Unified theory of relative turbulent diffusion

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Anomalous relative diffusion (pair separation) in the inertial, as well as in the viscous, subranges of fully developed turbulent flow is considered. Using the Lagrangian velocity-correlation function in lowest-order continued-fraction approximation based on the Navier-Stokes equations and a phenomenological closure assumption, we express the variance in terms of the static structure function. This closed form for the turbulent diffusivity unifies scaling concepts with fluid mechanics and is free of fitting parameters. We obtain the following conclusions: Various regimes of variance versus time t are identified, $\propto t^2$ initially, t^1 and exponential (viscous subrange), $t^{3+\gamma}$ (inertial subrange). Intermittency (well known to alter the scaling exponent) implies a fairly strong effect upon the magnitude of the diffusion depending on the initial separation and on the Reynolds number; diffusion is delayed with increasing intermittency. Incompressibility is expected to lead to differences in transverse versus parallel separation. Molecular diffusivity may show up even in the universal regime as a permanent enhancement of diffusion due to different incubation times.

I. INTRODUCTION

The understanding of the way in which gaseous and liquid wastes disperse in the atmosphere, oceans, lakes, and rivers may be achieved by studying turbulent diffusion.[1] The subject of turbulent diffusion is thus of great practical importance. On the other hand, this is a topic of great theoretical interest; turbulent diffusion is an "anomalous" diffusion process[2,3] which is affected by the fractal nature of turbulence;[4] its understanding is related to the very description of the dynamics and statistics of the turbulent fluid itself.[5] In addition it turns out that turbulent diffusion is in part responsible for the generation of self-similar structures like clouds, which seem to be fractals in their own right.[6,7] Consequently turbulent diffusion and its ramifications relate to a number of topics of current theoretical interest.

Theoretically, turbulent diffusion is best understood in the context of fully developed isotropic turbulence.[5] Even here problems abound. Single-particle diffusion is dominated by large eddies which have no universal properties, causing the diffusive process itself to be nonuniversal. The situation is better for two particles or "relative" turbulent diffusion. Here one is interested in interparticle distances. These are not affected by large eddies which convect pairs of particles together. They are not affected by very small eddies either, since these are poor in energy. Thus interparticle distances are mostly affected by eddies of sizes comparable to them. If the interparticle distance is within the inertial range, one can expect to find universal behavior.

Consider two particles which are released initially at points $\vec{r}_1$ and $\vec{r}_2$, respectively. Their interparticle distance $\vec{R}$, $\vec{R}\equiv\vec{r}_1-\vec{r}_2$, will change in time due to the fact that the velocities at position $\vec{r}_1$ and $\vec{r}_2$ are not the same. Denoting the relative velocity by $\vec{V}(t)$ we have

$$\vec{R}(t)=\vec{R}(0)+\int_0^t\vec{V}(\tau)d\tau\ . \tag{1.1}$$

In isotropic turbulence $\langle\vec{V}(t)\rangle=\vec{0}$. Consequently $\langle\vec{R}(t)\rangle=\langle\vec{R}(0)\rangle$. The variance, however, is changing, leading to the turbulent diffusivity[1–4]

$$\frac{d\langle R^2\rangle}{d\tau}=2\int_0^t\langle\vec{V}(t)\cdot\vec{V}(\tau)\rangle d\tau\ . \tag{1.2}$$

We see that in order to understand turbulent diffusion we have to estimate time-correlation functions of velocity differences across a length scale R. Recently, such estimates have been attempted.[4] The essence of the argument has been as follows: The correlation $\langle\vec{V}(t)\cdot\vec{V}(\tau)\rangle$ is known to be nonstationary. We can assert, however, that there exists a function of scaled time variables $g(x)$ such that

$$\langle\vec{V}(t)\cdot\vec{V}(\tau)\rangle=\langle\vec{V}(t)\cdot\vec{V}(t)\rangle g((t-\tau)/t_R)\ , \tag{1.3}$$

where t_R is the typical decay time of velocity differences across a length scale R. Substitution in Eq. (1.2) leads to the asymptotic predictions

$$\frac{d\langle R^2\rangle}{dt}\sim\begin{cases}\langle\vec{V}(t)\cdot\vec{V}(t)\rangle t, & t\ll t_R & (1.4a)\\ \langle\vec{V}(t)\cdot\vec{V}(t)\rangle t_R, & t\gg t_R\ . & (1.4b)\end{cases}$$

The lack of knowledge of $g(x)$ prohibits estimates of the turbulent diffusivities at all times. At this point one has to estimate $\langle\vec{V}(t)\cdot\vec{V}(t)\rangle$ and t_R. As long as R is in the inertial range, the estimate of $\langle\vec{V}(t)\cdot\vec{V}(t)\rangle$ is relatively easy. Within the "homogeneous fractal model" of turbulence[4,8,9] or the "log-normal model"[5] one finds

$$\langle \vec{V}(t)\cdot\vec{V}(t)\rangle \sim \bar{\epsilon}^{2/3}R^{2/3}(R/l_0)^{\mu/3} \text{ (fractal model)}, \quad (1.5a)$$

$$\langle \vec{V}(t)\cdot\vec{V}(t)\rangle \sim \bar{\epsilon}^{2/3}R^{2/3}(R/l_0)^{\mu/9} \text{ (log-normal model)}, \quad (1.5b)$$

where $\bar{\epsilon}$ is the mean energy dissipation per unit mass and unit time and μ is the intermittency exponent. l_0 is the outer length scale at which energy is injected. The calculation of t_R is, however, more ambiguous, since it can be estimated in a variety of ways. Taking, for example, the log-normal statistics, we can write[5] (with ϵ_R being the dissipation within a ball of radius R)

$$t_R^{(1)} \sim \langle \epsilon_R \rangle^{-1/3} R^{2/3} \quad (1.6a)$$

or

$$t_R^{(2)} \sim \langle \epsilon_R{}^{-1/3} \rangle R^{2/3}, \quad (1.6b)$$

to take just two examples. Dimensionally, both forms are correct. The intermittency corrections, however, are entirely different, leading to

$$t_R^{(1)} \sim \bar{\epsilon}^{-1/3}R^{2/3}, \quad (1.7a)$$

$$t_R^{(2)} \sim \bar{\epsilon}^{-1/3}R^{2/3}\left[\frac{R}{l_0}\right]^{-2\mu/9}. \quad (1.7b)$$

A similar ambiguity arises in the fractal model. Here one can guess

$$t_R \sim \frac{R}{V_R}, \quad (1.8)$$

where V_R is the typical velocity difference across a length scale R. Other guesses are, however, possible.

The approach presented in Ref. 4 leads, with the choices (1.7a) and (1.8), to the estimates

$$\frac{d\langle R^2\rangle}{dt} \sim \begin{cases} \bar{\epsilon}^{1/3}R^{4/3}\left[\dfrac{R}{l_0}\right]^{\alpha_1}, & t \ll t_R \quad (1.9a)\\ \bar{\epsilon}^{1/3}R^{4/3}\left[\dfrac{R}{l_0}\right]^{\alpha_2}, & t \gg t_R \quad (1.9b)\end{cases}$$

with $R=\langle R^2\rangle^{1/2}(t)$, and α_1 and α_2 being $\mu/6$ and $2\mu/3$ or $\mu/18$ and $2\mu/9$ in the fractal and log-normal models, respectively. These results are difficult to implement because it is not certain at what times t in a typical experiment a switch from the regime $t \ll t_R$ to $t \gg t_R$ takes place. (Remember that t_R grows with time and it might even happen that for the duration of some experiments t is always smaller or always larger than t_R.) Thus, showing that the intermittency corrections are interesting and observable these estimates leave a number of problems open.

(1) Is there a unique way for estimating the relevant timescales t_R?

(2) Can one derive a theory of turbulent diffusion that is acceptable for all times and not only asymptotically at $t \ll t_R$ and $t \gg t_R$?

(3) Can one derive a theory that would hold at all length scales in the viscous as well as in the inertial ranges?

All these questions can be answered in the affirmative if we can relate theoretically the two-time-correlation functions appearing in Eq. (1.2) to the one-time-correlation function. Such a relation, however, calls for an analysis of the fluid mechanical equations of motion. Fortunately such an analysis has been presented recently in Ref. 10. The aim of this paper is to unify the approach presented in Ref. 10 to the issue of turbulent diffusion in a way that yields a closed theory of turbulent diffusion. Within the approximations adopted in Ref. 10 we obtain a theory that appears to hold at all length scales and time scales and is in fact free of any parameter that cannot be estimated either theoretically or from existing experiments.

The structure of the paper is as follows. In Sec. II we derive the exact relationship between turbulent diffusivity and Lagrangian time-correlation functions. Next we review the theory of Ref. 10 and deduce expressions for t_R and approximate forms for the turbulent diffusivity at all times and length scales. Section III turns to quantitative treatment of the turbulent diffusivity, taking intermittency effects into account. Equations of motion for the variance of $\vec{R}(t)$ are set and solved in the inertial as well as in the viscous ranges. Finally the effects of molecular diffusivity are assessed. Section IV offers a summary and discussion. The main conclusion of Sec. IV, as far as basic theory is concerned, is that one can study turbulent diffusion with scaled variance as a function of scaled time in a way which leads to universal plots in the absence of intermittency. Thus, any deviation from universality is attributable to intermittency. It turns out that even within the log-normal model (which tends to underestimate intermittency effects) the existence of intermittency leads to a large effect rather than the small corrections that one usually refers to. Thus the study of turbulent diffusion can shed important light on the basic structure of turbulence.

II. TURBULENT RELATIVE DIFFUSION IN TERMS OF LAGRANGIAN CORRELATION FUNCTIONS

A. Exact relations

Consider two particles which are initially ($t=t_0$) at positions $\vec{s}_0$ and $\vec{s}_0+\vec{r}$ in a field of locally isotropic, homogeneous turbulence. We denote the positions of these two particles at time $t=t_0+\tau$ by $\vec{x}(\tau;s_0,t_0)$ and $\vec{x}(\tau;\vec{s}_0+\vec{r},t_0)$, respectively. The quantity of basic interest for relative diffusion is the interparticle vector distance $\vec{R}(\vec{r},\tau;\vec{s}_0,t_0)$ which is defined by

$$\vec{R}(\vec{r},\tau;\vec{s}_0,t_0)=\vec{x}(\tau;\vec{s}_0+\vec{r},t_0)-\vec{x}(\tau;\vec{s}_0,t_0). \quad (2.1)$$

Clearly,

$$\vec{R}(\vec{r},0;\vec{s}_0,t_0)=\vec{r}. \quad (2.2)$$

For any $\tau>0$ we can write

$$\vec{R}(\vec{r},\tau;\vec{s}_0,t_0)=\vec{r}+\int_0^\tau d\tau'[\vec{u}(\tau';\vec{s}_0+\vec{r},t_0)-\vec{u}(\tau';\vec{s}_0,t_0)]. \quad (2.3)$$

Here $\vec{u}(\tau;\vec{s}_0,t_0)$ is the Lagrangian velocity,

$$\vec{u}(\tau;\vec{s}_0,t_0)=d_\tau\vec{x}(\tau;\vec{s}_0,t_0) , \quad (2.4)$$

where total derivatives with respect to τ are denoted by d_τ. The velocity difference appearing in the integrand of Eq. (2.3) will be denoted by $\vec{v}(\vec{r},\tau';\vec{s}_0,t_0)$. This velocity difference is a "Lagrangian eddy." Introducing the average $\langle \cdots \rangle$ as an average over the initial conditions $\vec{s}_0,t_0$, *with a fixed* $\vec{r}$, we find that

$$\langle \vec{R}(\vec{r},t)\rangle=\vec{r} \text{ for all } \tau , \quad (2.5)$$

provided that

$$\langle \vec{v}(\vec{r},\tau)\rangle=\vec{0} \text{ for all } \tau . \quad (2.6)$$

The latter equation is indeed obtained in homogeneous, stationary turbulence due to the translational invariance of $\langle \vec{u}(\vec{s}_0,t_0)\rangle$.

The one time variance is now introduced by defining

$$\sigma_{ij}(\vec{r},\tau)=\langle \delta R_i(\vec{r},\tau)\delta R_j(\vec{r},\tau)\rangle=\int_0^\tau d\tau_1\int_0^\tau d\tau_2\langle v_i(\vec{r},\tau_1;\vec{s}_0,t_0)v_j(\vec{r},\tau_2;\vec{s}_0,t_0)\rangle , \quad (2.7)$$

where $\delta\vec{R}(\vec{r},\tau)=\vec{R}(\vec{r},\tau)-\vec{r}$.

Isotropy of the turbulent field leads to the symmetry $\sigma_{ij}=\sigma_{ji}$. We also have by construction $\sigma_{ij}(r,0)=0$. Since (in locally isotropic turbulence) $\vec{r}$ is the only vector available for the construction of the variance tensor, we can immediately write

$$\sigma_{ij}(\vec{r},t)=\sigma_{\parallel}(r)\hat{r}_i\hat{r}_j+\sigma_\perp(r)P_{ij}(\hat{r}) , \quad (2.8)$$

where $\hat{r}=\vec{r}/r$ and $\overleftrightarrow{P}=\overleftrightarrow{1}-\hat{r}\hat{r}$.

The relative turbulent diffusivity is defined now as

$$d_\tau\sigma_{ij}(\vec{r},\tau)=2\int_0^\tau d\tau'\langle v_i(\vec{r},\tau;\vec{s}_0,t_0)v_j(\vec{r},\tau-\tau';\vec{s}_0,t_0)\rangle_{\text{sym}} . \quad (2.9)$$

It is interesting to realize that the diffusivity can be decomposed again to longitudinal and transverse parts. In principle, one can detect experimentally the two parts. We return to this point in Sec. IV. Usually one is, however, interested only in the trace

$$d_\tau\sigma(\vec{r},\tau)=2\int\langle \vec{v}(\vec{r},\tau)\cdot\vec{v}(\vec{r},\tau-\tau')\rangle d\tau'$$
$$=d_\tau\sigma_{\parallel}+2d_\tau\sigma_\perp . \quad (2.10)$$

Equations (2.9) and (2.10) bring us as far as we can go with the exact theory. We now relate the time-correlation function to static structure functions, and this step requires approximations.

B. Approximations based on fluid mechanics

1. The decay of Lagrangian eddies

The correlation decay of Lagrangian velocity differences has been considered in Ref. 10. Here we summarize the ideas which are relevant for the present study.

Using the Navier-Stokes equations and the definition of $\vec{v}(\vec{r},\tau;\vec{s}_0,t_0)$ one derives the Lagrangian equation of motion

$$d_\tau v_n(\vec{r},\tau;\vec{s}_0,t_0)=\nu\Delta_x v_n(\vec{r},\tau;\vec{s}_0,t_0)-\partial_{x_n}\Pi(\vec{r},\tau;\vec{s}_0,t_0) . \quad (2.11)$$

Here Π denotes the pressure difference

$$\Pi(\vec{r},\tau;\vec{s}_0,t_0)=p(\vec{x}(\tau;\vec{s}_0+\vec{r},t_0),t_0+\tau)$$
$$-p(\vec{x}(\tau;\vec{s}_0,t_0),t_0+\tau) . \quad (2.12)$$

The dynamic correlation function

$$D_{ij}(\vec{r},\tau)=\langle v_i(\vec{r},0,\vec{s}_0,t_0)v_j(\vec{r},\tau;\vec{s}_0,t_0)\rangle \quad (2.13)$$

is now written as an inner product in a space M defined by

$$M=[\delta A(\vec{r},\tau;\vec{s}_0,t_0) \mid \langle A^2\rangle<\infty] , \quad (2.14)$$

where

$$\delta A(\vec{r},\tau;\vec{s}_0,t_0)=A(\vec{x}(\tau;\vec{s}_0+\vec{r},t_0),t_0+\tau)$$
$$-A(\vec{x}(\tau;\vec{s}_0,t_0),t_0+\tau) \quad (2.15)$$

and $A(\vec{x},t)$ may be any sum or product of the three components of $\vec{u}(\vec{x},t)$ and of $p(\vec{x},t)$. The inner product is written as

$$D_{ij}(\vec{r},\tau)=(v_i(\vec{r})\mid e^{\tau L}v_j(\vec{r})) . \quad (2.16)$$

Here L is defined by

$$L\,\delta A(\vec{r};\vec{s}_0,t_0)=d_\tau\delta A(\vec{r},\tau;\vec{s}_0,t_0)\mid_{\tau=0} . \quad (2.17)$$

The static correlation function is the structure function $D_{ij}(\vec{r})=D_{ij}(\vec{r},\tau=0)$ which can also be decomposed into longitudinal and transverse parts

$$D_{ij}(\vec{r})=D_{\parallel}(r)\hat{r}_i\hat{r}_j+D_\perp(r)P_{ij}(\vec{r}) . \quad (2.18)$$

Incompressibility implies the relation $D_\perp=D_{\parallel}+rD_{\parallel}'/2$. Next we introduce the projector $\mathscr{P}$ on $\vec{v}(\vec{r})$:

$$\mathscr{P}(\vec{r};\vec{s}_0,t_0)=\mid v_i(\vec{r};\vec{s}_0,t_0)N_{ij}(\vec{r})v_j(\vec{r};\vec{s}_0,t_0)\mid . \quad (2.19)$$

The matrix N_{ij} is the inverse of the static correlation functions

$$N_{ij}=\frac{1}{D_\perp(r)}P_{ij}(\vec{r})+\frac{1}{D_{\parallel}(r)}\hat{r}_i\hat{r}_j . \quad (2.20)$$

With the help of this projector one employs the standard Zwanzig-Mori[11] projection-operator technique generalized to dissipative systems in Ref. 12 to derive the exact equations of motion (Laplace transformed) for the normalized dynamic correlation functions, decomposed into the longitudinal and transverse parts:

$$C_{\parallel,\perp}(r,z)=[z+\gamma_{\parallel,\perp}(r)+m_{\parallel,\perp}(r,z)]^{-1} . \quad (2.21)$$

The following definitions are introduced:

$$\overleftrightarrow{C}(\vec{r},z)=\overleftrightarrow{D}(\vec{r},z)\cdot\overleftrightarrow{N}(\vec{r}) \text{ (correlation matrix)} , \quad (2.22)$$

$$\overleftrightarrow{\gamma}(\vec{r})=-(\vec{v}(\vec{r})\mid L\vec{v}(\vec{r}))\cdot\overleftrightarrow{N}(\vec{r}) \text{ (damping rate)} , \quad (2.23)$$

and

$$\overleftrightarrow{m}(\vec{r},z)=\overleftrightarrow{M}(\vec{r},z)\cdot\overleftrightarrow{N}(\vec{r})\text{ (memory tensor)}\ . \quad (2.24)$$

The major point of this analysis is that the damping rate $\overleftrightarrow{\gamma}(\vec{r})$ can be calculated exactly. The result of this calculation is[10]

$$\gamma_{\perp}(r)D_{\perp}(r)=\tfrac{2}{3}\bar{\epsilon}-\frac{\nu}{2r}[r^2D_{\parallel}'''(r)+6rD_{\parallel}''(r)+4D_{\parallel}'(r)]\ , \quad (2.25)$$

$$\gamma_{\parallel}(r)D_{\parallel}(r)=\tfrac{2}{3}\bar{\epsilon}-\frac{\nu}{r}[rD_{\parallel}''(r)+4D_{\parallel}'(r)]\ . \quad (2.26)$$

Unfortunately, the memory part cannot be calculated to any degree of controlled approximation. For this reason it has been assumed in Ref. 10 that the bare relaxation time contains most of the essential physics. A similar assumption is made here. Once the memory is neglected, the damping rate of eddies of size r is calculable unambiguously in terms of $\bar{\epsilon}$ and the static structure functions. Thus, the ambiguity of the simple dimensional analysis is removed. Furthermore, the extension to all r ranges is possible.

2. *The turbulent diffusivity*

Guided by the above analysis we can see that the correlation function of Eq. (2.10) can be written as

$$\langle v_i(\vec{r},\tau)v_i(\vec{r},\tau-\tau')\rangle \simeq \langle v_i(\vec{r},\tau)v_i(\vec{r},\tau)\rangle \exp[-\gamma(\vec{r},\tau)\tau']\ . \quad (2.27)$$

At this point we introduce the approximations

$$\langle v_i(\vec{r},\tau)v_i(\vec{r},\tau)\rangle \simeq \langle v_i(r_\tau)v_i(r_\tau)\rangle = D(r_\tau)\ , \quad (2.28a)$$

where

$$r_\tau=[r^2+\sigma(r,\tau)]^{1/2}=[R_i(r,\tau)R_i(r,\tau)]^{1/2}\ . \quad (2.28b)$$

In words, this approximation means that we replace the correlation function of Lagrangian velocity differences, evolved during the time τ from an r eddy (but averaged over initial conditions), by the correlation function of velocity differences across the distance r_τ which is the actual rms extension of a cloud of particles released initially at vector distance $\vec{r}$. This approximation relates to closure ideas and was taken naturally within the scaling analysis of Ref. 4. Consistently with this approximation we take[10]

$$\gamma_{\parallel}(r,\tau)\simeq\frac{1}{D_{\parallel}(r_\tau)}\left[\tfrac{2}{3}\bar{\epsilon}-\frac{\nu}{r}[rD_{\parallel}''(r_\tau)+4D_{\parallel}'(r_\tau)]\right] \quad (2.29)$$

and similarly $\gamma_{\perp}(r,\tau)\simeq\gamma_{\perp}(r_\tau)$. Focusing on $\sigma_{\parallel}$ (with an analogous equation for $\sigma_{\perp}$) we can perform the integration of Eq. (2.9) and derive the equation

$$d_\tau\sigma_{\parallel}(r,\tau)=2D_{\parallel}(r_\tau)\frac{1-e^{-\gamma_{\parallel}(r_\tau)\tau}}{\gamma_{\parallel}(r_\tau)}\ . \quad (2.30)$$

Written differently,

$$d_\tau\sigma_{\parallel}(r,\tau)=2D_{\parallel}(r_\tau)t_{\parallel}(r_\tau)\{1-\exp[-\tau/t_{\parallel}(r_\tau)]\}\ , \quad (2.31)$$

where the eddy correlation decay time $t_{\parallel}(r_\tau)=\gamma_{\parallel}^{-1}(r,\tau)$ was introduced. Notice that in Eq. (2.31) there is still a coupling to $\sigma_{\perp}(r,\tau)$ through

$$r_\tau=[r^2+\sigma(r,\tau)]^{1/2}\ .$$

Thus the equations for $\sigma_{\parallel}$ and $\sigma_{\perp}$ have to be solved simultaneously. For simplicity we adopt here a decoupling approximation in which we take

$$r_\tau\simeq[r^2+\sigma_{\parallel}(r,\tau)]^{1/2}$$

in the longitudinal equation and

$$r_\tau\simeq[r^2+\sigma_{\perp}(r,\tau)]^{1/2}$$

in the transverse one. Equation (2.31) becomes then, together with Eqs. (2.28) and (2.29), a closed equation for the variance $\sigma_{\parallel}(r_\tau)$ for all τ and all initial separations r, provided the static structure function $D_{\parallel}$ is given for eddies of all sizes. Equation (2.31) together with the analogous expression for $\sigma_{\perp}$ are the basic result: a set of two coupled first-order differential equations (which in the simple approximation adopted henceforth are decoupled) for the variance of two-particle diffusion, released at $\tau=0$ a distance r apart.

In the limit $\tau \gg t_{\parallel}(r_\tau)$ and $\tau \ll t_{\parallel}(r_\tau)$ we recover the estimates of Eq. (1.4) (in which the difference between $\sigma_{\parallel}$ and $\sigma_{\perp}$ has not been taken into account).

Notice that the results obtained here pertain to the viscous and the inertial ranges simultaneously. The theory is now set for the estimates of the intermittency effects and for the quantitative calculation of the turbulent diffusivity.

III. QUANTITATIVE TREATMENT OF TURBULENT DIFFUSIVITY

The all important quantity is the structure function $D_{\parallel}(r_\tau)$. In order to proceed we have to know this quantity for all r, in particular both in the inertial and viscous subranges. We begin with the inertial subrange.

A. The inertial subrange

Naive dimensional analysis in the inertial range predicts

$$D_{\parallel}(r_\tau)\sim\bar{\epsilon}^{2/3}r_\tau^{2/3}\ . \quad (3.1)$$

Intermittency introduces dimensionless corrections. These can be assessed according to either model of intermittency—the fractal model and the log-normal model.

1. *Fractally homogeneous turbulence*

In this model one distinguishes between velocity differences across *active* regions which belong to the fractal, and inactive regions which do not belong.[8,9] The probability that a distance r_τ belongs to a fractal of dimension D which is embedded in space of dimension d scales like $(r_\tau/l_0)^{d-D}=(r_\tau/l_0)^{\mu}$. Accordingly one writes[4]

$$\langle \vec{v}(r_\tau)\cdot\vec{v}(r_\tau)\rangle \sim v_{r_\tau}^2 (r_\tau/l_0)^\mu \,, \tag{3.2}$$

where v_{r_τ} is the velocity difference in an active region. The latter is related to the dissipation $\bar\epsilon$ via

$$\bar\epsilon \sim (v_{r_\tau}^3/r_\tau)(r_\tau/l_0)^\mu \,, \tag{3.3}$$

where the factor $(r_\tau/l_0)^\mu$ appears for the same reason; if r_τ does not belong to the fractal there is no contribution to $\bar\epsilon$ from that region. Combining Eqs. (3.2) and (3.3) we get

$$D_{||}(r_\tau) \sim \bar\epsilon^{2/3} r_\tau^{2/3} (r_\tau/l_0)^{\mu/3} \,. \tag{3.4}$$

For reasonable values of μ, $0.25 \lesssim \mu \lesssim 0.5$, the use of Eq. (3.4) in (2.26) shows that for $r_\tau \gg l_d$ the contribution of the viscosity term to $t_{||}(r_\tau)$ can be neglected in the inertial subrange and we get

$$t_{||}(r_\tau) = \frac{3}{2}\frac{D_{||}(r_\tau)}{\bar\epsilon} \sim \bar\epsilon^{-1/3} r_\tau^{2/3} (r_\tau/l_0)^{\mu/3} \,. \tag{3.5}$$

2. *The log-normal model*

Within this model one writes the average of the pth power of dissipation in a ball of radius r as

$$\langle \epsilon_r^p \rangle = \langle e^{pY_r} \rangle \,, \tag{3.6}$$

where $Y_r \equiv \ln \epsilon_r$. Using the log-normality assumption one then derives

$$\langle \epsilon_r^p \rangle \sim \bar\epsilon^p \left[\frac{l_0}{r} \right]^{\mu p (p-1)/2} \,. \tag{3.7}$$

The structure function $D_{||}(r_\tau)$ can now be written as

$$D_{||}(r_\tau) \sim \langle v_{r_\tau}^2 \rangle \sim \langle \epsilon_{r_\tau}^{2/3} r_\tau^{2/3} \rangle \sim \bar\epsilon^{2/3} r_\tau^{2/3} (r_\tau/l_0)^{\mu/9} \,. \tag{3.8}$$

Similarly we get for $t_{||}(r_\tau)$,

$$t_{||}(r_\tau) = \frac{3}{2}\frac{D_{||}(r_\tau)}{\bar\epsilon} \sim \bar\epsilon^{-1/3} r_\tau^{2/3} (r_\tau/l_0)^{\mu/9} \,. \tag{3.9}$$

3. *Asymptotic results for the diffusivity*

For $\tau \ll t_{||}(r_\tau)$ and $\tau \gg t_{||}(r_\tau)$ we can now combine Eqs. (3.4) and (3.5), or (3.8) and (3.9) and with the master formula (2.31) to obtain

$$d_\tau \sigma_{||}(r,\tau) \sim \begin{cases} \bar\epsilon^{1/3} r_\tau^{4/3} (r_\tau/l_0)^{\alpha_s}, & \tau \ll t_{||}(r_\tau) \quad (3.10\text{a}) \\ \bar\epsilon^{1/3} r_\tau^{4/3} (r_\tau/l_0)^{\alpha_l}, & \tau \gg t_{||}(r_\tau) \quad (3.10\text{b}) \end{cases}$$

where in the first regime $\alpha_s = \mu/6$ and $\mu/18$ in the fractal and log-normal models, respectively, whereas in the second regime $\alpha_l = 2\mu/3$ and $2\mu/9$ in the two models. Notice that for the fractal model these results agree with Ref. 4 (and $\alpha_{1,2}$ in Sec. I). For the log-normal model α_l is two times the result of Ref. 4. The difference stems from the choice (1.7a) in Ref. 4. In the present theory the ambiguity of $t_{||}(r_\tau)$ is removed and therefore we believe that the present result is the correct one.

4. *The diffusivity for all times*

In the present theory we are not limited to asymptotic results. Using the basic Eq. (2.31), we can write

$$\frac{d\sigma_{||}(r,\tau)}{d\tau} = C\bar\epsilon^{1/3} r_\tau^{4/3} (r_\tau/l_0)^\alpha (1 - e^{-\tau/t_{||}(r_\tau)}) \tag{3.11}$$

with a similar equation for $\sigma_\perp(r,\tau)$. Here $\alpha = 2\mu/3$, $2\mu/9$ in the fractal and log-normal models, respectively. Equation (3.11), which is the central result of the present approach in the inertial range, is a completely defined first-order differential equation. We rewrite it in a form suitable for numerical integration:

$$\frac{d\sigma_{||}(r,\tau)}{d\tau} = C\bar\epsilon^{1/3} [r^2 + \sigma_{||}(r,\tau)]^{2/3} \left[\frac{r^2 + \sigma_{||}(r,\tau)}{l_0^2} \right]^{\alpha/2} \{1 - \exp[-\tau/t_{||}(r_\tau)]\} \tag{3.12}$$

with

$$t_{||}(r_\tau) = C\bar\epsilon^{-1/3} (r^2 + \sigma_{||})^{1/3} \left[\frac{r^2 + \sigma_{||}}{l_0^2} \right]^{\alpha/4} \,. \tag{3.13}$$

These equations determine the whole temporal development of the variance $\sigma(r,\tau)$ for a pair of particles released initially at a distance $\vec{r}$ apart at time $\tau = 0$. The remaining Reynolds number dependent parameters (all denoted by the same symbol C) appearing in these equations can be determined from independent measurements. As an example we take the measurements reported in Ref. 13 and which are summarized by the formulas (cf. Ref. 10)

$$D_{||}(r_\tau) = b_{||} \mathscr{R}_\lambda^{-\mu/6} (r_\tau/l_d)^{2/3+\mu/9} v_{l_d}^2 \,, \tag{3.14}$$

$$t_{||}(r_\tau) = \tfrac{3}{2} b_{||} \mathscr{R}_\lambda^{-\mu/6} (r_\tau/l_d)^{(6+\mu)/9} \tau_{l_d} \,. \tag{3.15}$$

The relevant parameters for this experiment are displayed in Table I. With these numbers we integrated Eq. (3.12) numerically and obtained the results summarized in Figs. 1 and 2.

Some of the appearances of the curves in Figs. 1 and 2 can be obtained directly from Eqs. (3.10) and (3.11). For small τ, when $\sigma \ll r^2$ we find from Eq. (3.11) a regime

TABLE I. Parameters needed for the numerical integration leading to Figs. 1—4.

Quantity	Numerical value according to Ref. 13
b	2.7
$\mathscr{R}_\lambda$	4300
μ	0.25
ν (viscosity)	0.143 cm^2/sec
$\bar\epsilon$	300 cm^2/sec^3 (estimated)
$l_d = (\nu^3/\bar\epsilon)^{1/4}$	0.06 cm
$v_{l_d} = (\nu\bar\epsilon)^{1/4}$	2.6 cm/sec
$t_{l_d} = l_d/v_{l_d}$	2.3×10^{-2} sec

where

$$\text{(i)}\ \ \sigma_{\|}(r,\tau)\sim\bar{\epsilon}^{2/3}r_\tau^{2/3}\left[\frac{r_\tau}{l_0}\right]^{\xi_{2/3}}\tau^2\ ,\quad \sigma\ll r^2,\ \tau\ \text{small}\ .$$

Here $\xi_{2/3}=\mu/3$ and $\mu/9$ in the fractal and log-normal models, respectively. For $\sigma\gg r^2$ we can integrate Eqs. (3.10) to obtain

$$\text{(ii)}\ \ \sigma_{\|}(r,\tau)\sim\begin{cases}\bar{\epsilon}\tau^3[\tau/t(l_0)]^{\gamma_s}, & \tau\ll t_{\|}(r_\tau)\\ \bar{\epsilon}\tau^3[\tau/t(l_0)]^{\gamma_l}, & \tau\gg t_{\|}(r_\tau)\end{cases}$$

where $\gamma_s=3\mu/(4-\mu)$ or $3\mu/(12-\mu)$ and $\gamma_l=3\mu/(1-\mu)$ and $\gamma_l=3\mu/(3-\mu)$ in the fractal and log-normal models, respectively. $t(l_0)=\bar{\epsilon}^{-1/3}l_0^{2/3}$ denotes the stirring time scale. Regime (i) can be clearly seen in Figs. 1 and 2. For the parameters chosen, the regime $\tau\gg t_{\|}(r_\tau)$ is never reached in the cases plotted here. Thus the asymptotic (large-τ) curves are in accord with the first equation of regime (ii). It should be stressed that in other cases a crossover between the two regimes (ii) might occur.

B. Diffusion in the viscous subrange

In the viscous subrange the structure function $D_{\|}(r_\tau)$ depends on the viscosity explicitly. For $r\ll l_d$ analyticity requires $D_{\|}(r)\sim r^2$. Standard arguments show that (cf. Ref. 10)

$$D_{\|}(r)\simeq\tfrac{1}{15}(r/l_d)^2v_{l_d}^2,\quad r\ll l_d \tag{3.16}$$

where v_{l_d} is the Kolmogorov velocity. In addition one has a constant limiting value $t_{\|}(r)=t_{\perp}(r)\equiv t(r)$:

$$t(r)\simeq C_0t_{l_d}\equiv t_0,\quad r\ll l_d \tag{3.17}$$

where C_0 depends on the Reynolds number only. Combining Eqs. (2.31), (3.16), and (3.17) we find in the viscous subrange

$$\frac{d\sigma_{\|}(r,\tau)}{d\tau}=2ar_\tau^2t_0(1-e^{-\tau/t_0})\ , \tag{3.18}$$

where $a=\frac{1}{15}(v_{l_d}/l_d)^2$. This equation is integrated to yield

$$\ln\left[1+\frac{\sigma_{\|}(r,\tau)}{r^2}\right]=2at_0[\tau+t_0(e^{-\tau/t_0}-1)]\ . \tag{3.19}$$

As before, we identify three stages in the evolution of the cloud.

(i) $\tau\to0$. As before, we find that initially $\sigma_{\|}(r,\tau)\sim\tau^2$.
(ii) $\tau\gg t_0$ but still $\sigma_{\|}\ll r^2$.

There is no guarantee that such a regime always exists. If it does, then

$$\sigma_{\|}(r,\tau)\simeq2at_0r^2\tau\ . \tag{3.20}$$

This is a normal diffusive behavior with a diffusion constant

$$\mathcal{D}=at_0r^2=\tfrac{1}{15}C_0r^2/t_{l_d}\ . \tag{3.21}$$

Using $t_{l_d}=l_d^2/\nu$ we can write the diffusion constant also as

$$\mathcal{D}=\frac{C_0(\mathcal{R}_\lambda)}{15}\nu\left[\frac{r}{l_d}\right]^2\ . \tag{3.22}$$

(iii) $\tau\gg t_0$, $\sigma_{\|}\gtrsim r^2$.

This process gives rise to an exponential growth

$$\sigma_{\|}(r,\tau)=r^2\exp(2at_0\tau)\ . \tag{3.23}$$

Accordingly it cannot last long before r_τ exceeds l_d. Once this happens, the inertial range behavior takes over, and the formulas of Sec. III A have to be used.

It is important to realize that for $r<l_d$ the diffusion constant (3.22) may be smaller than its molecular counter-

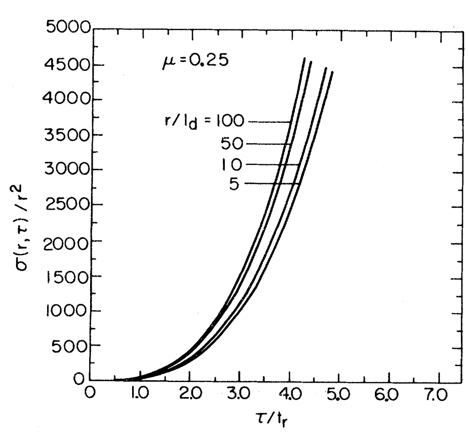

FIG. 1. Numerical solution of Eq. (3.12) with parameters as displayed in Table I, for four initial conditions: $r/l_d=5$, 10, 50, 100.

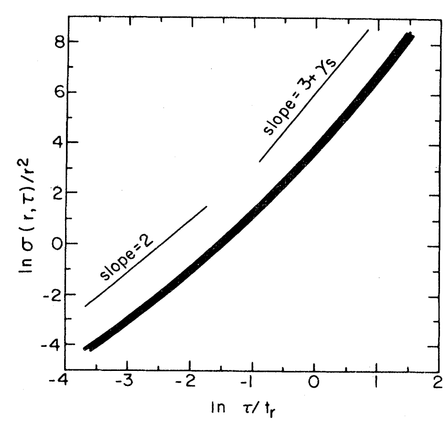

FIG. 2. A log-log plot of the data of Fig. 1. Here the asymptotic slopes of 2 and $3+\gamma_s$ are clearly seen.

part. Whether this is so or not depends also on the Reynolds number. At any rate there very well may be a competition between molecular and turbulent diffusivities as long as $r \lesssim l_d$. Consequently two materials of different molecular diffusivities χ might show markedly different turbulent diffusivities due to different incubation times that are needed to reach a cloud size r_τ for which the eddy diffusivity takes over, i.e., gets larger than χ. The quantitative aspects of this effect are discussed next.

C. The effects of molecular diffusivity

Without the turbulent activity all the contribution to the dispersion $\sigma(r,\tau)$ would have come from molecular diffusivity,

$$\frac{d\sigma(r,\tau)}{d\tau}=2\chi \ . \tag{3.24}$$

The simplest way of assessing the influence of molecular diffusivity is to assume that the molecular and turbulent diffusivities are uncorrelated. In this case we simply add 2χ to the right-hand side of Eq. (2.31). Since we expect the molecular diffusivity to be important in the viscous subrange but to become increasingly unimportant in the inertial range $r_\tau \gg l_d$, we want to integrate Eq. (2.31) such that $r_\tau \ll l_d$ initially but grows well into the inertial range finally. Therefore, the asymptotic forms for $D(r_\tau)$ and $t(r_\tau)$ are not sufficient. We need an expression for $D_{\|}(r_\tau)$ which holds for *all* r, including the crossover from the viscous to the inertial range. As an appropriate interpolation formula we use the following expression:[10]

$$D_{\|}(r_\tau)=\bar{a}(r_\tau/l_d)^2 v_{l_d}^2/[1+\bar{c}(r_\tau/l_d)^2]^q \ . \tag{3.25}$$

Here $\bar{a}=\frac{1}{15}$,

$$\bar{c}=(15b_{\|})^{-18/(12-\mu)}\mathscr{R}_\chi^{3\mu/(12-\mu)} \ ,$$

and $q=(12-\mu)/18$ in the log-normal model. A similar interpolation formula for the fractal model can be easily written down. We also have to interpolate $t(r_\tau)$ between Eqs. (3.9) and (3.17). This can be done by using Eq. (3.25) in Eq. (2.26) which expresses $t_{\|}(r)$ by $D_{\|}(r)$. For simplicity we used, however, the simpler interpolation

$$\hat{t}_{\|}(r_\tau)=t_0\left[1+\frac{1}{t_0}t_{\|}(r_\tau)\right] . \tag{3.26}$$

The results of the integration of Eq. (2.31) extended by $+2\chi$ on the right-hand side with these interpolation formulas are shown in Figs. 3 and 4. We have picked for χ the values 0.5×10^{-4}, 0.5×10^{-3}, 0.5×10^{-2}, 0.5×10^{-1} cm^2/sec, and the initial condition $r/l_d=10^{-2}$. We see that the effect of the molecular diffusivity is in determining how long it takes before the turbulent diffusivity sets in. As a result $\sigma(r,\tau)$ differs even at later times, if substances with different χ diffuse simultaneously. This "incubation time" can be understood as setting an effective initial time for the turbulent diffusion process, from which onward the molecular diffusivity can be neglected. Additional discussion of this effect and comparisons to experiments are discussed elsewhere.[14]

IV. DISCUSSION

One of the amusing results of the present study is the difference between $\sigma_{\|}$ and $\sigma_\perp$. Although we treat particle diffusion which occurs due to entrainment in chaotically moving fluid, the structure of the velocity field introduces infinitely long memory to the initial position difference $\vec{r}$ [cf. Eq. (2.1)]. The structure of the velocity field is represented by the different magnitude of $D_{\|}$ and $D_\perp$, a difference which reflects the different energies associated with longitudinal and transverse motion. Our theory predicts a transverse diffusivity larger than the longitudinal one. A jet of smoke injected to a turbulent medium is thus expected to become wider (in proportion to its length) as a function of time. It is appropriate to reiterate at this point that the equations for $\sigma_{\|}$ and $\sigma_\perp$ are coupled; we decoupled them for the sake of simplicity of analysis.

Within the stated approximations we achieved simple first-order differential equations for the variance of two-particle diffusion which hold equally well in the viscous and inertial subranges. Accordingly, these equations can be used in practical applications over a wide range of ex-

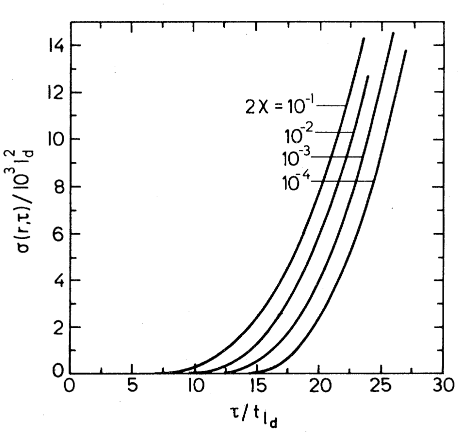

FIG. 3. The effect of molecular diffusivity. When the initial condition r is much smaller than l_d, molecular diffusivity wins over and determines the "incubation time" after which eddy diffusivity takes over.

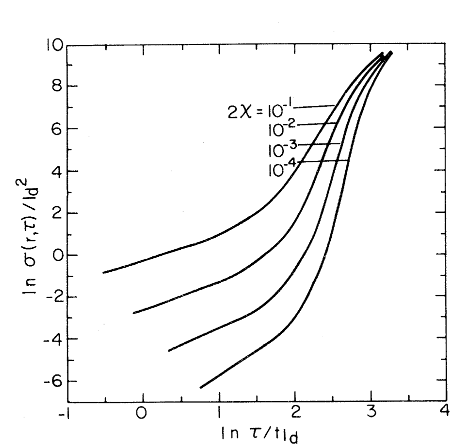

FIG. 4. A log-log plot of the data of Fig. 3. The asymptotic slopes considered in the text are easily seen.

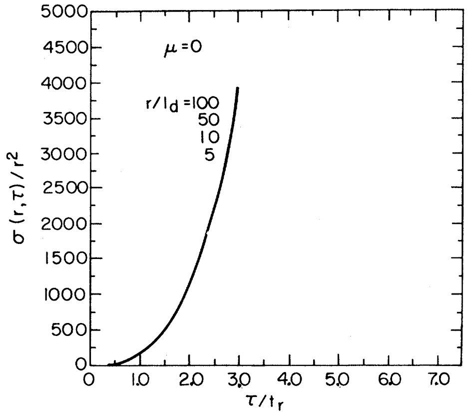

FIG. 5. The same as Fig. 1 but with $\mu=0$. It is $\bar{D}=88.6$. Relative diffusion shows no dependence on initial separation.

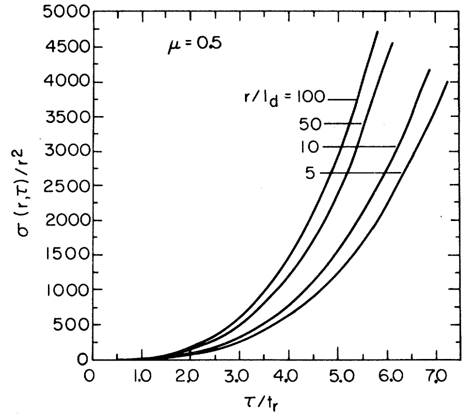

FIG. 6. The same as Figs. 1 and 5 but with $\mu=0.5$. Reynolds number $\mathscr{R}_\lambda=4300$. $\bar{D}$ is reduced to 14.3, 16.1, 21.0, 23.6. Notice that the magnitude of relative diffusion is reduced and delayed by intermittency, while the exponent of τ is increased by it. (Also seen in Fig. 1 with $\mu=0.25$, hence $\bar{D}=35.6$, 37.7, 43.1, 45.7).

perimental conditions. In addition to its practical importance, the analysis presented above should be useful for deepening our basic understanding of turbulence itself. As seen above, monitoring turbulent diffusion amounts to a measurement of the Lagrangian time-correlation functions. Done right, such measurements can be used to investigate intermittency *per se*. To stress this point we rewrite here the differential equation for $\sigma_{||}$ in a "quasi-universal" form that displays clearly the effects of $\mu\neq 0$ in the inertial range.

Start with Eq. (2.31). Use now scaled variables

$$\Gamma(r,t)=\sigma_{||}(r,\tau)/r^2 \tag{4.1}$$

and

$$t=\tau/t_{||}(r) \; . \tag{4.2}$$

Remembering Eqs. (3.14) and (3.15) we can derive straightforwardly the equation

$$\frac{d\Gamma}{dt}=\bar{D}(1+\Gamma)^{(6+\mu)/9}\left[1-\exp\left[-\frac{t}{(1+\Gamma)^{(6+\mu)/18}}\right]\right] \tag{4.3}$$

with

$$\bar{D}=\tfrac{9}{2}b_{||}^3\mathscr{R}_\lambda^{-\mu/2}(r/l_d)^{\mu/3} \; , \tag{4.4}$$

where the log-normal model has been used. An analogous equation with fractal statistics can be easily derived. The point is that now the dependence on initial conditions, i.e., r/l_d *and* on the Reynolds number appears only through $\bar{D}$, and *disappears for* $\mu=0$. Thus, in experiments the deviation of Γ from universality as a function of r and $\mathscr{R}_\lambda$ is only due to intermittency and seems therefore to be a particularly nice way of investigating intermittency, and the different models for it.

Figures 5 and 6 show a numerical solution of Eq. (4.3) for four values of r/l_d (5,10,50,100) where the only change is in the value of μ (0.5;0). Together with Fig. 1 which pertains to $\mu=0.25$, a dramatic effect is clearly seen. It thus seems tempting to suggest measurements of the intermittency exponent based on experiments of turbulent diffusivity as a function of initial conditions [i.e., changing r/l_d and monitoring $\Gamma(t)$].

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