Diffusive transport in spatially periodic hydrodynamic flows

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We calculate the effective diffusion coefficient in convective flows which are well described by one spatial mode. We use an expansion in the distance from onset and homogenization methods to obtain an explicit expression for the transport coefficient. We find that spatially periodic fluid flow enhances the molecular diffusion D by a term proportional to D^{-1}. This enhancement should be easy to observe in experiments, since D is a small number.

I. INTRODUCTION

Dispersion in fluids at rest is due to the Brownian motion of the molecules or particles. On the macroscopic level it is well described by the Fick-diffusion equation with a transport coefficient D, the molecular-diffusion coefficient. Diffusion, or dispersion, is obviously an important phenomenon in chemical reactions, mixing of fluids, and spreading of pollutants. It is therefore of fundamental and practical importance to understand how fluid flow affects dispersion. The fact that even laminar flow can considerably increase dispersion is far less well known than the enhancement due to turbulence. In 1953 Taylor[1] showed that the longitudinal dispersion in Poiseuille flow in a cylindrical tube is described by an effective diffusion coefficient D^*,

$$D^* = D + \frac{\bar{u}^2 R^2}{48D} .$$

Here $\bar{u}$ is the average flow velocity and R the radius of the tube. Since $D^* - D$ is proportional to the inverse of the molecular-diffusion coefficient, typically $D \sim 10^{-5}$ cm^2/s in liquids, the contribution from the fluid flow is by far the dominant effect.

Here we will investigate the effect of fluid flows with zero mean velocity, $\bar{u}=0$, on dispersion. This is the case if the fluid velocity is spatially periodic. Such situations arise frequently as a consequence of hydrodynamic instabilities, as for instance in the Rayleigh-Bénard system[2] and the circular Couette system.[3] Our study will thus address the effect of hydrodynamic instabilities on mass transport in fluids. We will specifically consider the case of the convective instability in the Rayleigh-Bénard system.

Convection plays a role in various natural phenomena. Cloud streets visualize convection cells in the atmosphere. Transport of salt and heat in the ocean are often influenced by convection cells arising from a double-diffusive instability.[4] Further applications can be found in astrophysics, convection cells in stars, and in chemical engineering. For instance, fast reactions will be very sensitive to any increase in the effective diffusion coefficient.

In this paper we calculate explicitly the effective diffusion coefficient for convective fluid motion, in which essentially only one mode is excited. This means that we consider situations close to the hydrodynamic instability. The problem of calculating effective transport coefficients in fluid flows is a difficult one. At present, there are some technical difficulties in treating flows farther from the onset of convection, where additional modes are excited. Even close to the onset of convective motion in the Rayleigh-Bénard system the calculation of the effective diffusion coefficient is not easy. Thus, we have found it convenient to look at this problem from different angles. To offset some of the drawbacks of any perturbation calculation, we have used two perturbative methods which have different limitations. The results from both calculations agree exactly with each other. This shows that their range of validity is actually larger than the limitations of each particular method suggest. Our main result is that flows with zero mean velocity also enhance dispersion and that again the contribution from the flow is proportional to D^{-1}.

The effect of periodic fluid flows with zero mean velocity was previously considered in a different context by Nadim, Cox, and Brenner[5] and by Moffat.[6] Nadim and co-workers study a two-dimensional, spatially periodic, vortexlike flow engendered by a square array of almost-touching parallel, infinitely long circular cylinders, each rotating steadily about its symmetry axis in a Newtonian fluid under the action of an external couple exerted upon it from outside of the system. They use generalized Taylor-dispersion theory[7,8] to calculate the effective diffusivity tensor of this flow. Using the first-order smoothing approximation or quasilinear approximation, Moffat derives an expression for the diffusivity tensor of a convected scalar field in a turbulent flow. He applies this result to a case of "frozen" turbulence, namely a space-periodic velocity field of the form considered in this paper. Our results agree with those of Moffat, who used a small Pèclet number expansion to calculate explicitly the effective diffusion coefficient. Our method is quite different, and not subject to the same limitations; it uses concepts originating from Brownian motion theory and concepts from the theory of homogenization.

The organization of this paper is as follows: In Sec. II we describe the motion of a particle in the fluid flow by a set of Langevin equations. We calculate the dispersion in the direction perpendicular to the convection roll axis via an expansion in the Rayleigh number. This method exploits the fact that the flow is close to the instability. In Sec. III we consider a periodic velocity field given by one Fourier mode and assume that the initial concentration of the particles in the flow varies on a scale much larger than the period. This allows us to use homogenization methods[9] to calculate the effective diffusion coefficient. In Sec. IV we summarize our results, compare them to the case of Taylor diffusion, and discuss the general structure of the enhanced diffusion.

II. EFFECTIVE DIFFUSION COEFFICIENT NEAR A HYDRODYNAMIC INSTABILITY

We consider a Rayleigh-Bènard system of infinite lateral extent and thickness d. Let z be the direction perpendicular to the plates, x the direction perpendicular to the roll axis. We nondimensionalize length and time in the standard way by measuring lengths in units of d and time in units of thermal diffusion time d^2/κ. Here κ is the thermal diffusivity. Writing the Rayleigh number for $R \gtrsim R_c$ as

$$R = R_c + R_2\epsilon^2 + O(\epsilon^3) , \tag{1}$$

we have for the velocity in the case of rolls[10]

$$\mathbf{U} = \epsilon \begin{bmatrix} -Aa^{-1}\pi\sin(ax)\cos(\pi z) \\ 0 \\ A\cos(ax)\sin(\pi z) \end{bmatrix} + O(\epsilon^3) = \epsilon \begin{bmatrix} u \\ 0 \\ v \end{bmatrix} + O(\epsilon^3) , \tag{2}$$

with $A^2 = 3\pi^2 R_2$ and $a = \pi/\sqrt{2}$.[11] Here we have chosen free-free boundary conditions for mathematical convenience. The motion of a (neutrally buoyant) particle is then given by the following set of Langevin equations:

$$\begin{aligned} \dot{x}(t) &= \epsilon u(x(t),z(t)) + \xi_x(t) , \\ \dot{y}(t) &= \xi_y(t) , \\ \dot{z}(t) &= \epsilon v(x(t),z(t)) + \xi_z(t) , \end{aligned} \tag{3}$$

with instantaneous reflection at $z=0$ and 1. Further,

$$\langle \xi_i(t)\xi_j(t') \rangle = 2D\delta_{ij}\delta(t-t') . \tag{4}$$

The effective diffusion coefficient D^*_{xx} in the x direction is given by

$$D^*_{xx} = \lim_{t\to\infty} \left[\frac{\langle [x(t) - \langle x(t) \rangle]^2 \rangle}{2t} \right] . \tag{5}$$

(For the remainder of this section, we will drop the subscript xx.) In order to evaluate this second moment, we need to solve the Fokker-Planck equation associated with (3)

$$\partial_t p(x,y,z,t) = -\epsilon[\partial_x u(x,z) + \partial_z v(x,z)]p(x,y,z,t) + D[\partial_{xx} + \partial_{yy} + \partial_{zz}]p(x,y,z,t) \tag{6}$$

with

$$p(x,y,z,0) = \delta(x)\delta(y)\delta(z-z_0) , \tag{7}$$

$$\partial_z p(x,y,z,t)\,|_{z=0,1} = 0 , \tag{8}$$

for the reflecting boundary at $z=0$ and 1,

$$p(x,y,z,t) \to 0 \quad \text{as } x \to \pm\infty \quad \text{or } y \to \pm\infty , \tag{9}$$

for the natural boundaries at $x = \pm\infty$ and $y = \pm\infty$. Clearly, we only need to find $p(x,z,t)$ which obeys

$$\partial_t p(x,z,t) = -\epsilon[\partial_x u(x,z) + \partial_z v(x,z)]p(x,z,t) + D[\partial_{xx} + \partial_{zz}]p(x,z,t) . \tag{10}$$

Since $u(x,z)$ is an odd function of x and since $p(x,z,0)$ is an even function of x, we have that $p(x,z,t)$ is even in x. This implies that

$$\langle x(t) \rangle = 0 . \tag{11}$$

Thus (5) is equivalent to

$$D^* = \lim_{t\to\infty} \left[\frac{\langle [x(t)]^2 \rangle}{2t} \right] , \tag{12}$$

or

$$\langle x(t)^2 \rangle = 2D^* t \quad \text{for large } t .$$

This implies that

$$D^* = \tfrac{1}{2}\langle [x(\dot{t})]^2 \rangle \quad \text{for } t \text{ large} . \tag{13}$$

From the Fokker-Planck equation (10), we obtain

$$\langle [x(\dot{t})]^2 \rangle = \int_{-\infty}^{\infty} dx \int_0^1 dz\, x^2[-\epsilon(\partial_x u + \partial_z v)p + D(\partial_{xx} + \partial_{zz})p] . \tag{14}$$

Using the expressions (2) for u and v and integrating by parts, we find that

$$\langle [x(\dot{t})]^2 \rangle = 2D + 2\epsilon \int_{-\infty}^{\infty} dx \int_0^1 dz\, x u(x,z) p(x,z,t) , \tag{15}$$

and thus

$$D^* = D + \epsilon \int_{-\infty}^{\infty} dx \int_0^1 dz\, x u(x,z) p(x,z,t)\,|_{t\to\infty} . \tag{16}$$

To calculate the contribution from the fluid flow to the dispersion, we seek a perturbative solution of the Fokker-Planck equation (10) near the instability,

$$p(x,z,t) = p^{(0)}(x,z,t) + \epsilon p^{(1)}(x,z,t) + O(\epsilon^2) . \tag{17}$$

Substituting this ansatz in (10), we find that to order ϵ^0,

$$\begin{aligned} &\partial_t p^{(0)}(x,z,t) = D(\partial_{xx} + \partial_{zz})p^{(0)}(x,z,t) , \\ &p^{(0)}(x,z,0) = \delta(x)\delta(z-z_0) , \\ &\partial_z p^{(0)}(x,z,t)\,|_{z=0,1} = 0 , \\ &p^{(0)}(x,z,t) \to 0 \quad \text{as } x \to \pm\infty . \end{aligned} \tag{18}$$

Clearly at this order the joint probability factorizes

$$p^0(x,z,t) = p^{(0)}(x,t)p^{(0)}(z,t) , \tag{19}$$

with

$$\partial_t p^{(0)}(x,t)=D\partial_{xx}p^{(0)}(x,t) \tag{20}$$

and

$$\partial_t p^{(0)}(z,t)=D\partial_{zz}p^{(0)}(z,t) . \tag{21}$$

The solution of (20) is

$$p^{(0)}(x,t)=\frac{1}{2\sqrt{D\pi t}}e^{-x^2/4Dt} . \tag{22}$$

Taking into account that $z=0$ and 1 are reflecting boundaries, we find that the solution of (21) is given by

$$p^{(0)}(z,t)=1+2\sum_{n=1}^{\infty} e^{-(n\pi)^2Dt}\cos(n\pi z_0)\cos(n\pi z) . \tag{23}$$

To order ϵ the Fokker-Planck equation (10) reads

$$\begin{aligned}\partial_t p^{(1)}(x,z,t)&=D(\partial_{xx}+\partial_{zz})p^{(1)}(x,z,t)-(\partial_x u+\partial_z v)p^{(0)}(x,z,t)\\ &=D(\partial_{xx}+\partial_{zz})p^{(1)}(x,z,t)+Aa^{-1}\pi\sin(ax)\cos(\pi z)\partial_x p^{(0)}(x,z,t)-A\cos(ax)\sin(\pi z)\partial_z p^{(0)}(x,z,t) .\end{aligned} \tag{24}$$

We substitute the expression for $p^{(0)}$, calculated above, and finally obtain

$$\begin{aligned}\partial_t p^{(1)}(x,z,t)=&D(\partial_{xx}+\partial_{zz})p^{(1)}(x,z,t)+Aa^{-1}\pi\sin(ax)\cos(\pi z)\\ &\times\left[1+2\left[\sum_{n=1}^{\infty} e^{-(n\pi)^2Dt}\cos(n\pi z_0)\cos(n\pi z)\right]\right]\partial_x\left[\frac{1}{2\sqrt{D\pi t}}e^{-x^2/4Dt}\right]\\ &+2A\cos(ax)\left[\frac{1}{2\sqrt{D\pi t}}e^{-x^2/4Dt}\right]\sin(\pi z)\left[\sum_{n=1}^{\infty}(n\pi)e^{-(n\pi)^2Dt}\cos(n\pi z_0)\sin(n\pi z)\right]\end{aligned} \tag{25}$$

with

$$p^{(1)}(x,z,0)=0 ,$$

$$\partial_z p^{(1)}(x,z,t)\,|_{z=0,1}=0 ,$$

$$p^{(1)}(x,z,t)\to 0 \quad \text{as } x\to\pm\infty .$$

Since we have reflection at $z=0$ and 1, we write

$$p^{(1)}(x,z,t)=p_0^{(1)}(x,t)+\sqrt{2}\sum_{m=1}^{\infty} p_m^{(1)}(x,t)\cos(m\pi z) . \tag{26}$$

Recall that we need to evaluate

$$\epsilon\int_{-\infty}^{\infty}dx\int_0^1 dz\, xu(x,z)p(x,z,t)\,|_{t\to\infty}=-\epsilon\int_{-\infty}^{\infty}dx\int_0^1 dz\, xAa^{-1}\pi\sin(ax)\cos(\pi z)p(x,z,t)\,|_{t\to\infty} .$$

Thus it is sufficient for our purpose to determine $p_1^{(1)}(x,t)$ explicitly in the long-term limit. Multiplying (25) by $\sqrt{2}\cos(\pi z)$ from the left and integrating over z, we obtain

$$\begin{aligned}\partial_t p_1^{(1)}(x,t)=&D\partial_{xx}p_1^{(1)}(x,t)-D\pi^2 p_1^{(1)}(x,t)\\ &+Aa^{-1}\pi\sin(ax)\partial_x\left[\frac{1}{2\sqrt{D\pi t}}e^{-x^2/4Dt}\right]\sqrt{2}[\tfrac{1}{2}+\tfrac{1}{2}e^{-4\pi^2Dt}\cos(2\pi z_0)]\\ &+A\cos(ax)\left[\frac{1}{2\sqrt{D\pi t}}e^{-x^2/4Dt}\right]\sqrt{2}\pi\cos(2\pi z_0)e^{-4\pi^2Dt} .\end{aligned} \tag{27}$$

Since we are interested in the long-term limit and since $[x(t),z(t)]$ is clearly metrically transitive, the choice of the initial condition will have no bearing on the final result. Thus for the sake of convenience we choose the initial value $z_0=\frac{1}{4}$ and find

$$\partial_t p_1^{(1)}(x,t)=D\partial_{xx}p_1^{(1)}(x,t)-D\pi^2 p_1^{(1)}(x,t)+A\frac{a^{-1}\pi}{\sqrt{2}}\sin(ax)\partial_x\left[\frac{1}{2\sqrt{D\pi t}}e^{-x^2/4Dt}\right] \tag{28}$$

with

$$p_1^{(1)}(x,0)=0 \quad\text{and}\quad p_1^{(1)}(x,t)\to 0 \quad\text{as } x\to\pm\infty .$$

We solve this equation via Laplace transform, restricting x to the positive half line,

$$s\hat{p}_1^{(1)}(x,s)=D\partial_{xx}\hat{p}_1^{(1)}(x,s)-D\pi^2 p_1^{(1)}(x,s)+A\frac{a^{-1}\pi}{\sqrt{2}}\sin(ax)\left[-\frac{1}{2D}e^{-\sqrt{s/D}x}\right] , \tag{29}$$

where

$$\hat{p}_1^{(1)}(x,s)=\int_0^\infty e^{-st}p_1^{(1)}(x,t)dt \ .$$

Since we are dealing with a situation close to the critical point, we have that $a=a_c=\pi/\sqrt{2}$ and (29) can be written as

$$\partial_{xx}\hat{p}_1^{(1)}(x,s)=D^{-1}(s+D\pi^2)\hat{p}_1^{(1)}(x,s)+(2D^2)^{-1}A\sin(ax)e^{-\sqrt{s/D}x},\quad x>0\ . \tag{30}$$

The solution of this homogeneous differential equation, which fulfills the condition $\hat{p}_1^{(1)}(x,s)\to 0$ as $x\to\infty$, is given by

$$\hat{p}_1^{(1)}(x,s)=\hat{C}(s)\exp\{-[D^{-1}(s+D\pi^2)]^{1/2}x\}-\frac{A[(a^2+\pi^2)\sin(ax)-2a\sqrt{s/D}\cos(ax)]e^{-\sqrt{s/D}x}}{8Da^2[s+D(a^2+\pi^2)^2/4a^2]}\ . \tag{31}$$

Here $\hat{C}(s)$ is an as yet undetermined function. Inverting the Laplace transform we find

$$\begin{aligned} p_1^{(1)}(x,t)=&\int_0^t d\tau\, C(t-\tau)e^{-D\pi^2\tau}\frac{x}{2(D\pi\tau^3)^{1/2}}e^{-x^2/4D\tau}\\ &-\frac{A}{8Da^2}\int_0^t d\tau\exp\left[-\frac{D(a^2+\pi^2)^2}{4a^2}(t-\tau)\right]\\ &\times\frac{1}{2(D\pi\tau^3)^{1/2}}e^{-x^2/4D\tau}[(a^2+\pi^2)x\sin(ax)-a\cos(ax)H_2(x/2\sqrt{D\tau})]\ . \end{aligned} \tag{32}$$

H_2 is the second order Hermite polynomial. This expression for $p_1^{(1)}(x,t)$ is valid for $x>0$. To obtain $p_1^{(1)}(x,t)$ for all values of x, we exploit the fact the $p(x,t)$ is an even function, i.e., $p_1^{(1)}(x,t)=p_1^{(1)}(-x,t)$. Thus we find

$$\begin{aligned} p_1^{(1)}(x,t)=&\int_0^t d\tau\, C(t-\tau)e^{-D\pi^2\tau}\frac{|x|}{2(D\pi\tau^3)^{1/2}}e^{-x^2/4D\tau}\\ &-\frac{A}{8Da^2}\int_0^t d\tau\exp\left[-\frac{D(a^2+\pi^2)^2}{4a^2}(t-\tau)\right]\\ &\times\frac{1}{2(D\pi\tau^3)^{1/2}}e^{-x^2/4D\tau}[(a^2+\pi^2)x\sin(ax)-a\cos(ax)H_2(x/2\sqrt{D\tau})]\ . \end{aligned} \tag{33}$$

Further, $p_1^{(1)}(x,t)$ has to be at least twice continuously differentiable. The continuity of $\partial_x p_1^{(1)}(x,t)|_{x=0}$ requires that $C(s)$ vanishes identically. Thus we find that the solution of the Fokker-Planck equation (10) is, to first order in ϵ,

$$\begin{aligned} p(x,z,t)=\cos(\pi z)\Bigg[&\frac{1}{\sqrt{D\pi t}}e^{-x^2/4Dt}\cos\frac{\pi}{4}e^{-D\pi^2 t}\\ &-\epsilon\frac{A}{8Da^2}\int_0^t d\tau\exp\left[-D\frac{(a^2+\pi^2)^2}{4a^2}(t-\tau)\right]\frac{1}{(2D\pi\tau^3)^{1/2}}e^{-x^2/4D\tau}\\ &\times[(a^2+\pi^2)x\sin(ax)-a\cos(ax)H_2(x/2\sqrt{D\tau})]\Bigg]+\text{terms orthogonal to }\cos(\pi z)\ . \end{aligned} \tag{34}$$

So, the effective diffusion coefficient is given by

$$\begin{aligned} D^*=D+\epsilon\int_{-\infty}^{\infty}dx\int_0^1 dz\, x[-Aa^{-1}\pi\sin(ax)\cos(\pi z)]\\ \times\Bigg[&\frac{1}{2\sqrt{D\pi t}}e^{-x^2/4Dt}2\cos(\pi/4)e^{-D\pi^2 t}\cos(\pi z)\\ &-\epsilon\frac{A}{8Da^2}\int_0^t d\tau\exp\left[-D\frac{(a^2+\pi^2)^2}{4a^2}(t-\tau)\right]\frac{1}{2(D\pi\tau^3)^{1/2}}e^{-x^2/4D\tau}\\ &\times[(a^2+\pi^2)x\sin(ax)-a\cos(ax)H_2(x/2\sqrt{D}\,\tau)]\sqrt{2}\cos(\pi z)\Bigg]+O(\epsilon^3)\ . \end{aligned} \tag{35}$$

which needs to be evaluated in the long-time limit. Writing $D^*=D+\epsilon D_1+\epsilon^2 D_2+O(\epsilon^3)$, we find that

$$D_1=-A(D\pi t)\cos(\pi/4)e^{-D(a^2+\pi^2)t}\to 0 \quad \text{as } t\to\infty \tag{36}$$

and

$$D_2=\frac{A^2}{2}\int_0^t d\tau\, e^{-9D\pi^2(t-\tau)/8}\left[\tfrac{3}{4}-e^{-2D\pi^2\tau}\left(\tfrac{11}{4}-7D\pi^2\tau\right)\right]$$
$$\to\frac{1}{3}\frac{A^2}{D\pi^2} \quad \text{as } t\to\infty\ . \tag{37}$$

So, our final result is

$$D^*=D+\frac{1}{3}\frac{\epsilon^2A^2}{D\pi^2}+O(\epsilon^3)\ . \tag{38}$$

Note that ϵ^2A^2 is proportional to $\langle(\epsilon u)^2\rangle$, where $\langle\ \rangle$ denotes integration over the period cell. Thus (38) can be written as

$$D^*-D\propto\frac{\langle(\epsilon u)^2\rangle}{D}+O(\epsilon^3)=\frac{\langle U_x^2\rangle}{D}+O(\epsilon^3)\ . \tag{39}$$

On the other hand, using $A^2=3\pi^2R_2$ we obtain from (38)

$$D^*=D+\frac{\epsilon^2R_2}{D}+O(\epsilon^3)\ .$$

Close to onset we may write this, neglecting higher-order terms, as

$$D^*=D+(R-R_c)/D\ .$$

Using a typical value of $D=10^{-5}$ cm^2/s, we find that in the nondimensionalized units used here, $D=10^{-2}$. Thus, convective flow, even close to onset, can appreciably enhance dispersion ($R_c=657.5$ for free-free boundary conditions),

$$D^*=10^{-2}+10^2R_c\frac{R-R_c}{R_c}$$
$$=10^{-2}+6.575\times10^4\frac{R-R_c}{R_c}\ . \tag{40}$$

A result completely similar to (38) is obtained for the case of rigid-rigid boundaries.[12]

III. EFFECTIVE DIFFUSION COEFFICIENT IN PERIODIC FLOWS: THE HOMOGENIZATION METHOD

In Sec. II we have obtained an explicit expression for the enhancement of dispersion by convective flows. We have used a perturbation expansion in the distance from the critical point to approach this problem. In this section we will attack the problem from a different viewpoint. This will also furnish an indication as to the range of validity of expression (39). Let $\varphi(t,\mathbf{r})$ be the concentration of a passive (neutrally buoyant) contaminant. Then its temporal evolution is governed by the convection-diffusion equation

$$\partial_t\varphi(t,\mathbf{r})+\nabla\cdot[\mathbf{u}(\mathbf{r})\varphi(t,\mathbf{r})]=D\nabla^2\varphi(t,\mathbf{r}),\quad \mathbf{r}\in\mathbf{R}^3\ , \tag{41}$$

with initial condition

$$\varphi(0,\mathbf{r})=\varphi_0(\mathbf{r})\ . \tag{42}$$

(In this section we do not use the nondimensionalization of Sec. II.) Here $\mathbf{u}(\mathbf{r})$ is an arbitrary periodic velocity field which is divergence free,

$$\nabla\cdot\mathbf{u}(\mathbf{r})=0 \tag{43}$$

and has mean value zero,

$$\langle\mathbf{u}(\mathbf{r})\rangle=\mathbf{0}\ . \tag{44}$$

Recall that $\langle\ \rangle$ denotes integration over the period cell. We will consider the case that the initial condition varies on a length scale l_m that is large compared to the length scale l_f of the velocity field,

$$\varphi(0,\mathbf{r})=\varphi_0(\eta\mathbf{r})\ , \tag{45}$$

where η is the ratio of the two scales, $\eta=l_f/l_m$. It is exactly for problems of this type that homogenization methods were developed.[9] This particular problem was treated by McLaughlin *et al.*[9(c)] We briefly summarize their procedure here. Changing space and time scales by letting $\mathbf{r}\to\mathbf{r}/\eta$ and $t\to t/\eta^2$, we transform (41) into

$$\partial_t\varphi^\eta+\frac{1}{\eta}\nabla[\mathbf{u}(\mathbf{r}/\eta)\varphi^\eta]=D\nabla^2\varphi^\eta\ , \tag{46}$$

where $\varphi^\eta(t,\mathbf{r})=\varphi(t/\eta^2,\mathbf{r}/\eta)$ and

$$\varphi^\eta(0,\mathbf{r})=\varphi_0(\mathbf{r})\ .$$

We make the following ansatz for φ^η:

$$\varphi^\eta(t,\mathbf{r})=\bar\varphi(t,\mathbf{r})+\eta\varphi^{(1)}(t,\mathbf{r},\mathbf{r}/\eta)+\eta^2\varphi^{(2)}(t,\mathbf{r},\mathbf{r}/\eta)+\cdots\ . \tag{47}$$

In the spirit of a multiple-scale method, we treat $\mathbf{r}$ and $\boldsymbol{\rho}=\mathbf{r}/\eta$ as independent variables. We have thus $\nabla=\nabla_\mathbf{r}+\eta^{-1}\nabla_\rho$. We insert (47) into (46) and collect powers of η. To order η^{-2} we obtain

$$\mathbf{u}(\boldsymbol{\rho})\nabla_\rho\bar\varphi=\nabla_\rho^2\bar\varphi\ , \tag{48}$$

which is trivially satisfied, since $\bar\varphi$ is independent of the small scale $\boldsymbol{\rho}$. The coefficients of η^{-1} lead to the equation

$$D\nabla_\rho^2\varphi^{(1)}-\mathbf{u}(\boldsymbol{\rho})\cdot\nabla_\rho\varphi^{(1)}-\mathbf{u}(\boldsymbol{\rho})\cdot\nabla_\mathbf{r}\bar\varphi=0\ . \tag{49}$$

We denote the unit vectors in the x, y, and z direction by $\mathbf{e}_k$, $k=1,2,3$. Define functions $\chi_k(\boldsymbol{\rho})$ by

$$D\nabla_\rho^2\chi_k(\boldsymbol{\rho})-\mathbf{u}(\boldsymbol{\rho})\cdot\nabla_\rho\chi_k(\boldsymbol{\rho})-\mathbf{u}(\boldsymbol{\rho})\cdot\mathbf{e}_k=0\ . \tag{50}$$

Then it is easily verified that

$$\varphi^{(1)}(t,\mathbf{r},\boldsymbol{\rho})=\sum_{k=1}^{3}\chi_k(\boldsymbol{\rho})\frac{\partial\bar\varphi(t,\mathbf{r})}{\partial_{x_k}}\ , \tag{51}$$

with $\mathbf{r}=(x_1,x_2,x_3)$ and $\boldsymbol{\rho}=(x,y,z)=1/\eta(x_1,x_2,x_3)$ is a solution of (49). To order η^0 we obtain

$$D\nabla_\rho^2\varphi^{(2)}-\mathbf{u}\cdot\nabla_\rho\varphi^{(2)}-\mathbf{u}\cdot\nabla_\mathbf{r}\varphi^{(1)}$$
$$+2D\nabla_\mathbf{r}\cdot\nabla_\rho\varphi^{(1)}+D\nabla_\mathbf{r}^2\bar\varphi-\partial_t\bar\varphi=0\ . \tag{52}$$

We are looking for a solution of this equation which for each t and $\mathbf{r}$ fixed is periodic in $\boldsymbol{\rho}$, since $\mathbf{u}(\rho)$ is periodic. For periodic solutions

$$\langle \nabla_\rho^2 \varphi^{(2)} \rangle = 0$$

and

$$\langle \mathbf{u}\cdot\nabla_\rho \varphi^{(2)} \rangle = \langle \nabla\cdot(\mathbf{u}\varphi^{(2)}) \rangle = 0 \; . \tag{53}$$

The first equality in (53) follows from (43). Thus the solution of (52) can be periodic only if

$$-\langle \mathbf{u}\cdot\nabla_{\mathbf{r}}\varphi^{(1)}\rangle + 2D\langle \nabla_{\mathbf{r}}\cdot\nabla_\rho \varphi^{(1)}\rangle + D\langle \nabla_{\mathbf{r}}^2\bar{\varphi}\rangle - \langle \partial_t \bar{\varphi}\rangle = 0 \; . \tag{54}$$

Now $\langle \nabla_{\mathbf{r}}\cdot\nabla_\rho\varphi^{(1)}\rangle = \nabla_{\mathbf{r}}\cdot\langle \nabla_\rho\varphi^{(1)}\rangle = 0$, where the last equality follows from the periodicity of $\varphi^{(1)}$. So, the solvability condition (54) reads

$$\partial_t\bar{\varphi} = D\nabla_{\mathbf{r}}^2\bar{\varphi} - \langle \mathbf{u}\cdot\nabla_{\mathbf{r}}\varphi^{(1)}\rangle \; . \tag{55}$$

Inserting (51) into (55) we finally obtain

$$\partial_t\bar{\varphi} = \sum_{i,j=1}^{3} (D\delta_{ij} + D_{ij}^F)\frac{\partial^2}{\partial x_i\,\partial x_j}\bar{\varphi} \; , \tag{56}$$

where

$$D_{ij}^F = -\langle u_i \chi_j \rangle \; . \tag{57}$$

This shows that the average concentration $\bar{\varphi}$ obeys indeed a diffusion equation and that the enhancement of the dispersion due to the periodic flow is given by $-\langle u_i\chi_j\rangle$.

It is in general a formidable task to explicitly solve (50) to obtain the χ_k and thus to calculate D^F. Note, however, that we need to know "only" a "moment," namely $\langle u_i\chi_j\rangle$. Thus it is sufficient to solve (50) in the weak sense: Let V be a closed function space of periodic functions, such that $u_i \in V$. Then χ_k is a weak solution of (50) if

$$\langle fD\nabla_\rho^2\chi_k\rangle - \langle f\mathbf{u}\cdot\nabla_\rho\chi_k\rangle - \langle f\mathbf{u}\cdot\mathbf{e}_k\rangle = 0 \tag{58}$$

for an arbitrary element f of V. Now, if $\mathbf{u}\cdot\mathbf{e}_k$ is periodic and $\langle \mathbf{u}\cdot\mathbf{e}_K\rangle = 0$, then (58) has a periodic solution and uniqueness is guaranteed if $\langle \chi_k\rangle = 0$.[13]

To apply these results to convective flows let us consider velocity fields of the form

$$\mathbf{u} = \begin{bmatrix} -\tilde{A}\dfrac{\pi}{d}\sin\left[a\dfrac{x}{d}\right]\cos\left[\pi\dfrac{z}{d}\right] \\ 0 \\ \tilde{A}\dfrac{a}{d}\cos\left[a\dfrac{x}{d}\right]\sin\left[\pi\dfrac{z}{d}\right] \end{bmatrix} , \tag{59}$$

where $x \in (-\infty,\infty)$ and $z \in (-\infty,\infty)$. As in Sec. II we consider only convective flows that are well described by one spatial mode.

If we want to apply these considerations to the Rayleigh-Bénard system, then the velocity field (59) corresponds to an infinite stack of such systems. The application of homogenization methods to periodic flows in finite or semiinfinite geometries requires this mathematical trick. It is somewhat awkward to take boundary conditions into account using homogenization techniques. It is for this reason that (41) was formulated on an infinite domain. It will turn out, however, that the dispersions in the three coordinate directions are independent. Thus the result for D_{xx}^F is not affected by the above mathematical trick and can be applied directly to the Rayleigh-Bénard system.

For convective flows of the form (59), the solution of (58) is given by

$$\chi_i = -\frac{1}{D}\frac{d^2}{a^2+\pi^2}u_i \; . \tag{60}$$

To prove this, note that $D\nabla^2\chi_i = u_i$. So, $\langle fD\nabla^2\chi_i\rangle = \langle fu_i\rangle$ for arbitrary f. Thus, the above assertion is true if $\langle f\mathbf{u}\cdot\nabla\chi_i\rangle = 0$. Let V be the function space whose elements are

$$f = \sum_{m,n=1}^{\infty}\left[Q_{m,n}\sin\left[ma\frac{x}{d}\right]\cos\left[n\pi\frac{z}{d}\right] + P_{m,n}\cos\left[ma\frac{x}{d}\right]\sin\left[n\pi\frac{z}{d}\right]\right] . \tag{61}$$

Clearly u_1, u_2, and u_3 are elements of V. We have

$$\begin{aligned}\mathbf{u}\cdot\nabla u_1 &= (u_1\partial_x + u_3\partial_z)u_1 \\ &= \tilde{A}^2\frac{a\pi^2}{d^3}\sin\left[a\frac{x}{d}\right]\cos\left[\pi\frac{z}{d}\right]\cos\left[a\frac{x}{d}\right]\cos\left[\pi\frac{z}{d}\right] + \tilde{A}^2\frac{a\pi^2}{d^3}\cos\left[a\frac{x}{d}\right]\sin\left[\pi\frac{z}{d}\right]\sin\left[a\frac{x}{d}\right]\sin\left[\pi\frac{z}{d}\right] \\ &= \tilde{A}^2\frac{a\pi^2}{d^3}\sin\left[a\frac{x}{d}\right]\cos\left[a\frac{x}{d}\right]\end{aligned} \tag{62}$$

and

$$\begin{aligned}\mathbf{u}\cdot\nabla u_3 &= (u_1\partial_x + u_3\partial_z)u_3 \\ &= \tilde{A}^2\frac{a^2\pi}{d^3}\sin\left[a\frac{x}{d}\right]\cos\left[\pi\frac{z}{d}\right]\sin\left[a\frac{x}{d}\right]\sin\left[\pi\frac{z}{d}\right] + \tilde{A}^2\frac{a^2\pi}{d^3}\cos\left[a\frac{x}{d}\right]\sin\left[\pi\frac{z}{d}\right]\cos\left[a\frac{x}{d}\right]\cos\left[\pi\frac{z}{d}\right] \\ &= \tilde{A}^2\frac{a^2\pi}{d^3}\sin\left[\pi\frac{z}{d}\right]\cos\left[\pi\frac{z}{d}\right] .\end{aligned} \tag{63}$$

To evaluate $\langle f\mathbf{u}\cdot\nabla\chi_i\rangle$ we have to calculate

$$\left\langle \sin\left[ma\frac{x}{d}\right]\cos\left[n\pi\frac{z}{d}\right]\sin\left[a\frac{x}{d}\right]\cos\left[a\frac{x}{d}\right]\right\rangle ,$$

$$\left\langle \cos\left[ma\frac{x}{d}\right]\sin\left[n\pi\frac{z}{d}\right]\sin\left[a\frac{x}{d}\right]\cos\left[a\frac{x}{d}\right]\right\rangle ,$$

$$\left\langle \sin\left[ma\frac{x}{d}\right]\cos\left[n\pi\frac{z}{d}\right]\sin\left[\pi\frac{z}{d}\right]\cos\left[\pi\frac{z}{d}\right]\right\rangle ,$$

and

$$\left\langle \cos\left[ma\frac{x}{d}\right]\sin\left[n\pi\frac{z}{d}\right]\sin\left[\pi\frac{z}{d}\right]\cos\left[\pi\frac{z}{d}\right]\right\rangle .$$

Clearly all expectations vanish which implies that indeed

$$\langle f\mathbf{u}\cdot\nabla\chi_i\rangle=0 .$$

Since $\langle\chi_i\rangle$ vanishes, the solution (60) is unique and can be used to calculate D_{ij}^F. We find

$$D_{xx}^F=-\left\langle u_1\left[-\frac{1}{D}\frac{d^2}{a^2+\pi^2}\right]u_1\right\rangle=\frac{\langle u_1^2\rangle d^2}{D(a^2+\pi^2)} , \tag{64}$$

$$D_{yy}^F=0 , \tag{65}$$

$$D_{zz}^F=-\left\langle u_3\left[-\frac{1}{D}\frac{d^2}{a^2+\pi^2}\right]u_3\right\rangle=\frac{\langle u_3^2\rangle d^2}{D(a^2+\pi^2)} . \tag{66}$$

The off-diagonal terms of the diffusion matrix D^F all vanish,

$$D_{xy}^F=D_{yz}^F=0, \text{ since } u_2=0 \tag{67}$$

$$D_{xz}^F=-\left\langle u_1\left[-\frac{1}{D}\frac{d^2}{a^2+\pi^2}\right]u_3\right\rangle$$

$$=0, \text{ since } \langle\sin\xi\cos\xi\rangle=0 . \tag{68}$$

The final result for the effective diffusion coefficient in the x direction is

$$D^*=D+\frac{\langle u_1^2\rangle d^2}{D(a^2+\pi^2)} . \tag{69}$$

Note that D^* is dependent on the wavelength of the convective flow. Now $\langle u_1^2\rangle=\frac{1}{4}(\tilde{A}^2\pi^2/d^2)$ and if $a=a_c=\pi/\sqrt{2}$, we find

$$D^*=D+\frac{\tilde{A}^2}{6D} .$$

In order to compare this expression with (38) in Sec. II we need to nondimensionalize length and time. In these units, the relation between A in (2) and $\tilde{A}$ in (59) is

$$\tilde{A}=\epsilon A a^{-1}=\epsilon A\left[\frac{\pi}{\sqrt{2}}\right]^{-1} ,$$

so that

$$D^*=D+\frac{\epsilon^2A^2}{3D\pi^2} ,$$

which is exactly the result obtained in Sec. II.

IV. CONCLUSIONS

Using two different techniques, we have calculated the effective diffusion coefficient for simple convective flows. We find that

$$D_{ii}^*=D+\frac{\langle u_i^2\rangle d^2}{D(a^2+\pi^2)} \tag{70}$$

for spatially periodic flows which are well described by one spatial mode. Note that the limit of vanishing molecular diffusion is a singular limit and (70) is not applicable in this limit. This is very evident in both approaches. In the treatment of Sec. II we need to take a long-time limit in order to calculate D^*. As is clear from, for instance, (36), this limit is attained only for times such that $t=O(1/D)$, i.e., after the dispersing material has been spread over a roll by molecular diffusion, or, in other words, after the particles have sampled the velocity field transverse to the direction of dispersion considered. Thus, as the molecular diffusion becomes smaller and smaller, the time scale on which the effective diffusive behavior is established becomes longer and longer. Replacing the nondimensionalized quantities of Sec. II by the original ones, we obtain from this consideration the following condition for the observation of effective diffusive behavior: If at any time the dispersing material is spread over a region of dimension L in the x_i direction, convection will have given rise to effective diffusive behavior if

$$L^2\gg\left[D+\frac{\langle u_i^2\rangle d^2}{D(a^2+\pi^2)}\right]\frac{d^2}{D} ,$$

or, with $U\equiv(\langle u_i^2\rangle)^{1/2}$,

$$\frac{L}{d}\gg\frac{Ud}{D}=\mathcal{P} , \tag{71}$$

where $\mathcal{P}$ is the Pèclet number of the flow. As the molecular-diffusion coefficient D goes to zero or the Pèclet number to infinity, the spatial scale, on which effective diffusive behavior is observed, diverges. The inequality (71) establishes a relation between the macroscopic diffusive scale and the Pèclet number; condition (71) is exactly the same as the one derived by Taylor for enhanced diffusion in Poiseuille flow [see (16) of Ref. 1]. If the molecular diffusion is strictly zero, there can be no dispersion in a space-periodic flow with zero mean velocity, so $D^*=0$. This follows indeed quite easily from (5) and (10). In the homogenization method used in Sec. III the limit of vanishing molecular diffusion is also a singular limit; D enters the equation for χ_k, (50), multiplying the highest-order derivative. Again, we obtain that $D^*=0$ if the molecular diffusion is zero. Indeed, $\chi_x(\tau)=x$ and $\chi_z(\tau)=z$ for $D=0$, and thus $D_{ij}^F=-\langle u_i x_j\rangle=0$.

Our result (70) shows that spatially periodic flows enhance dispersion, though their average velocity is zero.

In fact, we find that the excess diffusion has exactly the same form as in the case of Poiseuille flow and linear shear flows, which have a nonzero average velocity. We are thus led to conjecture that it is a general feature of laminar flows that dispersion is enhanced in proportion to the square of a characteristic velocity and in inverse proportion to the molecular diffusion. This form of the excess diffusion can be understood heuristically in the following way: The smaller the molecular diffusion, the longer each particle will follow a given streamline of the laminar flow. If there exists a velocity gradient, then the different velocities between the streamlines will give rise to a dispersion of the particles, which will be larger the longer each particle stays near a given streamline. This is the situation for small molecular diffusion. If the molecular diffusion is large, then each particle will randomly sample many streamlines, i.e., many different velocities. Thus, for large D a velocity gradient will have only a negligible effect on dispersion; the velocity gradient is masked by the large random motion of the particles. We therefore expect the excess diffusion to be proportional to an inverse power of D, the simplest case being D^{-1}. It then follows simply from dimensional arguments that the excess diffusion is also proportional to the square of a characteristic velocity times a characteristic length.

Since molecular diffusion in liquids is small, typically $D = 10^{-5}$ cm^2/s, the flow-induced dispersion dominates molecular diffusion even for small amplitudes of the convective motion. Thus, experimental tests of our theoretical predictions should be relatively easy. Recall, however, that our theoretical calculations are carried out for two-dimensional flows and appropriate care has to be taken in experiments to avoid any significant deviation from this approximation.

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[1]G. I. Taylor, Proc. R. Soc. London A **219**, 186 (1953).

[2]For a recent review, see, e.g., F. H. Busse in *Hydrodynamic Instabilities and the Transition to Turbulence,* edited by H. L. Swinney and J. P. Gollub (Springer, Berlin, 1981).

[3]For a recent review, see, e.g., R. C. Di Prima and H. L. Swinney, in *Hydrodynamic Instabilities and the Transition to Turbulence,* edited by H. L. Swinney and J. P. Gollub (Springer, Berlin, 1981).

[4]See, e.g., H. E. Huppert and D. R. Moore, J. Fluid Mech. **78**, 821 (1976).

[5]A. Nadim, R. G. Cox, and H. Brenner, J. Fluid Mech. **164**, 185 (1986).

[6]H. K. Moffat, Rep. Prog. Phys. **46**, 621 (1983).

[7]H. Brenner, Philos. Trans. R. Soc. London A **297**, 81 (1980).

[8]H. Brenner and P. M. Adler, Philos. Trans. R. Soc. London A **307**, 149 (1982).

[9](a) A. Bensoussan, J. L. Lions, and G. Papanicolaou, *Asymptotic Analysis for Periodic Structures* (North-Holland, Amsterdam, 1978); (b) G. Papanicolaou and O. Pironneau, in *Stochastic Nonlinear Systems,* edited by L. Arnold and R. Lefever (Springer, Berlin, 1981); (c) D. W. McLaughlin, G. C. Papanicolaou, and O. R. Pironneau, SIAM J. Appl. Math. **45**, 780 (1985).

[10]A. Schlüter, D. Lortz, and F. Busse, J. Fluid Mech. **23**, 129 (1965).

[11]A. C. Newell and J. A. Whitehead, J. Fluid Mech. **38**, 279 (1969).

[12]P. McCarty (unpublished).

[13]G. C. Papanicolaou and S. R. S. Varadhan, in *Random Fields,* Vol. 27 of *Colloquid Mathematica Societatis Janos Bolyai 27,* edited by J. Fritz, J. L. Lebowitz, and D. Szasz (North-Holland, Amsterdam, 1981).