delta x^2 \rangle = 2\sigma^2 \frac{1}{u} \frac{\varphi(u)}{1 - \varphi(u)}. \quad (1)$$

This result will be our main connection with the Langevin equation, which we propose in the form

$$\frac{dx}{dt} = v(t), \quad (2)$$

\*Electronic address: albert@telemaco.uab.es

where  $v(t)$  is a fluctuating velocity with vanishing mean  $\langle v(t) \rangle = 0$ . From Eq. (2) the mean square displacement  $\langle \Delta x^2 \rangle$  can be written as

$$\langle \Delta x^2 \rangle = \int_0^t dt' \int_0^t dt'' \langle v(t') v(t'') \rangle, \quad (3)$$

whence

$$\frac{d\langle \Delta x^2 \rangle}{dt} = 2 \int_0^t dt' \langle v(0) v(t') \rangle, \quad (4)$$

where we assume that the velocity correlation function  $F(t-t') = \langle v(t) v(t') \rangle$  depends just on the difference of times and is a symmetric function of its argument (assumption of stationarity). At this point our aim is to find an expression for the velocity correlation function  $F(t)$  for a walker obeying a statistics of waiting times given by  $\varphi(t)$ . To this end we can write Eq. (4) in the Laplace domain and obtain

$$u \langle \Delta x^2 \rangle(u) = 2 \frac{F(u)}{u}, \quad (5)$$

assuming a vanishing initial dispersion. We now combine Eqs. (1) and (5) to get the Laplace inversion of the velocity correlation function of a CTRW with a Gaussian distribution of step lengths and an arbitrary distribution of waiting times  $\varphi(t)$ ,

$$F(u) = \sigma^2 u \frac{\varphi(u)}{1 - \varphi(u)}. \quad (6)$$

We first prove this expression for a Brownian random walker, where  $\varphi(t) = \delta(t - \tau)$ ,  $\tau$  being the fixed waiting time between steps. We thus have  $\varphi(u) = \exp(-u\tau)$  and the velocity correlation function given by Eq. (6) turns out to be, in the Laplace domain,

$$F(u) = D_1 \frac{u\tau}{e^{u\tau} - 1},$$

with  $D_1 = \sigma^2/\tau$ . To compute the Laplace inverse of this expression we just keep the three lowest orders of the exponential as  $u\tau \rightarrow 0$  (we therefore assume observation times much longer than the microscopic waiting time  $\tau$ :  $t \gg \tau$ ) and get

$$F(t) = \frac{2D_1}{\tau} e^{-2t/\tau}. \quad (7)$$

This result can now be compared with the velocity correlation function for a Brownian walker as described by the standard Langevin equation for a particle in a viscous medium [13] and we identify the friction coefficient in Eq. (7) as  $2/\tau$ .

We now want to find the velocity correlation function associated with a FTRW. To this aim we only have to introduce in Eq. (6) a waiting time distribution function  $\varphi(t)$  with an infinite first moment (undefined mean waiting time). The absence of a characteristic waiting time scale suffices to introduce anomalies in the diffusion process, following the properties of Lévy statistics [14]. Many choices are possible

and here we will propose one that most directly generalizes our previous derivations: We take the stable law  $\varphi(u) = \exp[-(u\tau)^\gamma]$ , with  $0 < \gamma < 1$ . One then has the asymptotic subdiffusive behavior typical for FTRW's:  $\langle \Delta x^2 \rangle = 2D_\gamma t^\gamma/\Gamma(1+\gamma)$ , with  $D_\gamma = \sigma^2/\tau^\gamma$ . The Laplace transform of the associated velocity correlation function is given by formula (6) and, keeping as before just the three lowest orders in  $u\tau$  to compute  $\exp(u\tau)^\gamma$ , one has

$$F(u) = D_\gamma u^{1-\gamma} \frac{u^{1-\gamma}}{1 + \frac{1}{2}\tau^\gamma u^\gamma}.$$

The Laplace inversion of this function is carried out with the help of generalized Mittag-Leffler functions (see, for instance, [6])

$$F(t) = \frac{2}{\tau^\gamma} D_\gamma t^{2\gamma-2} E_{\gamma, 2\gamma-1} \left[ -2 \left( \frac{t}{\tau} \right)^\gamma \right], \quad \frac{1}{2} < \gamma < 1, \quad (8)$$

where we restrict further our parameter  $\gamma$  in order to express  $F(t)$  in terms of Mittag-Leffler functions; in the range  $0 < \gamma < \frac{1}{2}$  we instead obtain more complex functions (Fox's  $H$  functions; see [6]). Therefore, expression (8) describes the second moments of the stochastic variable  $v$  in our Eq. (2). The result (8) has the property of having at long times a negative long tail  $F(t) \sim -t^{\gamma-2}$ , a known result in the theory of anomalous diffusion [15].

### III. FTRW'S IN TAYLOR DISPERSION

We shall now follow [12] to derive the dispersion properties of FTRW's undergoing Taylor dispersion. To this aim we imagine a longitudinal steady flow between two parallel plates at  $y=0$  and  $y=l$  given by the velocity field  $V_x(y)$ , where we release a tracer whose diffusion is governed by Lévy statistics as a FTRW. The objective is to establish the longitudinal dispersion behavior of the tracer, that is, in the flow direction. As in [12], independent motion in the  $x$  and  $y$  directions is assumed and diffusion is taken to be isotropic in a fluid at rest, whence the same diffusivity  $D_\gamma$  applies to each direction. So one incorporates in the Langevin equation (2) for the  $x$  direction the drag due to the solvent

$$\frac{dx}{dt} = V_x(y(t)) + v(t), \quad (9)$$

which leads to

$$\begin{aligned} \langle \Delta x^2 \rangle = & \int_0^t dt' \int_0^t dt'' [\langle V_x(y(t')) V_x(y(t'')) \rangle - \langle V_x(y(t')) \rangle \\ & \times \langle V_x(y(t'')) \rangle] + \int_0^t dt' \int_0^t dt'' \langle v(t') v(t'') \rangle. \end{aligned} \quad (10)$$

As seen in Sec. II, the second term in Eq. (10) provides  $2D_\gamma t^\gamma/\Gamma(1+\gamma)$ , namely, the diffusion in the absence of a velocity field. The first integral, on the other hand, describes the dispersion due to the coupling between transverse diffusion and the velocity profile, i.e., Taylor dispersion. In order to perform the averages appearing in Eq. (10), we need to use the two-time probability distribution function

$$P_2(y, t; y', t') = P(y, t - t' | y') P(y', t'),$$

with  $P(y, t - t' | y')$  the conditional probability distribution and  $P(y', t')$  the one-time probability density. For the sake of simplicity, we consider a uniform initial distribution of tracer particles along  $y$  at  $x=0$ , so that for all times one has  $P(y', t') = 1/l$ .

Therefore, our first calculations will be aimed at obtaining the conditional probability distribution of particles with coordinate  $y$  at time  $t$  given the fact that at time  $t'$  they were in  $y'$ :  $P(y, t - t' | y')$ . We then start by using the diffusion equation. Since we are considering Lévy statistics in time, this diffusion equation must correspondingly be the one associated with FTRW's, as obtained in [16,17], and written for the  $y$  direction,

$$\frac{\partial P(y, t - t' | y')}{\partial t} = D_\gamma \frac{\partial^{1-\gamma}}{\partial t^{1-\gamma}} \frac{\partial^2 P(y, t - t' | y')}{\partial y^2}, \quad (11)$$

where the operator  $\partial^\alpha / \partial t^\alpha$  stands for the Riemann-Liouville fractional derivative of order  $\alpha$  [18]. We now solve Eq. (11) for the initial condition

$$P(y, 0 | y') = \delta(y - y') \quad (12)$$

and the boundary condition of impenetrability at the plates

$$\left. \frac{\partial P(y, t | y')}{\partial y} \right|_{y=0, l} = 0. \quad (13)$$

The solution will be a superposition of modes, whose spatial dependences in the present problem are  $\cos(n\pi y/l)$ ,  $n=0, 1, 2, \dots$ . Then, we define the Fourier coefficients of  $P(y, t | y')$  as

$$P_n(t | y') = \frac{2}{l} \int_0^l P(y, t | y') \cos \frac{n\pi y}{l} dy$$

and compute their first derivative with the help of Eqs. (11) and (13) and repeated integration by parts to obtain

$$\frac{\partial P_n(t | y')}{\partial t} = -D_\gamma \frac{n^2 \pi^2}{l^2} \frac{\partial^{1-\gamma} P_n(t | y')}{\partial t^{1-\gamma}}. \quad (14)$$

Thus  $\tau_n \equiv (n^2 \pi^2 D_\gamma / l^2)^{-1/\gamma}$  gives the decaying time of the  $n$ th diffusive mode. Equation (14) is now easy to solve in the Laplace domain, yielding

$$P_n(u | y') = \frac{u^{-1}}{1 + (\tau_n u)^{-\gamma}} P_n(t=0 | y'),$$

which has a direct inversion in the form of a generalized Mittag-Leffler function (see, for instance, [6]) as

$$P_n(t | y') = \frac{2}{l} E_{\gamma, 1} \left[ - \left( \frac{t}{\tau_n} \right)^\gamma \right] \cos \frac{n\pi y'}{l}, \quad (15)$$

where  $E_{\gamma, n}(x) = \sum_{m=0}^{\infty} x^m / \Gamma(\gamma m + n)$ . In Eq. (15) we have written already the explicit form of  $P_n(t=0 | y')$  for our particular initial condition (12). The solution for our conditional probability distribution is therefore

$$P(y, t | y') = \frac{1}{l} + \frac{2}{l} \sum_{n=1}^{\infty} E_{\gamma, 1} \left[ - \left( \frac{t}{\tau_n} \right)^\gamma \right] \cos \frac{n\pi y}{l} \cos \frac{n\pi y'}{l}. \quad (16)$$

Now, with the help of Eq. (16), the averages in Eq. (10) yield

$$\langle V_x(t) \rangle = \overline{V_x}, \quad (17)$$

$$\langle V_x(t) V_x(t') \rangle = \overline{V_x^2} + \frac{1}{2} \sum_{n=1}^{\infty} v_n^2 E_{\gamma, 1} \left( - \frac{|t-t'|^\gamma}{\tau_n^\gamma} \right), \quad (18)$$

where we drop the condition  $t > t'$  by introducing the absolute value in Eq. (18),  $\overline{V_x}$  stands for the section average of the velocity field  $V_x(y)$  and  $v_n$  is the  $n$ th Fourier coefficient of  $V_x(y)$ ,

$$v_n = \frac{2}{l} \int_0^l V_x(y) \cos \frac{n\pi y}{l} dy.$$

Combining Eq. (10) with the results (17) and (18) we find

$$\begin{aligned} \langle \Delta x^2 \rangle(t) &= \sum_{n=1}^{\infty} v_n^2 \int_0^t dt'' \int_{t''}^t dt' E_{\gamma, 1} \left[ - \left( \frac{t' - t''}{\tau_n} \right)^\gamma \right] \\ &\quad + \frac{2D_\gamma t^\gamma}{\Gamma(1 + \gamma)}. \end{aligned}$$

By first resorting to the Laplace domain and then inverting the resulting expressions, the dispersion of the solute becomes

$$\langle \Delta x^2 \rangle(t) = \frac{2D_\gamma t^\gamma}{\Gamma(1 + \gamma)} + \sum_{n=1}^{\infty} v_n^2 t^2 E_{\gamma, 3} \left[ - \left( \frac{t}{\tau_n} \right)^\gamma \right]. \quad (19)$$

Let us note that at long times, i.e., for  $t \gg (\pi^2 D_\gamma / l^2)^{-1/\gamma}$ , one can retain only the leading term in the function  $E_{\gamma, 3}(x) \approx x^{-1} / \Gamma(3 - \gamma)$ , so that Eq. (19) yields

$$\langle \Delta x^2 \rangle(t) = \frac{2D_\gamma t^\gamma}{\Gamma(1 + \gamma)} + \frac{2}{\Gamma(3 - \gamma)} D_T(\gamma) t^{2-\gamma} + O(t^{2-2\gamma}), \quad (20)$$

with  $D_T(\gamma) \equiv \frac{1}{2} \sum_{n=1}^{\infty} v_n^2 \tau_n^\gamma$  a generalized Taylor dispersion coefficient [19], since it involves both the velocity of the flow and the diffusion constant  $D_\gamma$  and it therefore represents the typical coupling of advection and transverse diffusion in Taylor dispersion. It is interesting to notice that the Taylor dispersion contribution is not diffusive, but it grows as  $t^{2-\gamma}$  in contrast to what is found for Brownian diffusion or persistent random walks [20].

We observe in Eq. (20) that the regime is superdiffusive since  $0 < \gamma < 1$ . It may seem at first sight paradoxical that the slower the transverse diffusion (smaller  $\gamma$ ), the faster the longitudinal dispersion. Let us remember that this situation is analogous to the inverse proportionality of the Taylor dispersion coefficient  $D_T$  with the molecular diffusivity  $D$  in standard Taylor dispersion:  $D_T \propto D^{-1}$ . The interpretation is also the same: The slower the transverse diffusion, the longer the fast particles remain in the fast layers and the slow particles

in the slow ones, so that the rate at which they separate longitudinally from each other is faster.

Let us also note that the dispersion coefficient  $D_T(\gamma)$  is basically the same as in standard Taylor dispersion except that  $D_\gamma$  replaces  $D$ . Then we have, in general,  $\Delta x^2 \sim \overline{V_x^2} l^2 D_\gamma^{-1} t^{2-\gamma}$ , which establishes how  $\Delta x^2$  scales with the section size, the average velocity, and the transverse diffusion coefficient. It can also be observed that, contrarily to Brownian Taylor dispersion,  $D_T$  and  $D_\gamma$  do not have the same dimensions, this being a consequence of the fact that they are associated with different diffusion regimes (superdiffusion and subdiffusion, respectively).

Let us examine the limit cases. As  $\gamma \rightarrow 1$ , one recovers the diffusive dispersion, namely,  $\langle \Delta x^2 \rangle = 2(D_1 + D_T)t$ . As  $\gamma \rightarrow 0$ , that is, when the jumps between layers are rare, one expects that the tracer essentially follows the velocity profile and hence  $\langle \Delta x^2 \rangle \propto t^2$ , as it indeed ensues from Eq. (20).

At short times, on the other hand, one has  $E_{\gamma,3}(x \rightarrow 0) \rightarrow 1$  and Eq. (19) yields

$$\langle \Delta x^2 \rangle = \frac{2D_\gamma t^\gamma}{\Gamma(1+\gamma)} + \overline{V_x^2} t^2 + O(t^{2\gamma}).$$

The first term corresponds to diffusion in the absence of a flow and the second one is the drag for an initially uniform distribution of solute at  $x=0$ .

At this point we can now compare with the results of [6], where the leading term for the dispersion at long times [again  $t \gg (\pi^2 D_\gamma / l^2)^{-1/\gamma}$ ] grew as  $t^{2\gamma}$ . The macroscopic setup is very much the same in this paper and in [6], but the essential difference lies on the stochastic model used to implement the FTRW: In [6] the diffusing particles remain fixed in space, transparent to the fluid stream, during the waiting periods between successive steps. This was effectively seen in [6] to explain the asymptotic behavior  $\langle \Delta x^2 \rangle \propto t^{2\gamma}$  on the grounds of purely advective phenomena, so that the essential ingredients of Taylor dispersion were not present in this leading term. In the current paper, in contrast, the tracer particles are continuously being dragged by the flow and this yields a different dispersion law  $\langle \Delta x^2 \rangle \propto t^{2-\gamma}$ , always superdiffusive since  $0 < \gamma < 1$  and associated with the coupling of advection and transverse diffusion (Taylor dispersion). Interesting from this comparison is the fact that  $\gamma = 2/3$  marks the transition from a situation in which the model of [6] is more efficient in dispersing the tracer ( $\gamma > 2/3$ ) than the model presented here, and the converse situation for  $\gamma < 2/3$ . The existence of such a transition is indeed logical since, assuming the extremal values for  $\gamma$ , it becomes clear that for a FTRW with  $\gamma \approx 1$  (almost Brownian) pure convection is much more efficient for dispersion

than Taylor dispersion and, conversely, when  $\gamma \approx 0$  the tracer in [6] is practically permanently static whereas here it keeps advancing with the stream in an almost convective manner. The fact that this ballistic limit is attained for different extremal values of  $\gamma$  here and in [6] is quite remarkable. It is also to be noted that in [6] a macroscopic parameter  $A$  must be included in order to ensure a good correspondence between the stochastic and the macroscopic results. This parameter does not appear here and should be accounted for when treating the model of this article with the stochastic formalism of [6].

Finally, we stress that the results obtained in the present work are not restricted to flows between parallel plates, but the calculations can be easily generalized to arbitrary section geometries. The spatial dependence of the diffusive modes and their decaying times would change accordingly, but the dispersion would still be given by Eqs. (19) and (20).

#### IV. CONCLUSIONS

We have calculated the mean square displacement for a fractal time random walker that is suspended in a solvent flowing through a tube. To this end, we have performed a stochastic analysis by using a Langevin equation and a Fokker-Planck-like equation for the transverse diffusion of the walker. One finds for asymptotic long times that the dispersion is mainly due to the coupling of advection and transverse diffusion, whence we have an example of Taylor dispersion. Furthermore,  $\langle \Delta x^2 \rangle$  has been seen to grow asymptotically as  $t^{2-\gamma}$  and hence the dispersion is superdiffusive for  $\gamma < 1$ , in contrast to the diffusive growth usually displayed in Taylor dispersion, where the solvent diffuses in the solvent in a Brownian way. When  $\gamma$  tends to 1 (Brownian diffusion) one recovers the standard diffusive behavior. For  $\gamma \rightarrow 0$  the dispersion approaches a ballistic regime, as corresponds to the absence of transverse diffusion. Although the calculations have been performed in this work for flows between parallel plates for the sake of simplicity, they can be easily adapted for arbitrary section geometries and the results are therefore general.

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