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TAYLOR DISPERSION ON A FRACTAL*

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Taylor dispersion is the greatly enhanced diffusion in the direction of a fluid flow caused by ordinary diffusion in directions orthogonal to the flow. It is essential that the system be bounded in space in the directions orthogonal to the flow. We investigate the situation where the medium through which the flow occurs has fractal properties so that diffusion in the orthogonal directions is anomalous and non-Fickian. The effective diffusion in the flow direction remains normal; its width grows proportionally with the time. However, the proportionality constant depends on the fractal dimension of the medium as well as its walk dimension.

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1. Introduction

Dynamical processes on fractals are rather different from similar process taking place on Euclidean supports. Differences can arise either because the elementary dynamics are fractal (Levy walks, for example) or because the processes take place in a fractal background. In the present paper we shall discuss a particular case of the second of these possibilities.

Diffusion is an important example of a transport process. Normal diffusion is characterized by a dispersion of position of diffusing particles given by $\langle r^2 \rangle \sim t$. Diffusion on fractals is anomalous, with $\langle r^2 \rangle \sim t^{2/d_w}$. d_w is called the dimension of the random walk underlying the diffusion; for walks on a fractal substrate, generally $d_w > 2$. This relation is the definition of d_w; it's value must, of course, be determined individually for each case.

It is of interest to study how diffusion interacts with other transport processes. The case when diffusion takes place in a flowing fluid with a

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velocity gradient has been studied by Taylor [1]. Briefly put, when a solute is placed in a fluid flowing in a tube, the center of the solute distribution moves with the average velocity of the tube cross section. For a delta function initial distribution of solute, the width of the distribution at subsequent long times increases linearly with the time, and the distribution of solute itself is very close to gaussian. I have given a more detailed description of Taylor dispersion at the IV Zakopane Symposium [2].

The physics behind this phenomenon is that as the solute particles get carried along by the fluid flow they also perform diffusive motions with respect to the barycentric motion of the fluid. The diffusive motion in the direction of the fluid flow gives rise to a negligible effect, and we ignore it here. Diffusion perpendicular to the flow, however, means that over the course of time, solute particles are exposed to many different fluid velocities. After a time long compared to the time necessary for a particle to sample the entire velocity distribution of the fluid, each particle acquires a distribution of velocities that mirrors the distribution over the cross section. It is this interaction of transverse diffusion with longitudinal flow which generates the effect described in the preceding paragraph.

It is essential that the system be finite in extent in the orthogonal directions. This is because the longitudinal dispersion only appears to be pseudo-Fickian after the solute has had time to traverse the entire cross section. In an infinite system, this never occurs. For times short compared to the traversal time of the system, when the boundaries have not yet had any appreciable effect on the transverse diffusion, there is anomalous dispersion in the flow direction. We shall not consider that case in this paper, restricting our attention to the long time regime.

Taylor dispersion is interesting because it is a nontrivial example of how a random process (the diffusion) interacts with a deterministic process (the flow). It is also interesting because it is the basis for a good method for measuring diffusion coefficients. Diffusion coefficients in liquids are small. Therefore diffusion experiments of classical type take a long time, and it is difficult to keep the system isothermal, vibration free, *etc.* for long periods. When the solute particles are large, *e.g.* polymers, inelastic light scattering provides a useful alternative method, but for low molecular weight solutes, the optical contrast between solute and solvent is usually not sufficient for an accurate measurement by this method. Taylor dispersion, relying on the greatly enhanced value of the Taylor diffusion coefficient relative to the molecular diffusion coefficient, has been used successfully in this latter case [3].

It is therefore worthwhile to consider variations on Taylor's original idea, extending it to new and different situations. Indeed, there is a large engineering literature on Taylor dispersion, and a growing literature on the phenomenon from a fundamental physicochemical point of view. In this paper we treat the long time longitudinal dispersion of a solute where the transverse dimensions are fractal. We shall see that, in the long time limit, the dispersion in the longitudinal dimension becomes Fickian. However, the effective diffusion coefficient depends on the physical parameters of the system in a different way than is the case when the ambient medium is Euclidean.

2. Diffusion on fractals and scaling considerations

We first review what is known about diffusion on fractals. What we are after is the probability distribution for being at a certain position on the fractal at a certain time. In fact, this is a multifractal, a fractal measure. It has too much fine detail for our purposes; it is irregular on all scales. We need a smoothed, averaged version of this distribution. Knowledge of this averaged distribution is primarily based on numerical simulations of random walks and on scaling arguments.

Actually, the probability distribution for the fractal without boundaries is not of major interest in the present context, although it is the quantity on which all previous authors have focused their attention. We need the equation of evolution satisfied by the distribution function. The infinite space distribution function is of interest only insofar as it indicates whether or not a proposed evolution equation has a possibility of being correct. We shall adopt the evolution equation advocated by O'Shaughnessy and Procaccia [4, 5]. It is

$$\frac{\partial P}{\partial t} = \frac{1}{r^{d-1}} \frac{\partial}{\partial r} \left(K r^{d-d_w+1} \frac{\partial P}{\partial r} \right) . \tag{1}$$

Here, d is the fractal dimension of the fractal substrate, d_w is the walk dimension defined by

$$\langle r^2 \rangle \sim t^{2/d_w} , \tag{2}$$

and K is a constant. This equation has been controversial, and we must justify our choice.

Everyone agrees that $P(r,t)$ has the scaling form

$$P(r,t) = t^{-d_w/2} F(r/t^{1/d_w}) . \tag{3}$$

This is indicated by two considerations. First, the probability of return to the origin is given by $P(0,t) \sim t^{-dw/2}$. $P(0,t)$ clearly determines the prefactor of F, and is determined by its relation to the vibrational density of states of the fractal (regarded as a vibrating body) [6]. Second, (2) gives the second moment of P, so that dimensional considerations suggest that F should have the indicated argument. The question is, what is the form of F?

Banavar and Willemsen [7] proposed, on the basis of dimensional analysis and the Chapman–Kolmogorov equation, that $F(\xi) \sim \exp(-c\xi)$. O'Shaughnessy and Procaccia obtained the same result on the basis of the differential equation (1). Shortly thereafter, Guyer [8] proposed the form $F(\xi) \sim \exp(-c'\xi^{\alpha})$ with $\alpha = d_w/d_w - 1$. His results were based on numerical simulations in a range of values of ξ larger than those used by O'Shaughnessy and Procaccia [4] in checking their conclusions; Guyer, in fact, suggested that different results may be valid in different time regimes. The exponent α has been further discussed by Aharony [9] and by Van den Broeck [10]. The situation as of 1987 has been reviewed by Havlin and Ben-Avraham [11].

The situation has been considerably clarified by numerical work of Klafter *et al.* [12]. These authors find that the O'Shaughnessy-Procaccia form describes their numerical work well for $\xi \ll 1$. For $\xi \gg 1$, they find that $F(\xi) \sim \xi^{\lambda} \exp(-c'\xi^{\alpha})$ with α given by the Guyer form and $\lambda = (d - d_w/2)/(d_w - 1)$. As stated above, the detailed form of F does not concern us here since it refers to an infinite system. The system we deal with is bounded, and we are interested in long time results. Consequently, the variable $\xi = r/t^{1/d_w}$ is small. While we cannot just take over the small ξ form of F because of the effects of the boundaries, it is reasonable to use the evolution equation known to be valid for small ξ, Eq. (1), supplemented by appropriate boundary conditions. The boundary conditions which we shall adopt are that the flux of solute vanishes on the boundaries, or $\partial P/\partial r = 0$ (assuming radial symmetry).

Of the authors whom we have quoted thus far, only Ref. [4] introduces an evolution equation. We should however, mention an equation proposed by Metzler *et al.* [13]. This is a differential equation which is of fractional order with respect to the time; said another way, it is an integrodifferential equation. Metzler *et al.* find an exact solution of their equation in terms of special functions called H functions [14]. This solution has the Guyer asymptotic form for $r/t^{1/d_w} \gg 1$ but without the additional ξ^{λ} term observed in the numerical work of Klafter *et al.* The solution does not have the O'Shaughnessy-Procaccia form for $r/t^{1/d_w} \ll 1$; Furthermore, it does not appear to have the form of an equation of continuity for the probability current. The authors did not derive this equation from physical arguments, but proposed it as a mathematical construct which would yield the previously proposed asymptotic result. Consequently we have not adopted this equation as the starting point for the work reported here.

So far, we have been discussing pure diffusion with no convection. In Taylor dispersion we envisage unidirectional fluid flow with diffusion in directions transverse to the flow. To visualize this, one might think of a Sierpinski gasket moved parallel to itself (a "Toblerone" tube with a Sierpinski gasket cross section). We shall take the evolution equation for this system to be the convective diffusion equation

$$\frac{\partial P}{\partial t} + v_x(r)\frac{\partial P}{\partial x} = \frac{1}{r^{d-1}}\frac{\partial}{\partial r}\left(Kr^{d+1-d_w}\frac{\partial P}{\partial r}\right). \tag{4}$$

Here x is the coordinate in the direction of the flow and r the radial variable in the transverse directions; we assume cylindrical symmetry. The goal of this paper is to derive an evolution equation for the probability density $n(x,t)$, which is the density $P(r,x,t)$ averaged over the cross section of the flowing system.

Before attempting this derivation, we use scaling arguments to see what results might be expected from a more detailed calculation. I am indebted to my colleague John Toner for the following arguments.

Ben-Naim, Redner, and ben-Avraham [15] have shown that a particle undergoing normal diffusion in a flow $v_x = u_0\hat{e}_x y^\beta \mathrm{sgn}(y)$ undergoes anomalous dispersion in the x direction varying as $\langle\delta x^2\rangle \sim u_0^2 D^\beta t^{2+\beta}$. This can be shown exactly for $\beta = 1$ and by approximate arguments for $\beta \neq 1$. It holds when the direction normal to the flow, the y direction is *unbounded.* When boundaries are inserted in the y direction, one expects that proportionality to t will be recovered. Let us assume this. Then, in the presence of boundaries

$$\langle\delta x^2\rangle = u_0^2 D^\beta t^{2+\beta} f(Dt/l^2). \tag{5}$$

Here f is a dimensionless function of the indicated argument. The argument is the only dimensionless variable one can construct from the parameters of the diffusion problem, and we have assumed that f can only depend on the diffusional variable, not the flow variables.

In the long time limit, $Dt \gg l^2$, we are supposing that $\langle\delta x^2\rangle \sim t$. For this to be true, along with Eq. (5), it must be the case that $f(z) \sim z^\nu$ for large z with $\nu = -1-\beta$. Consequently

$$\langle\delta x^2\rangle \sim u_0^2 l^{2(1+\beta)} t/D. \tag{6}$$

It is of interest to express this in terms of the maximum flow velocity, $v_{x,m} = u_0 l^\beta$,

$$\langle\delta x^2\rangle \sim v_{x,m}^2 l^2 t/D. \tag{7}$$

Note that the dispersion is independent of β when expressed in terms of the maximum velocity. The linear dependence on t in Eqs (6) and (7) has been assumed here, not derived. What has been derived is the dependence of the coefficient of t on the physical parameters of the system, the physical size, l, the diffusion coefficient, D, and the velocity v.

The preceding argument has assumed that the diffusion in the transverse dimension is normal. We now carry through the analogous calculation for the case when the transverse diffusion is anomalous, $\langle \delta y^2 \rangle = Bt^\alpha$. Arguments like those of Ben-Naim *et al.* yield, for the case when the y direction is unbounded

$$\langle \delta x^2 \rangle \sim u_0^2 B^\beta t^{2+\alpha\beta} \,. \tag{8}$$

For the bounded case, we assume $\langle \delta x^2 \rangle \sim u_0^2 B^\beta t^{2+\alpha\beta} \phi(Bt^\alpha / l^2)$ and that this must be proportional to t for large t. This can only happen if $\phi(z) \sim z^\mu$ for $z \gg 1$. This implies that $\mu = -(\beta + 1/\alpha)$. Consequently

$$\langle \delta x^2 \rangle \sim u_0^2 l^{2(\beta+1/\alpha)} t / B^{1/\alpha} = v_{x,m}^2 l^{2/\alpha} t / B^{1/\alpha} \,. \tag{9}$$

So here, while the t dependence is normal (by hypothesis), the size dependence is anomalous.

Of course, scaling arguments of the sort used here will never yield the numerical coefficient of the proportionality. One must go to a full solution of the problem for this, and incidentally also to justify the assumption that the x dispersion in the bounded case, the Taylor dispersion, is indeed proportional to t. This is the subject of the next section.

3. The convective diffusion equation

The basic model is that the fluid flows in the x direction with a velocity $v_x(r)$ where r is the radial variable normal to x. We take as our basic evolution equation the convective diffusion equation (4), and want to derive from this an equation for $p(x,t)$ alone. We regard this as a problem in the elimination of fast variables. To identify r as the fast variable, we write Eq. (4) in dimensionless form. Set $\rho = r/l$, $\zeta = x/L$, $\tau = v_{x,m} t/L$, $\varepsilon^{-1} = Kl^{-d_w} L / v_{x,m}$, $p(\rho, \zeta, \tau) = P(r, x, t)$, where l is the transverse dimension of the system, L is a characteristic length in the longitudinal direction, which is determined by the experimental conditions, and $v_{x,m}$ is the maximum flow velocity. One could, of course, reduce z by the length l instead of L. However, we feel it is more physical to scale a longitudinal variable by a longitudinal characteristic length, even if this length is arbitrary. We are no longer assuming that the flow velocity is a simple power law and consequently write $v_x = v_{x,m} g(r)$. Then the convective diffusion equation becomes

$$\frac{\partial p}{\partial \tau} + g(\rho) \frac{\partial p}{\partial \zeta} = \varepsilon^{-1} \frac{1}{\rho^{d-1}} \frac{\partial}{\partial \rho} \left(\rho^{d+1-d_w} \frac{\partial p}{\partial \rho} \right) . \tag{10}$$

We shall regard ε as a small parameter; that is, we are in the low velocity limit. L is, of course, arbitrary; it should be of the order of the width of the solute distribution, perhaps a few centimeters. According to Taylor [16], ε should be at most about 0.3 for the validity of his approximation. If we regard Kl^{2-d_w} as the analog of the diffusion coefficient in the nonfractal case, we can then treat ε as small (even though 0.3 is not so very small). According to the general principles of multiple time scale analysis [17] radial motion can be identified as the fast variable, and axial motion as the slow variable. This idea is implicit in the original paper of Taylor [1].

The method for elimination of fast variables which we use is essentially the Chapman–Enskog method. Its application to the problem of Taylor dispersion has been discussed in two earlier publications [2, 18], so that we can proceed directly to the case at hand. The density of solute, averaged over a cross section of the system is

$$n(\zeta, t) = V \int_0^1 p(\rho, \zeta, \tau) \rho^{d-1} d\rho \,, \tag{11}$$

where V is the factor coming from the angular integration in d dimensions. From (11) and (10), one finds

$$\frac{\partial n}{\partial \tau} = -V v_{x,m} \frac{\partial}{\partial \zeta} \int_0^1 g(\rho) p \rho^{d-1} d\rho \,. \tag{12}$$

Now we make the crucial hypothesis that p depends on time only through its functional dependence on n. This is supposed to hold after some induction period during which transients due to initial conditions die out. This hypothesis is the same as the familiar hypothesis in the kinetic theory of gases [19]. Application yields

$$\int \frac{\delta p}{\delta n(y)} \frac{\partial n(y, \tau)}{\partial \tau} d^d y = \varepsilon^{-1} \frac{1}{\rho^{d-1}} \frac{\partial}{\partial \rho} \rho^{d+1-d_w} \frac{\partial p}{\partial \rho} - g(\rho) \frac{\partial p}{\partial \zeta} \,, \tag{13}$$

where δ denotes functional derivative. Now we assume that p can be expanded in the form

$$p = p^0 + \varepsilon p^1 + \varepsilon^2 p^2 + \cdots \tag{14}$$

This is not necessarily a power series. The p^i may depend on ε also, but not in ways that affect their orders of magnitude; *e.g.* they may contain ε in an exponent. Nevertheless, we equate terms in (13) with the same apparent power of ε to zero. Furthermore, we require that

$$\begin{aligned} V \int p^0 \rho^{d-1} d\rho &= n \,, \\ V \int p^j \rho^{d-1} d\rho &= 0; j > 0 \,. \end{aligned} \tag{15}$$

To order ε^{-1}

$$\frac{1}{\rho^{d-1}}\frac{\partial}{\partial\rho}\,\rho^{d+1-d_w}\,\frac{\partial p^0}{\partial\rho}=0\,, \tag{16}$$

with solution

$$p^0=(a\rho^{d_w-d}+b)h(\zeta,\tau)\,, \tag{17}$$

where a and b are constants, and h is an arbitrary function. d_w is generally greater than d, so that p^0 is finite at the origin. However, there will be an unphysical cusp at the origin unless a vanishes (recall that p is a smoothed distribution). Application of (15) then determines h as

$$p^0=\frac{d}{V}n\,(\zeta,\tau)\,. \tag{18}$$

Next we look at terms of order ε^0. These are

$$\frac{\partial p^0}{\partial\tau}+g(\rho)\frac{\partial p^0}{\partial\zeta}=\frac{1}{\rho^{d-1}}\frac{\partial}{\partial\rho}\left(\rho^{d+1-d_w}\frac{\partial p^1}{\partial\rho}\right)\,. \tag{19}$$

Using (18) for p^0 and (12) for $\partial n/\partial\tau$, one finds

$$\frac{\partial}{\partial\rho}\left(\rho^{d+1-d_w}\frac{\partial p^1}{\partial\rho}\right)=\frac{d}{V}\frac{\partial n}{\partial\zeta}\alpha(\rho)\,, \tag{20}$$

where

$$\alpha(\rho)=\rho^{d-1}\left(g(\rho)-d\int_0^1 g(\rho)\rho^{d-1}d\rho\right)\,. \tag{21}$$

This equation can easily be solved for $p^1(\rho)-p^1(0)$. $p^1(0)$ is determined by (15) with the final result

$$\begin{aligned}p^1 \;=\; & \frac{d}{V}\frac{\partial n}{\partial\zeta}\left(\int_0^\rho x^{d_w-1-d}dx\int_0^x \alpha(y)dy\right.\\ & \left.-d\int_0^1 \rho^{d-1}d\rho\int_0^\rho x^{d_w-1-d}dx\int_0^x \alpha(y)dy\right)\,.\end{aligned} \tag{22}$$

Finally, this expression must be substituted in (12).

If we now return to ordinary, instead of dimensionless, coordinates, our final result to order ε is

$$\frac{\partial n}{\partial t}+\bar{v}\frac{\partial n}{\partial x}=D_{\text{eff}}\frac{\partial^2 n}{\partial x^2}\,, \tag{23}$$

where $\bar{v}$ is the cross sectional averaged flow velocity, and

$$D_{\text{eff}} = \frac{v_{x,m}^2 l^{d_w}}{K} Z \,, \tag{24}$$

$$\begin{aligned} Z \;=\; & d \int_0^1 \rho^{d-1} g(\rho) \left(d \int_0^1 \rho^{d-1} d\rho \int_0^\rho x^{d_w-1-d} dx \int_0^x \alpha(y) dy \right. \\ & \left. - \int_0^\rho x^{d_w-1-d} dx \int_0^x \alpha(y) dy \right) d\rho \,. \end{aligned} \tag{25}$$

This is consistent with the scaling arguments of the previous section, except that here we have explicitly shown the proportionality to t, and we have a definite expression for the proportionality coefficient, Z.

4. Discussion

Equations (24) and (25) are the main formal results of this paper. To evaluate Z and hence D_{eff} in any given case is now merely a question of quadratures. As examples, we have taken the case of a cylindrical container. For a parabolic velocity profile, $g(r) = 1 - (r/l)^2$, and for $d = d_w = 2$, we are back to Taylor's original problem, and obtain the original answer $D_{\text{eff}} = v_{x,m}^2 l^2 / 192 D_{\text{molecular}}$ if we equate $D_{\text{molecular}}$ with $K l^{2-d_w}$. The numerical coefficient is precisely that of Taylor [1]. For the same model of a cylindrical tube with parabolic velocity profile, carrying out the quadrature (25) is elementary, and yields

$$\begin{aligned} Z \;=\; & \frac{d}{d+2} \left[\frac{d}{d+2} \left(\frac{1}{d_w (d+d_w)} - \frac{1}{(d_w+2)(d+d_w+2)} \right) \right. \\ & \left. - \frac{1}{d_w (d+d_w+2)} + \frac{1}{(d_w+2)(d+d_w+4)} \right] . \end{aligned} \tag{26}$$

Although this formula is not very enlightening, the point to be made is that it can easily be computed. The interesting aspect of the result is the dependence on physical parameters, as shown in (24).

A parabolic velocity profile, used in the example above, is appropriate for Poiseuille flow in a tube of ordinary liquid. When the tube cross section is fractal, the velocity distribution will be different, and will probably depend on the pressure distribution at the inlet end of the tube. Nevertheless, once the velocity distribution is known, Z can be computed by simple quadratures. The parabolic profile was chosen only for the sake of example.

Regarding the choice of the evolution equation in the (r, x) space with which we started, we have discussed our reasons for this choice in Section 2. If, however, these reasons should be found not compelling, it is my belief that any evolution equation equivalent to an equation of continuity ($\partial P/\partial t = -$divergence(probability current)) will give rise to an effective diffusion equation in the x space when the fast, r, variable is eliminated. This will, I conjecture, be the case when the current does not depend on the history of the system, *i.e.* when the description is markovian.

In the situation considered here, classical diffusion on a fractal substrate, the dispersion (in the absence of convection) is subdiffusional, *i.e.* $d_w > 2$. We conjecture that similar behavior will occur when the dispersion in the absence of convection is superdiffusional, $d_w < 2$. This would be the case, for example, if the diffusion were based on a Levy walk instead of a Pearson walk. To verify whether this conjecture is correct, one would have to know the diffusion equation for Levy walks.

An interesting topic for future research is, what happens if the velocity distribution is a random variable? Matheron and Marsily [20] have studied this question in the case of normal diffusion with superposed convection for infinite systems; they were interested in applications where the time was short relative to the time necessary for the boundaries to be important. These authors found an anomalous longitudinal dispersion, as did Mazo and Van den Broeck [21] for a finite system with periodic boundary conditions. We conjecture that the longitudinal dispersion for a finite system with random velocities will again be linear in t, but a very interesting part of the problem will be to redo the Matheron–Marsily calculation for the case of anomalous diffusion in the transverse direction.

In this entire area of study, it is noteworthy how the presence of boundaries completely changes the character of the longitudinal dispersion. The reason is physically clear. Without boundaries, the transverse dispersion grows without bound. This indefinite growth, when fed back into the longitudinal motion by the convection gives rise the an anomalous dispersion. On the other hand, when the system is transversely bounded there is an upper bound on the transverse dispersion, the transverse dimension of the system. So the amount of spread fed back into the longitudinal motion is limited. This is the fundamental mechanism behind Taylor dispersion, regardless of the mechanism and time dependence of the transverse motion.

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