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Generalized Langevin equation for relative turbulent dispersion

By BURKHARD M. O. HEPPE

Department of Applied Mathematics and Theoretical Physics, University of Cambridge, Silver Street, Cambridge CB3 9EW, UK

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The relative velocity of two fluid particles in homogeneous and stationary turbulence is considered. Looking for reduced dynamics of turbulent dispersion, we apply the nonlinear Mori–Zwanzig projector method to the Navier–Stokes equations. The projector method decomposes the Lagrangian acceleration into a conditionally averaged part and a random force. The result is an exact generalized Langevin equation for the Lagrangian velocity differences accounting for the exact equation of the Eulerian probability density. From the generalized Langevin equation, we obtain a stochastic model of relative dispersion by stochastic estimation of conditional averages and by assuming the random force to be Gaussian white noise. This new approach to dispersion modelling generalizes and unifies stochastic models based on the well-mixed condition and the moments approximation. Furthermore, we incorporate viscous effects in a systematic way. At a moderate Reynolds number, the model agrees qualitatively with direct numerical simulations showing highly non-Gaussian separation and velocity statistics for particle pairs initially close together. At very large Reynolds numbers, the mean-square separation obeys a Richardson law with coefficient of the order of 0.1.

1. Introduction

Turbulent dispersion of a contaminant, for example pollutant dispersion in the atmosphere, is conveniently described in terms of Lagrangian statistics sampled along the paths of fluid particles (Taylor 1921). In practice, however, the Eulerian statistics sampled at fixed points in space are far better known from experiments. Hence, the basic problem of turbulent dispersion is to calculate Lagrangian statistics from given Eulerian statistics. Lagrangian dispersion models address the problem by statistically characterizing particle paths from an Eulerian input. Only recently have Lagrangian statistics become immediately accessible through direct numerical simulations (DNS) of the Navier–Stokes equation, however with the well-known limitation to moderate Reynolds numbers.

In contrast, most Lagrangian dispersion models have been developed for large or infinite Reynolds numbers in accordance with Kolmogorov inertial-range theory. In order to compare modelled dispersion behaviour to DNS data, one therefore needs to incorporate finite Reynolds number effects and hence include the viscous range explicitly. In this work we propose a new approach to a certain class of Lagrangian dispersion models, so-called stochastic models. Starting from the Navier–Stokes equation, we describe relative dispersion in stationary, homogeneous, and locally isotropic turbulence on all scales including the small viscous scales.

1.1. *Stochastic models of turbulent dispersion*

Lagrangian stochastic models formulate the time evolution of the particle velocity in terms of stochastic differential equations. Analogous to the Langevin equation of Brownian motion, turbulent fluctuations are represented by a stochastic force. The models are best understood for the description of one-particle ensembles, which contain only one-point statistical information and hence lack any information about the different scales of turbulent eddies. Two alternative modelling approaches include Eulerian statistics in Lagrangian one-particle models: the well-mixed approach of Thomson (1987) and the moments approximation method of Kaplan & Dinar (1993) (only the well-mixed approach guarantees consistency, see Du, Wilson & Yee 1994).

One-particle stochastic models have also received a great deal of attention from the combustion community, since they provide simple closure models for the Eulerian (one-point) probability density function (p.d.f.). The transport equation of the Eulerian p.d.f. contains conditional averages which are non-closed and require modelling (Pope 1985). In Lagrangian p.d.f. methods, stochastic models for the fluid particle paths are used to determine both the Lagrangian and the Eulerian p.d.f.s (Pope 1994). The model coefficients are expressed in terms of low-order Eulerian moments according to certain consistency requirements. Whereas the exact but unclosed p.d.f. transport equations have been known for a long time, the corresponding exact Langevin equations in the one-particle case have only recently been formulated (Heinz 1997).

A modelled ensemble of single particles allows the calculation of mean concentrations, whereas an ensemble of particle pairs allows the calculation of concentration fluctuations. To this end, a one-dimensional two-particle stochastic model has been proposed by Durbin (1980). Three-dimensional models at infinite Reynolds and Péclet numbers have been considered in Thomson (1990) and Borgas & Sawford (1994) based on the well-mixed criterion.

A moments approximation method for relative dispersion at high Reynolds numbers has been investigated by Novikov and co-workers (Novikov 1989; Pedrizzetti & Novikov 1994). Their work can be regarded as a Lagrangian p.d.f. method: given a Lagrangian model one can investigate the corresponding Eulerian p.d.f. In the work of Pedrizzetti & Novikov (1994), this is the p.d.f. of spatial velocity increments which has received much attention by experimentalists. It is well established experimentally and numerically that the p.d.f. of velocity increments shows exponential tails on sub-integral scales corresponding to anomalous scaling of higher-order velocity structure functions. Anomalous scaling is regarded as an important signature of turbulent intermittency (Frisch 1995). Although Pedrizzetti & Novikov (1994) find their stochastic model to be compatible with exponential tails of the Eulerian p.d.f., it remains open whether the model reliably represents small-scale relative dispersion.

The infinite Reynolds number models of Thomson and Novikov are briefly discussed in § 3 and generalized in §§ 4 and 5 by means of an exact generalized Langevin equation for the relative dispersion of two fluid particles. The dispersion model of Durbin (1980) has been extended to include finite Reynolds and Péclet number effects, but lacks certain consistency requirements guaranteed by the well-mixed approach (Sawford & Hunt 1986). Recently, Borgas & Sawford (1996) investigated the effects of molecular diffusion and viscosity on the concentration statistics produced by decaying turbulence behind a grid. Their marked molecule approach, in which time histories of tracer molecules are modelled, rather than those of fluid particles, yields an improved description of the experimental data. However, the inclusion of viscous and diffusive effects is *ad hoc* and in order to satisfy the well-mixed condition they have to include

an unphysical drift term in the advection equation. In this paper we demonstrate that viscous effects cannot be taken into account by merely extending the energy spectrum or second-order structure function to include the viscous range (Sawford & Hunt 1986; Borgas & Sawford 1996).

1.2. *The projector approach to dispersion modelling*

We restrict ourselves to the relative motion of two fluid particles and look for a reduced statistical Lagrangian description of relative dispersion by constructing a stochastic model valid for arbitrary turbulent Reynolds numbers. We apply the nonlinear Mori–Zwanzig projection operator method (Grabert 1982) to the Navier–Stokes equations for an ensemble of particle pairs. As known from the theory of Brownian motion in non-equilibrium statistical mechanics, the deterministic equations of motion with random initial conditions can be rewritten in the form of exact generalized Langevin equations (Lindenberg & West 1990). The linear Mori–Zwanzig projector method has been introduced in the context of turbulent dispersion by Grossmann & Thomae (1982, henceforth referred to as GT82).

In an original attempt to relate the correlation at two different times of velocity differences between two fluid particles to Eulerian structure functions, GT82 apply the linear Mori–Zwanzig projector formalism directly to the Navier–Stokes equations. Their results have been used by Grossmann & Procaccia (1984) together with several assumptions to derive closed ordinary differential equations for the mean-square separation of two fluid particles. The nonlinearity due to the Lagrangian paths is taken into account only phenomenologically, but Grossmann & Procaccia (1984) not only describe the mean-square separation in the inertial range, but also find the correct asymptotic behaviour in the viscous subrange.

In this work, we consider the problem setting of GT82, but we make use of the nonlinear Mori–Zwanzig projector method in order to capture the inherent nonlinearities. The difference between the linear and the nonlinear theory lies in the choice of the projection operator P to be defined in § 4. In the linear case P projects onto the initial velocities. In the nonlinear case P is essentially the conditional average for fixed velocity increments. Rather than looking for equations for the mean separation, we formally derive exact generalized Langevin equations, which we then approximate by stochastic differential equations. This yields nonlinear stochastic models for the relative separation and velocity of two fluid particles, thus extending the models of Thomson and Novikov. Furthermore, the nonlinear theory readily allows the exact equation for the Eulerian p.d.f. of velocity increments to be recovered.

The projector method decomposes the time rate of change of the dynamical variables, which in this context are the components of relative acceleration, into a systematic drift and a random force representing turbulent fluctuations. The correlation of the random force is related to the drift by a fluctuation–dissipation theorem, which has to be fulfilled to guarantee consistency with the Eulerian statistics even if one introduces approximations (see the discussion in § 3). The nonlinear projector method formulates drift and random force as conditional averages which we approximate by lowest-order stochastic estimation. The coefficients of the Langevin equation are expressed by Eulerian velocity and pressure structure functions known from experiments and analytically represented by interpolation functions. The scale dependence on viscous, inertial and integral scales thus follows from the theory including the approximations. The amplitudes of the stochastic force in the three scaling ranges, however, are free model parameters and have to be determined by comparison to

numerical data. We note that we do not account for any intermittency corrections of the Eulerian velocity statistics as is usually done in stochastic dispersion studies (Borgas & Sawford 1994, but see the discussion in § 7). Assuming the random force to be Gaussian distributed and delta-correlated in time (white noise) finally yields a simple Markovian dispersion model.

The Markovian dispersion model is solved numerically for separation and velocity statistics. For the first time we compare the relative dispersion results of a stochastic model to DNS (Yeung 1994). The Markov model describes the mean-square separation reasonably well on all scales. The velocity statistics suggests that the viscous range can effectively be modelled by choosing the free parameter of the model, the amplitude of the stochastic force, to be larger in the viscous range than in the inertial range. The Richardson coefficient is found to be smaller than for the well-mixed models and of $O(0.1)$.

The probability densities of relative separation and longitudinal velocity difference show a strong deviation from a Gaussian distribution in the transition region from viscous to integral scales. The model captures the qualitative trends in the dependence on the initial separation, but underestimates peak values of skewness and flatness.

In the main body of this paper we consider the evolution of particle velocities only and we neglect intermittency effects. Furthermore, our Lagrangian modelling approach can also be applied to the time histories of concentration differences between two fluid particles. Whereas traditional two-particle models describe only the variance of a passive scalar field, our approach allows higher-order two-point statistics such as scalar structure functions of arbitrary order and Péclet number to be extracted. In Appendix B we demonstrate how this yields Kraichnan's statistical theory of a passive scalar advected by a Gaussian white-noise velocity field. Kraichnan (1994) showed that the mutual effect of random advection and molecular diffusion causes the scalar field to become intermittent. Scalar intermittency appears as anomalous scaling of scalar structure functions established for the highly simplified case that the advecting velocity field is non-intermittent and Gaussian.

This paper is organized as follows. In § 2 we formulate the statistical description in terms of Eulerian and Lagrangian variables. The stochastic models of Thomson and Novikov are introduced in § 3. After summarizing the results from the linear projector method (GT82) we derive the nonlinear generalized Langevin equation in § 4. Section 5 discusses the principal approximations of the different terms in the Langevin equation. The numerical simulation of the stochastic dispersion model is given in § 6 and compared to the DNS data of Yeung (1994). We discuss possible improvements of the dispersion model and conclude in § 7.

2. Eulerian versus Lagrangian variables

In the Eulerian representation the incompressible velocity field is described as a three-dimensional vector field $\boldsymbol{u}(\boldsymbol{x}, t)$ with $\partial_{x_i} u_i(\boldsymbol{x}, t) = 0$ at a fixed position $\boldsymbol{x}$ governed by the Navier–Stokes equation

$$\partial_t u_i(\boldsymbol{x}, t) + u_m(\boldsymbol{x}, t)\partial_{x_m} u_i(\boldsymbol{x}, t) = \nu \partial_{x_m}\partial_{x_m} u_i(\boldsymbol{x}, t) - \partial_{x_i} p(\boldsymbol{x}, t), \tag{2.1}$$

plus appropriate initial and boundary conditions. In (2.1), ∂_t and ∂_{x_i} denote partial differentiation with respect to time and space coordinates respectively, ν is the kinematic viscosity, p is pressure divided by mass density and repeated indices are summed over.

Turbulent transport is most conveniently described in terms of Lagrangian statistics

based on the representation of the turbulent velocity field by fluid particles. Particles are points lying on a trajectory $\boldsymbol{X}(t;\boldsymbol{x}_0)$ which are labelled by their initial position $\boldsymbol{X}(0;\boldsymbol{x}_0)=\boldsymbol{x}_0$ at the reference time $t=0$. The particle trajectories satisfy

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{X}(t;\boldsymbol{x}_0)=\boldsymbol{u}(t;\boldsymbol{x}_0), \tag{2.2}$$

where by definition the Lagrangian velocity $\boldsymbol{u}(t;\boldsymbol{x}_0)$ and the Eulerian velocity at the position of the fluid particle are identical: $\boldsymbol{u}(t;\boldsymbol{x}_0)=\boldsymbol{u}(\boldsymbol{X}(t;\boldsymbol{x}_0),t)$. Motivated by the Galilean invariance of the Navier–Stokes equations, as natural dynamical variables to study turbulent dispersion we choose the relative velocities of two fluid particles. At time t the particles are separated by a distance

$$\boldsymbol{r}(t)\equiv\boldsymbol{r}(t;\boldsymbol{r}_0,\boldsymbol{x}_0)=\boldsymbol{X}(t;\boldsymbol{x}_0+\boldsymbol{r}_0)-\boldsymbol{X}(t;\boldsymbol{x}_0), \tag{2.3}$$

where all particle pairs have the initial separation $\boldsymbol{r}(0)=\boldsymbol{r}_0$. The relative velocities are

$$\boldsymbol{v}(t)\equiv\boldsymbol{v}(t;\boldsymbol{r}_0,\boldsymbol{x}_0)=\boldsymbol{u}(\boldsymbol{X}(t;\boldsymbol{x}_0+\boldsymbol{r}_0),t)-\boldsymbol{u}(\boldsymbol{X}(t;\boldsymbol{x}_0),t), \tag{2.4}$$

where initially $\boldsymbol{v}_0\equiv\boldsymbol{v}(0;\boldsymbol{r}_0,\boldsymbol{x}_0)=\boldsymbol{u}(\boldsymbol{x}_0+\boldsymbol{r}_0,0)-\boldsymbol{u}(\boldsymbol{x}_0,0)$. Although we will often use the shorter notation, the dependence of $\boldsymbol{v}(t)$ and $\boldsymbol{v}_0$ on $\boldsymbol{r}_0$ and $\boldsymbol{x}_0$ is important for what follows. The dynamics of $\boldsymbol{r}(t)$ is obviously given by

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{r}(t)=\boldsymbol{v}(t). \tag{2.5}$$

The Navier–Stokes equation in terms of the Lagrangian velocity differences reads

$$\frac{\mathrm{d}}{\mathrm{d}t}v_i(t)=\nu\partial_{x_m}\partial_{x_m}v_i(t)-\partial_{x_i}\pi(t), \tag{2.6}$$

where $\pi(t)$ is the pressure difference

$$\pi(t)\equiv\pi(t;\boldsymbol{r}_0,\boldsymbol{x}_0)=p(\boldsymbol{X}(t;\boldsymbol{x}_0+\boldsymbol{r}_0),t)-p(\boldsymbol{X}(t;\boldsymbol{x}_0),t). \tag{2.7}$$

In equation (2.6) the partial derivatives with respect to x are to be interpreted such that first one has to expand the differences and then differentiate with respect to the first argument of the Eulerian variables. The time derivative $\mathrm{d}/\mathrm{d}t$ in (2.6) will be further investigated in § 4.

Describing the statistical state of the flow in terms of $\boldsymbol{v}$ and π is equivalent to specifying the static velocity structure functions such as $D_{ij}(\boldsymbol{r}_0)=\langle v_{0i}v_{0j}\rangle$, $D_{ijk}(\boldsymbol{r}_0)=\langle v_{0i}v_{0j}v_{0k}\rangle$, or the pressure structure function $D_{\pi\pi}(\boldsymbol{r}_0)=\langle\pi_0\pi_0\rangle$. We assume that the ensemble average $\langle\cdot\cdot\rangle$ is well defined for all moments of velocity and pressure up to sufficiently high order. The notation readily reflects the assumption that the ensemble is homogeneous and stationary with respect to the Eulerian coordinates $\boldsymbol{x},t$. Furthermore, owing to homogeneity the ensemble average is assumed to be equal to a space average $\langle(\cdot\cdot)\rangle=(1/V)\int_V\mathrm{d}\boldsymbol{x}_0(\cdot\cdot)$ over the initial positions of the particle pairs in the fluid volume V. Additionally, assuming reflection invariance and isotropy with respect to $\boldsymbol{r}$ allows the structure functions to be decomposed into longitudinal and transversal parts, for instance

$$D_{ij}(\boldsymbol{r})=D_{NN}(r)(\delta_{ij}-n_in_j)+D_{LL}(r)n_in_j, \tag{2.8}$$

where $r=|\boldsymbol{r}|$ and $\boldsymbol{n}=\boldsymbol{r}/r$.

The mean rate of energy dissipation per unit volume ϵ determines the Kolmogorov length, time, and velocity scales of the turbulence: $\eta=(\nu^3/\epsilon)^{1/4}$, $\tau_\eta=(\nu/\epsilon)^{1/2}$, $v_\eta=(\nu\epsilon)^{1/4}$, respectively. The Taylor–Reynolds number $Re_\lambda=u'\lambda/\nu$ is defined with the root-mean-square turbulence velocity u' and the Taylor microscale $\lambda=u'/(\langle\partial_{x_1}u_1\rangle)^{1/2}$.

As is well-known from the kinematics of homogeneous and isotropic turbulence, transversal structure functions can be expressed in terms of longitudinal ones owing to incompressibility (Monin & Yaglom 1975). Hence, the most important statistical quantity is the second-order longitudinal structure function $D_{LL}(r)$, which is very well approximated by a Batchelor type of interpolation function (Sawford & Hunt 1986)

$$D_{LL}(r) = 2u'^2\Big(\frac{r^2}{r_d^2+r^2}\Big)^{2/3}\Big(\frac{r^2}{r^2+r_L^2}\Big)^{1/3}, \tag{2.9}$$

here written down without any intermittency corrections.

In (2.9) the length scale $r_d = (15b_{LL})^{3/4}\eta$ determines the crossover from the viscous to the inertial subrange, where b_{LL} denotes the Kolmogorov constant. The crossover r_L from inertial to integral scales can be related to the integral scale L_1:

$$L_1 \equiv \int_0^\infty \Big(1 - \frac{D_{LL}(r)}{2u'^2}\Big)\mathrm{d}r \approx Br_L. \tag{2.10}$$

For large r ($r_d \approx 0$) the integral can be evaluated exactly: $B = \Gamma(\frac{5}{6})\Gamma(\frac{1}{2})/\Gamma(\frac{1}{3}) \approx 0.75$ (Borgas & Sawford 1994) and B increases slightly for finite r_d. The three asymptotic ranges of (2.9) for viscous, inertial, and integral scales can readily be identified.

The normalized energy dissipation rate c_ϵ defined as $\epsilon = c_\epsilon(Re_\lambda)u'^3/L_1$ is known to be Reynolds-number dependent for $Re_\lambda \leqslant 100$ (Sreenivasan 1984; Lohse 1995). The experimental data of Sreenivasan (1984) suggest a constant asymptotic value $c_\epsilon(\infty) \approx 1.0$ where saturation takes place around $Re_\lambda \approx 50$. Other authors give lower values, e.g. $c_\epsilon(\infty) \approx 0.8$ (Townsend 1976). DNS suggests $c_\epsilon \propto Re_\lambda^{-1/2}$ without a clear saturation in the range of Reynolds numbers ($38 \leqslant Re_\lambda \leqslant 93$) investigated by Yeung & Pope (1989). Since no fully consistent picture has yet emerged, we specify our numerical model by assuming the usual Kolmogorov constant $b_{LL} = 2.0$ and $B = 0.8$ which fixes $c_\epsilon = B(2/b_{LL})^{3/2} = 0.8$.

The pressure structure function $D_{\pi\pi}(r)$ depends only on $r = |\boldsymbol{r}|$ if one assumes local isotropy. Taylor expansion for small arguments yields for $r \ll \eta$

$$D_{\pi\pi}(r) = c_p(r/\eta)^2 v_\eta^4 + O(r^4). \tag{2.11}$$

The inertial range behaviour follows from dimensional analysis: $D_{\pi\pi}(r) \propto \epsilon^{4/3}r^{4/3}$. The variance of the pressure gradient $c_p = \frac{1}{3}\langle(\nabla p)^2\rangle\tau_\eta/\epsilon$ has been found to be strongly Reynolds-number dependent. Yeung & Pope (1989) give $c_p \propto Re_\lambda^{1/2}$ in the range of their DNS study. There is hardly any indication that c_p approaches a constant asymptotic value at high Reynolds numbers.

3. Stochastic modelling of relative dispersion

The key statistical quantity to study relative turbulent dispersion is the Lagrangian p.d.f. $P_L(\boldsymbol{v}, \boldsymbol{r}, t|\boldsymbol{r}_0)$. For example, P_L determines the mean-square separation of two fluid particles $\langle \boldsymbol{r}^2(t)\rangle$ averaged over ensembles of particle trajectories, which is related to the size of a pollutant cloud (Batchelor 1952). The p.d.f. P_L characterizes the probability of two fluid particles at time t having the velocity difference $\boldsymbol{v}$ and separation $\boldsymbol{r}$ given that they were separated by $\boldsymbol{r}_0$ initially and is defined as

$$P_L(\boldsymbol{v}, \boldsymbol{r}, t|\boldsymbol{r}_0) = \langle \delta(\boldsymbol{v} - \boldsymbol{v}(t))\delta(\boldsymbol{r} - \boldsymbol{r}(t))\rangle. \tag{3.1}$$

The connection between Lagrangian and Eulerian descriptions of turbulence is established via the fundamental relation due to Novikov (1969) (see also Pope 1985,

equation 4.24). In a homogeneous and incompressible flow the Eulerian p.d.f. $P_E(\boldsymbol{v}|\boldsymbol{r},t)$ of a velocity increment $\boldsymbol{v}$ between two fixed points in a distance $\boldsymbol{r}$ at time t is related to the Lagrangian p.d.f. P_L by

$$P_E(\boldsymbol{v}|\boldsymbol{r},t) = \int \mathrm{d}\boldsymbol{r}_0 P_L(\boldsymbol{v},\boldsymbol{r},t|\boldsymbol{r}_0), \tag{3.2}$$

where in a stationary state P_E is independent of time and initially

$$P_L(\boldsymbol{v},\boldsymbol{r},0|\boldsymbol{r}_0) = P_E(\boldsymbol{v}|\boldsymbol{r}_0)\delta(\boldsymbol{r}-\boldsymbol{r}_0). \tag{3.3}$$

As is shown in Appendix A, $P_E(\boldsymbol{v}|\boldsymbol{r})$ obeys the exact continuity equation

$$v_i \partial_{r_i} P_E + \partial_{v_i}(A_i P_E) = 0, \tag{3.4}$$

where $\boldsymbol{A}$ denotes the conditionally averaged relative acceleration vector for fixed velocity increments $\boldsymbol{v}$. Since the exact evolution of P_L in terms of P_E remains unknown, models are needed.

Two related modelling approaches have been suggested: the well-mixed condition of Thomson (1990) and a hierarchy of constraints of the model coefficients introduced by Novikov (1989). They both start from the assumption that the Lagrangian velocity difference evolves as a continuous Markov process obeying the (Ito-) stochastic differential equation

$$\mathrm{d}v_i(t) = a_i(\boldsymbol{r},\boldsymbol{v},t)\,\mathrm{d}t + b_{ij}(\boldsymbol{r},\boldsymbol{v},t)\,\mathrm{d}W_j(t), \tag{3.5}$$

where the component $\mathrm{d}W_j$ is the increment of a Wiener process or Gaussian white noise with correlation averaged over the ensemble of realizations of the stochastic force:

$$\langle \mathrm{d}W_i(t)\,\mathrm{d}W_j(t+\tau)\rangle = \delta_{ij}\delta(\tau)\,\mathrm{d}t\,\mathrm{d}\tau. \tag{3.6}$$

In §§ 4 and 5 we try to clarify the assumptions underlying a stochastic Lagrangian description such as (3.5).

The Lagrangian p.d.f. P_L then satisfies a Fokker–Planck equation equivalent to (3.5):

$$\partial_t P_L + v_i \partial_{r_i} P_L + \partial_{v_i}(a_i P_L) = \partial^2_{v_i v_j}(M_{ij} P_L), \tag{3.7}$$

where $M_{ij} = \frac{1}{2} b_{ik} b_{jk}$. Owing to the fundamental relation (3.2) the same equation is satisfied by the Eulerian p.d.f. P_E, but the first term in (3.7) then vanishes owing to stationarity.

Thomson's well-mixed criterion identifies the necessary and sufficient condition for the model coefficients $\boldsymbol{a}$ and $\boldsymbol{b}$ to satisfy

$$a_i = A_i + \frac{1}{P_E}\partial_{v_i}(M_{ij} P_E), \tag{3.8}$$

such that the conditional acceleration $\boldsymbol{A}$ as calculated from (3.8) satisfies the continuity equation (3.4). If P_E is assumed to be Gaussian models consistent with the well-mixed condition (3.8) have a conditional acceleration $\boldsymbol{A}$ quadratic in the velocities $\boldsymbol{v}(t)$ (Thomson 1990; Borgas & Sawford 1994). The stochastic force is assumed to be isotropic:

$$b_{ij} = b\delta_{ij}, \tag{3.9}$$

but (3.8) does not specify the models unambiguously.

The moments approximation method of Pedrizzetti & Novikov (1994) instead does

not explicitly assume a certain shape of P_E and employs the ansatz $a_i = -\gamma_{ij} v_j$. Using (3.9) they derive a hierarchy of constraints for the model coefficients by taking the moments equations implied by (3.4).

We have seen that once a model for the random trajectories is known it determines the Lagrangian and Eulerian statistics. The latter, i.e. P_E, is assumed to be given and hence imposes constraints on the model coefficients. Note that P_E cannot be Gaussian since the skewness or third-order structure function does not vanish by means of the Kolmogorov structure equation (Frisch 1995, chap. 6.2). Moreover, in practice only a few low-order moments of P_E are known to some accuracy.

Hence, in order to formulate a stochastic dispersion model one has to compromise. Either one assumes P_E to be Gaussian or of any other approximated form, for example the maximum missing information p.d.f. corresponding to the given information (Du *et al.* 1994), and constructs the model fully consistent with (3.8) and (3.4) (Thomson 1987, 1990), although one can solve (3.4) analytically only for particular forms of P_E. Or, the model coefficients are chosen such that only the known low-order moments are consistent with the Eulerian constraints (Pedrizzetti & Novikov 1994). We will follow the second approach since this allows the third-order structure function representing the mean energy transfer from the large to the small turbulent scales to be included. The theory developed in §§ 4 and 5 generalizes the stochastic model (3.5) by providing a systematic approach starting from the Navier–Stokes equation. Moreover, in the Markov approximation we obtain further information on the model coefficients $\boldsymbol{a}$ and $\boldsymbol{M}$. The reader only interested in the the dispersion properties of our model might skip the somewhat formal §§ 4 and 5 and continue with § 6. However, we try to keep the theoretical derivation self-consistent and do not presume any prior knowledge of the projector formalism.

Note that there are two different kinds of non-uniqueness for the construction of stochastic dispersion models: different Lagrangian models with different dispersion statistics can be consistent with given Eulerian statistics. The first non-uniqueness is well-known and refers to the determination of $\boldsymbol{A}$. The conditional acceleration $\boldsymbol{A}$ requires information about the joint p.d.f. of relative velocities and accelerations. Many choices of $\boldsymbol{A}$ can be consistent with the marginal p.d.f. $P_E(\boldsymbol{v}|\boldsymbol{r})$. The second non-uniqueness is due to the memory $\boldsymbol{M}$. As one can see from (3.4) any choice of $\boldsymbol{M}$ is consistent with the Eulerian constraint if the model is formulated according to the well-mixed criterion (3.8). The exact Langevin equation in § 4 generalizes the well-mixed condition; however, the (exact) Eulerian constraint (3.4) remains unchanged. Hence, it is not possible to determine the memory from Eulerian consistency requirements, which explains the persistent lack of a rigorous theory for turbulent transport. Fortunately, highly simplifying assumptions on $\boldsymbol{M}$ already permit many dynamical features of turbulent dispersion to be included as shown in the numerical simulations of § 6.

4. The generalized Langevin equation

We first define the spaces of the relevant variables and then investigate their time derivatives, i.e. the $\mathrm{d}/\mathrm{d}t$ in (2.6). The relative velocities (2.4) and pressure (2.7) are differences of u_i and p at time t at the positions $\boldsymbol{x}_1 = \boldsymbol{X}(t; \boldsymbol{x}_0 + \boldsymbol{r}_0)$ and $\boldsymbol{x}_2 = \boldsymbol{X}(t; \boldsymbol{x}_0)$. Consequently, we introduce the space of variables

$$\mathscr{M} = \{A(\boldsymbol{x}_1, \boldsymbol{x}_2, t) : \boldsymbol{x}_1 = \boldsymbol{X}(t; \boldsymbol{x}_0 + \boldsymbol{r}_0), \boldsymbol{x}_2 = \boldsymbol{X}(t; \boldsymbol{x}_0)\}, \tag{4.1}$$

where $A(t)$ may be any sum of products of velocity components $u_i(\boldsymbol{x}, t), i = 1, 2, 3$, and $p(\boldsymbol{x}, t)$. This definition includes all powers $\boldsymbol{v}^n(t)$ and their generating function, i.e. the three-dimensional delta-function $\delta(\boldsymbol{v} - \boldsymbol{v}(t))$ by means of $\boldsymbol{v}^n(t) = \int \mathrm{d}\boldsymbol{v}\boldsymbol{v}^n\delta(\boldsymbol{v} - \boldsymbol{v}(t))$.

Important for the following will be the subspace of the relevant variables at the reference time $t = 0$

$$\mathscr{M}_0 = \{A_0 \equiv A(0) : A(t) \in \mathscr{M}\}. \tag{4.2}$$

Although we assume the Eulerian statistics to be stationary, the relative dispersion of two fluid particles is a non-stationary process and the particle-pair mean-square separation increases with time. GT82 realized that this implies a non-symmetric time evolution operator L and gave an explicit representation of L.

This evolution in time is governed by the Liouville operator $L : \mathscr{M}_0 \to \mathscr{M}_0$ defined as

$$LA_0 = \frac{\mathrm{d}}{\mathrm{d}t}A(t)|_{t=0}. \tag{4.3}$$

Furthermore

$$A(t) = \sum_{n=0}^{\infty} \frac{t^n}{n!}\frac{\mathrm{d}^n}{\mathrm{d}t^n}A(t)|_{t=0} = \mathrm{e}^{Lt}A_0, \tag{4.4}$$

where differentiation of the last equation gives

$$\frac{\mathrm{d}}{\mathrm{d}t}A(t) = \mathrm{e}^{Lt}LA_0. \tag{4.5}$$

Equation (4.5) will be the starting point for the projector method.

An explicit representation of L in $\mathscr{M}_0$ can be obtained from

$$\frac{\mathrm{d}}{\mathrm{d}t}A(t) = \left(\partial_t + u_m(\boldsymbol{x}_1, t)\partial_{x_{1m}} + u_m(\boldsymbol{x}_2, t)\partial_{x_{2m}}\right)A(t)\Big|_{\boldsymbol{x}_2=\boldsymbol{X}(t;\boldsymbol{x}_0)}^{\boldsymbol{x}_1=\boldsymbol{X}(t;\boldsymbol{x}_0+\boldsymbol{r}_0)}, \tag{4.6}$$

and in the limit $t \to 0$ where $\boldsymbol{x}_1 = \boldsymbol{x}_0 + \boldsymbol{r}_0$ and $\boldsymbol{x}_2 = \boldsymbol{x}_0$,

$$L = \partial_t + u_m(\boldsymbol{x}_0, 0)\partial_{x_m} + v_m(0; \boldsymbol{r}_0, \boldsymbol{x}_0)\partial_{r_m}, \tag{4.7}$$

together with a skewness formula expressing the non-stationarity or time reversal asymmetry of relative dispersion $\boldsymbol{a}_0, \boldsymbol{b}_0 \in \mathscr{M}_0$

$$\langle \boldsymbol{a}_0 L\boldsymbol{b}_0\rangle + \langle (L\boldsymbol{a}_0)\boldsymbol{b}_0\rangle = \partial_{r_m}\langle v_{m0}\boldsymbol{a}_0\boldsymbol{b}_0\rangle. \tag{4.8}$$

Note that incompressibility implies $\partial_{x_j}u_j(\boldsymbol{x}, t) = 0$, but only $\partial_{r_j}v_{0j} = 0$ at $t = 0$ with no general statement for $t \neq 0$.

4.1. *The linear projector method*

In order to emphasize the difference between our approach and the work of GT82, we will now briefly summarize their results. GT82 introduce the ensemble average as an inner product in $\mathscr{M}$, where they defined $\mathscr{M}$ more restrictively, and define a projector P_{GT} in $\mathscr{M}_0$ on the initial velocity differences

$$P_{GT}A_{i0} = v_{j0}\langle v_{j0}v_{k0}\rangle^{-1}\langle v_{k0}A_{i0}\rangle \tag{4.9}$$

in accordance with the linear Mori–Zwanzig theory (Lindenberg & West 1990). The projector method was applied in the form of a resolvent decomposition of the Laplace-transformed Lagrangian correlation function. Neglecting memory effects in the so-called relaxation approximation the Lagrangian correlation then decays exponentially:

$$\langle v_{i0}\mathrm{e}^{Lt}v_{j0}\rangle = D_{ik}(\boldsymbol{r}_0)(\mathrm{e}^{-\gamma(\boldsymbol{r}_0)t})_{kj}, \tag{4.10}$$

where the relaxation matrix is given by

$$\gamma_{ij}(\boldsymbol{r}_0) = -\tfrac{1}{2}\partial_{r_m} D_{mik}(\boldsymbol{r}_0) D^{-1}_{kj}(\boldsymbol{r}_0). \tag{4.11}$$

By the same reasoning to be applied below, the linear theory yields a linear Langevin equation where the coefficients depend on the initial separation $\boldsymbol{r}_0$. By using the usual white noise approximations for the random force, the linear theory is thus incapable of capturing an intermediate scaling range between the initial ballistic and the final Taylor diffusive behaviour of the dispersion. However, we can overcome the restrictions of the linear theory, if we phenomenologically replace the dependence on $\boldsymbol{r}_0$ by a dependece on the instantaneous $\boldsymbol{r}(t)$. This replacement is not supported by the linear theory, but yields the same nonlinear model of § 6 (Heppe 1997).

The nonlinear theory of the next section allows us to obtain a nonlinear stochastic model generalizing the relative dispersion models of Thomson (1990) and Pedrizzetti & Novikov (1994). The linear theory although formally exact is unsuitable for simple approximations since the nonlinear couplings are included implicitly via a complicated dynamics of the random force.

4.2. *The nonlinear projector method*

A nonlinear Langevin equation results from treating the nonlinear interactions among the relevant dynamical variables $\boldsymbol{v}(t)$ explicitly, especially those due to the Lagrangian paths. Those nonlinear couplings are always of importance if the steady state itself is a state far from thermal equilibrium (Grabert, Hanggi & Talkner 1980). The main difference between the linear and the nonlinear theory lies in the choice of the projector P. In the linear theory P was a projection onto the initial velocities $\boldsymbol{v}_0$ (4.9). According to the nonlinear theory we project onto the generating function of all polynomials in $\boldsymbol{v}_0$, a larger subset of $\mathscr{M}_0$. Projection onto the subspace spanned by the set $\delta(\boldsymbol{v}-\boldsymbol{v}_0)$ is the same as projection onto the subspace of all functions of the $\boldsymbol{v}_0$.

The generating function is obviously related to the fine-grained Lagrangian p.d.f. defined as $\delta(\boldsymbol{v}-\boldsymbol{v}(t))\delta(\boldsymbol{r}-\boldsymbol{r}(t))$. Its ensemble average gives the Lagrangian p.d.f. P_L (3.1).

As in the linear theory, for $a, b \in \mathscr{M}$ the ensemble average of particle pairs with a fixed initial separation $\boldsymbol{r}_0$ is introduced as an inner product $\langle ab\rangle$. Because of the fundamental relation (3.2) and from the delta-function property

$$\delta(\boldsymbol{v}-\boldsymbol{v}_0)\delta(\boldsymbol{v}'-\boldsymbol{v}_0) = \delta(\boldsymbol{v}-\boldsymbol{v}')\delta(\boldsymbol{v}-\boldsymbol{v}_0), \tag{4.12}$$

and $\langle\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle = P_E(\boldsymbol{v}|\boldsymbol{r}_0)$ we have the following identity:

$$\langle\delta(\boldsymbol{v}-\boldsymbol{v}_0)\delta(\boldsymbol{v}'-\boldsymbol{v}_0)\rangle = P_E(\boldsymbol{v}|\boldsymbol{r}_0)\delta(\boldsymbol{v}'-\boldsymbol{v}). \tag{4.13}$$

We can now define a projection operator $P : \mathscr{M} \to \mathscr{M}_0$ as

$$P\boldsymbol{b}(t;\boldsymbol{r}_0,\boldsymbol{x}_0) = \int \mathrm{d}\boldsymbol{v}\, \frac{\langle \boldsymbol{b}(t)\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle}{P_E(\boldsymbol{v}|\boldsymbol{r}_0)}\delta(\boldsymbol{v}-\boldsymbol{v}_0). \tag{4.14}$$

Here, the dependence of $P\boldsymbol{b}(t;\boldsymbol{r}_0,\boldsymbol{x}_0)$ on the initial values $\boldsymbol{r}_0$ and $\boldsymbol{x}_0$ via $\delta(\boldsymbol{v}-\boldsymbol{v}_0)$ should be noted. In the following we will make use of the conditional average of $\boldsymbol{b}(t)$ for a given velocity increment $\boldsymbol{v}$ (and a fixed $\boldsymbol{r}_0$) defined as $\langle\boldsymbol{b}(t)|\boldsymbol{v}\rangle = \langle\boldsymbol{b}(t)\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle/P_E(\boldsymbol{v}|\boldsymbol{r}_0)$. The definition (4.14) then identifies the projector as an average conditional upon the initial velocity difference $P\boldsymbol{b}(t) = \langle\boldsymbol{b}(t)|\boldsymbol{v}_0\rangle$ from which the properties of P as an orthogonal projector, $P^2 = P$ and $\langle aPb\rangle = \langle(Pa)b\rangle$, can readily be verified. Furthermore, P projects the initial generating function onto itself: $P\delta(\boldsymbol{v}-\boldsymbol{v}_0) =$

$\delta(\boldsymbol{v}-\boldsymbol{v}_0)$. All averages thus depend parametrically on $\boldsymbol{r}_0$, but this dependence will not be expressed explicitly in the following.

The projector method exactly rewrites the Liouville equation (4.5) in a form that more closely resembles a Langevin equation by means of the projector P and its complement $Q \equiv 1 - P$:

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{v}(t) = L\boldsymbol{v}(t) = \mathrm{e}^{Lt}L\boldsymbol{v}_0 = \mathrm{e}^{Lt}PL\boldsymbol{v}_0 + \mathrm{e}^{Lt}QL\boldsymbol{v}_0. \tag{4.15}$$

The second term of (4.15) can be rewritten by use of the operator identity

$$\mathrm{e}^{Lt} = \int_0^t \mathrm{e}^{L(t-\tau)}PL\mathrm{e}^{QL\tau}\,\mathrm{d}\tau + \mathrm{e}^{QLt}, \tag{4.16}$$

which can be verified by Laplace transform. Inserting (4.16) into (4.15) yields the generalized Langevin equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{v}(t) = \mathrm{e}^{Lt}PL\boldsymbol{v}_0 + \int_0^t \mathrm{e}^{L(t-\tau)}PL\mathrm{e}^{QL\tau}QL\boldsymbol{v}_0\,\mathrm{d}\tau + \mathrm{e}^{QLt}QL\boldsymbol{v}_0. \tag{4.17}$$

The first term in (4.17) is the conditional average of the relative acceleration:

$$\mathrm{e}^{Lt}PL\boldsymbol{v}_0 = \int \mathrm{d}\boldsymbol{v}\langle L\boldsymbol{v}_0|\boldsymbol{v}(\boldsymbol{r})\rangle\delta(\boldsymbol{v}-\boldsymbol{v}(t)) = \langle L\boldsymbol{v}_0|\boldsymbol{v}(t)\rangle. \tag{4.18}$$

In (4.18) we have used $\mathrm{e}^{Lt}\delta(\boldsymbol{v}-\boldsymbol{v}_0) = \delta(\boldsymbol{v}-\boldsymbol{v}(t))$ which follows from (4.4) and the fact that L is a first-order differential operator (4.7). The last equality in (4.18) means that first one has to calculate the conditional average $\langle L\boldsymbol{v}_0|\boldsymbol{v}\rangle$ and then replace $\boldsymbol{v}$ by $\boldsymbol{v}(t)$.

We first discuss the third term in (4.17), the so-called random force

$$\boldsymbol{f}(t) \equiv \boldsymbol{f}(t;\boldsymbol{r}_0,\boldsymbol{x}_0) = \mathrm{e}^{QLt}QL\boldsymbol{v}_0, \tag{4.19}$$

which in general is very difficult to evaluate exactly. By construction $\boldsymbol{f}(t)$ is orthogonal to all functions of $\boldsymbol{v}_0$ at all times $P\boldsymbol{f}(t) = 0$, which implies that $\langle \boldsymbol{f}(t)\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle = 0$ and after integration over $\boldsymbol{v}$ that the ensemble average of $\boldsymbol{f}(t)$ vanishes:

$$\langle \boldsymbol{f}(t)\rangle = 0, \tag{4.20}$$

which suggests that $\boldsymbol{f}(t)$ might successfully be modelled as a stochastic force.

The time integral in the second term in (4.17) allows us to identify this term as a memory contribution since the integrand depends on all times $0 \leqslant \tau \leqslant t$. The memory term is related to the random force by definition of the projection operator (cf. (4.18)):

$$\int_0^t \mathrm{d}\tau\mathrm{e}^{L(t-\tau)}\int \mathrm{d}\boldsymbol{v}\langle L\boldsymbol{f}(\tau)|\boldsymbol{v}\rangle\delta(\boldsymbol{v}-\boldsymbol{v}_0) = \int_0^t \mathrm{d}\tau\langle L\boldsymbol{f}(\tau)|\boldsymbol{v}(t-\tau)\rangle. \tag{4.21}$$

As shown in Appendix A, a physically more suggestive form of the memory kernel can be derived which holds only in stationary and homogeneous turbulence:

$$\langle Lf_i(\tau)|\boldsymbol{v}\rangle = \frac{1}{P_E}\partial_{v_j}\Big(\langle f_i(\tau)f_j(0)|\boldsymbol{v}\rangle P_E\Big). \tag{4.22}$$

Hence, the random force and the memory are related by a fluctuation–dissipation relation. In other words, the memory is essentially the conditionally averaged correlation function of the random forces.

Thus we have seen that for each choice of the projection operator P the Mori–Zwanzig method yields a unique decomposition of the time rate of change of the

Lagrangian velocity differences into a systematic and a random part. The random part has the property that its statistical average over realizations of the stochastic process vanishes. The systematic part consists of the relative acceleration conditionally averaged over the (stationary) Eulerian ensemble and a memory term induced by the random forces.

We have transformed the dynamical system (2.5) and (2.6) for an ensemble of particle pairs into an exact generalized Langevin equation (4.17) which including the above definitions can be rewritten as

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{r}(t) = \boldsymbol{v}(t), \tag{4.23}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{v}(t) = \langle L\boldsymbol{v}_0|\boldsymbol{v}(t)\rangle + \int_0^t \mathrm{d}\tau \Big[\frac{1}{P_E}\partial_{v_j}\Big(\langle \boldsymbol{f}(\tau) f_j(0)|\boldsymbol{v}\rangle P_E\Big)\Big]_{\boldsymbol{v}=\boldsymbol{v}(t-\tau)} + \boldsymbol{f}(t). \tag{4.24}$$

Alternatively, the generalized Langevin equation (4.24) can be derived from the evolution equation of $\delta(\boldsymbol{v}-\boldsymbol{v}(t))$ (Heppe 1997). Only the conditional acceleration appears in the continuity equation for the Eulerian p.d.f. $P_E(\boldsymbol{v}|\boldsymbol{r})$ (3.4):

$$v_m \partial_{r_m} P_E + \partial_{v_m}(\langle L v_{0m}|\boldsymbol{v}\rangle P_E) = 0. \tag{4.25}$$

Note that this equation is an exact consequence of the skewness formula (4.8) for homogeneous and stationary turbulence, i.e. it holds for arbitrary approximations of the memory, which will be discussed in the next section.

5. Approximations of the model coefficients

For the derivation of the generalized Langevin equation (4.24) in the previous section we have only assumed homogeneity and stationarity of the turbulent ensemble. In order to obtain useful stochastic models from (4.24) we have to approximate the drift and the memory term and need to make further assumptions about the time evolution of the random forces. Additionally, for simplicity we will assume local isotropy such that scalar functions of the separation depend only on its magnitude $r(t) = |\boldsymbol{r}(t)|$ and tensors can be decomposed into longitudinal and transversal parts (e.g. (2.8)).

5.1. *The conditional acceleration*

In applications of the nonlinear projector method, one part of the systematic drift, the conditionally averaged acceleration, is usually approximated by expanding $\delta(\boldsymbol{v}-\boldsymbol{v}_0)$ in $\langle L\boldsymbol{v}_0|\boldsymbol{v}\rangle$ in complete orthogonal polynomials (Zwanzig 1980). If $P_E(\boldsymbol{v}|\boldsymbol{r})$ were Gaussian, Hermite polynomials would be the natural choice corresponding to a Gram–Chalier expansion (Lumley 1970) of the joint p.d.f. of $L\boldsymbol{v}$ and $\boldsymbol{v}$. This yields an expansion of the form

$$A_i(\boldsymbol{v}) = \langle L\boldsymbol{v}_0|\boldsymbol{v}\rangle = \alpha_i + \beta_{ij}v_j + \gamma_{ijk}v_j v_k + \ldots. \tag{5.1}$$

Rather than expanding $\delta(\boldsymbol{v}-\boldsymbol{v}_0)$ we regard (5.1) as an ansatz for the nonlinear least-mean-square estimate of the conditional average $\boldsymbol{A}$ (Adrian 1996). Minimizing the mean-square error of the estimate yields a system of equations for the unknown (non-random) coefficients α_i, β_{ij}, and γ_{ijk}

$$\left.\begin{aligned} &\langle (A_i - \alpha_i - \beta_{ij}v_j - \gamma_{ijk}v_j v_k)\rangle = 0, \\ &\langle (A_i - \alpha_i - \beta_{ij}v_j - \gamma_{ijk}v_j v_k)v_l\rangle = 0, \\ &\langle (A_i - \alpha_i - \beta_{ij}v_j - \gamma_{ijk}v_j v_k)v_l v_n\rangle = 0, \end{aligned}\right\} \tag{5.2}$$

and so forth. Noting that ensemble averaging is equivalent to integrating the Eulerian p.d.f. P_E over velocity increments, the first term in each equation of (5.2) gives the joint moment or structure function of the relative acceleration Lv_i and 1, v_l, $v_l v_n \ldots$ due to $\langle\langle L\boldsymbol{v}_0|\boldsymbol{v}\rangle h(\boldsymbol{v})\rangle = \langle (L\boldsymbol{v}_0)h(\boldsymbol{v}_0)\rangle$ where h is an arbitrary function of its argument. Since we assume the mean values of $L\boldsymbol{v}$ and $\boldsymbol{v}$ to vanish, the first equation of (5.2) gives

$$\alpha_i + \gamma_{ijk} D_{jk} = 0. \tag{5.3}$$

The skewness formula (4.8) determines the joint acceleration–velocity structure function, which by isotropy equals $\langle (Lv_i)v_j\rangle = \frac{1}{2}\partial_{r_m} D_{mij}$. The linear estimate of $\boldsymbol{A}$ therefore yields

$$A_i^{(1)} = \tfrac{1}{2}\{\partial_{r_m} D_{mij}\} D_{jk}^{-1} v_k. \tag{5.4}$$

This is exactly the result of GT82 for their linear drift term (4.11). The divergence of the third-order structure function is well known from the Kolmogorov structure equation and is essentially the energy dissipation rate on inertial-range scales (see (6.1), (6.2)). This implies that the local Lagrangian time scale t_r of two-particle dispersion in the inertial range is given by $t_r \propto D_{LL}(r)/\epsilon \propto \epsilon^{-1/3} r^{2/3}$.

On the other hand, if we assume P_E to be Gaussian distributed, $P_E \propto \exp(-\frac{1}{2} v_i D_{ij}^{-1} v_j)$, the linear estimate (5.4) vanishes and one has to include terms quadratic in the velocities in (5.1). In this case there is no unique solution of (5.2). The solution which takes $\langle (Lv_i)v_j v_k\rangle$ to be fully symmetric and hence equal to $\frac{1}{3}\partial_{r_m} D_{mijk}$ is

$$A_i^G = -\tfrac{1}{6}\{\partial_{r_m} D_{mijk}\} D_{jk}^{-1} + \tfrac{1}{6}\{\partial_{r_m} D_{mijk}\} D_{jl}^{-1} D_{kn}^{-1} v_l v_n. \tag{5.5}$$

By use of $\partial_{r_m} D_{mi} = 0$ and the Gaussian property $D_{mijk} = D_{mi}D_{jk} + D_{mj}D_{ik} + D_{mk}D_{ij}$ (5.5) can be rewritten as

$$A_i^G = \tfrac{1}{6} D_{mi} D_{jk} \partial_{r_m} D_{jk}^{-1} + \tfrac{1}{6}\Big(-D_{mi}\partial_{r_m} D_{nl}^{-1} + D_{kn}^{-1}\partial_{r_l} D_{ik} + D_{jl}^{-1}\partial_{r_n} D_{ij}\Big) v_l v_n. \tag{5.6}$$

Note that the order of the matrices does not matter owing to the assumed isotropy. Each of the three contributions in the quadratic term in (5.6) are known from the well-mixed approach (Borgas & Sawford 1994). The least-mean-square estimate taken up to second order for arbitrary P_E yields much more complicated coefficients. For simplicity we will include only the linear drift term $\boldsymbol{A}^{(1)}$ in the numerical simulations in § 6.

5.2. *The memory term*

The first obvious approximation of the memory term in the generalized Langevin equation (4.24) is a coarse graining in time or equivalently the Markov assumption. The stochastic process in the separation–velocity phase space is assumed to be Markovian, i.e. the values of $\boldsymbol{r}(t)$ and $\boldsymbol{v}(t)$ at times greater than t depend only on the present separation and velocity and are independent of the history of values at times less than t. While a rigorous justification for the Markov assumption in turbulent transport is still lacking, it has been well known since the work of Taylor (1921). For the assumption to hold, the relative acceleration correlations have to decay rapidly. There is some indication that the acceleration of one fluid particle de-correlates on a time scale proportional to the Kolmogorov time scale τ_η (Sawford 1991), i.e. diminishes with increasing Reynolds number Re_λ. However, Yeung (1997) has recently found surprisingly long-time persistence of the two-particle acceleration correlations in his DNS at moderate Reynolds number ($Re_\lambda = 140$). Hence, on small

scales at moderate Re_λ the Markov approximation has to be regarded as a first guess only.

We assume that the characteristic time scale τ_f of the random force $\boldsymbol{f}(t)$ is distinctly shorter than the time scale for an appreciable change of the velocity difference $\boldsymbol{v}(t)$, and we expect at least for $t \gg \tau_f$ the approximation to be reasonable. If $\tau_f \propto \tau_\eta$, the approximation becomes better for increasing Re_λ. At times of the order of τ_η the Markov assumption is likely to fail and we have to expect qualitative deviations of a Markovian model from the dispersion physics. However, the Markov approximation might be valid in the inertial range of scales $\tau_\eta \ll t \ll T_L$, where T_L is an integral time scale, and allows efficient numerical solutions of the Langevin equation (4.24).

In the Markov approximation the memory $\int_0^t \mathrm{d}\tau \langle L\boldsymbol{f}(\tau)|\boldsymbol{v}(t-\tau)\rangle$ is replaced by

$$\int_0^\infty \mathrm{d}\tau \langle Lf_i(\tau)|\boldsymbol{v}(t)\rangle = \Big[\frac{1}{P_E}\partial_{v_j}\Big(M_{ij}(\boldsymbol{v})P_E\Big)\Big]_{\boldsymbol{v}=\boldsymbol{v}(t)}, \tag{5.7}$$

where we have introduced the memory matrix

$$M_{ij}(\boldsymbol{v}) = \int_0^\infty \mathrm{d}\tau \langle f_i(\tau) f_j(0)|\boldsymbol{v}\rangle. \tag{5.8}$$

Equivalently, coarse graining in time means $\langle f_i(\tau) f_j(0)|\boldsymbol{v}\rangle = 2\delta(\tau) M_{ij}(\boldsymbol{v})$.

We still need an explicit expression for the memory matrix $\boldsymbol{M}$. Previous Markov models are recovered by assuming the memory to be independent of the separation and velocity. Consistency with Kolmogorov theory in the inertial range yields

$$M_{ij} \propto \epsilon \delta_{ij}, \tag{5.9}$$

or with slightly greater generality $M_{ij} = \epsilon(an_{ij} + b(\delta_{ij} - n_i n_j))$, where a and b are constant. Here we obtain a dynamical justification of (5.9) from the operator expression for the force (4.19) or the memory.

If one assumes the relative acceleration $L\boldsymbol{v}_0$ to be a small quantity, which is hard to justify in turbulence because of the lack of statistical information on the pressure term in (2.6), an explicit expression for $\boldsymbol{M}$ can be obtained via an expansion in $L\boldsymbol{v}_0$ (Zwanzig 1961; Grabert 1982). Retaining only terms up to third order, the memory reads

$$M_{ij}(\boldsymbol{v}) = \int_0^\infty \mathrm{d}\tau \langle [Lv_i(\tau) - \langle Lv_i(\tau)|\boldsymbol{v}\rangle][Lv_{0j} - \langle Lv_{0j}|\boldsymbol{v}\rangle]|\boldsymbol{v}\rangle + O((L\boldsymbol{v}_0)^3). \tag{5.10}$$

The lowest-order stochastic estimate of (5.10) is independent of the velocity increments since the mean relative acceleration vanishes:

$$M_{ij} \approx \tau_a \langle Lv_{0i} Lv_{0j}\rangle, \tag{5.11}$$

where $\tau_a = t_a \tau_\eta \approx \tau_f$ is the correlation time of the relative accelerations. The structure function of the relative acceleration in (5.11) can be evaluated by replacing $L\boldsymbol{v}_0$ with the right-hand side of the Navier–Stokes equation (2.6), (GT82). We can see now that an external force, which maintains stationarity by forcing the large scales only, does not contribute for particle separations on sub-integral scales, because its structure function vanishes by definition for $r \ll L_1$. Of course, on integral scales the forcing contributes and has to be included in the theoretical description. Without the external force we have

$$\langle Lv_{0i} Lv_{0j}\rangle = v^2 \nabla_r^4 D_{ij}|_0^{r_0} - \nabla_r^2 D_{\pi\pi}|_0^{r_0}, \tag{5.12}$$

where we set $\partial_{r_m}\partial_{r_m} = \nabla_r^2$. Neglecting the viscous term in the inertial range and on

integral scales, the longitudinal and transversal components equal $\frac{2}{3}\langle(\nabla p)^2\rangle v_\eta^2/\tau_\eta^2$ and hence

$$M_{ij} = \tfrac{2}{3} t_a \epsilon \langle(\nabla p)^2\rangle \delta_{ij}. \tag{5.13}$$

In the viscous range both terms in (5.12) contribute and scale as r^2. Therefore, consistency requires t_a to be independent of the separation in the viscous as well as the inertial range.

5.3. *The random force*

The nonlinear Langevin equation (4.24) determines the stochastic process for the Lagrangian velocity differences in term of the process of the random force $\boldsymbol{f}(t)$. Except for very simple cases the operator expression (4.19) does not allow the stochastic process of $\boldsymbol{f}(t)$ to be evaluated completely (Zwanzig 1980). Even if we knew the memory, i.e. the force correlation $\langle f_i(\tau) f_j(0)|\boldsymbol{v}\rangle$ together with the vanishing mean (4.20), this would not determine the stochastic process of $\boldsymbol{f}(t)$ completely. Indeed, we must know all time correlation functions involving more than two f like

$$\langle f_{j_1}(t_1) f_{j_2}(t_2) \dots f_{j_n}(t_n)|\boldsymbol{v}\rangle. \tag{5.14}$$

Those higher-order correlations had to be extracted from (4.19) which seems to be intractable. The only sensible thing to do for practical models is to assume that $\boldsymbol{f}(t)$ obeys a Gaussian stochastic process and is independent of $\boldsymbol{v}$. The conditional average $\langle\cdot|\boldsymbol{v}\rangle$ is then equivalent to the ensemble average and it is enough to specify

$$\langle f_i(t) f_j(s)\rangle = \langle f_i(t-s) f_j(0)\rangle \tag{5.15}$$

since higher-order correlations factorize:

$$\langle f_{j_1}(t_1) f_{j_2}(t_2) \dots f_{j_n}(t_n)\rangle = \sum_{\text{perm}} \prod_{m=1}^{n} \langle f_{j_m}(t_m) f_{j'_m}(t'_m)\rangle, \tag{5.16}$$

where the set $\{m'\}$ is obtained from $\{m\}$ by a permutation of j_{n+1}, $j_{n+2}, \dots j_{2n}$ and the sum is over all permutations. Note that a similar property to (5.15) does not hold in general for the conditional average $\langle f_i(t-s) f_j(0)|\boldsymbol{v}\rangle$.

The usual interpretation of a Langevin equation

$$\frac{\mathrm{d}}{\mathrm{dt}} a(t) = g(a(t)) + \zeta(t) \tag{5.17}$$

is that of an equation in the state space Σ_a and two kinds of averages are explained: the average over the random forces ζ, which is done with fixed values of the dynamical variables a and which is specified by the correlations $\langle\zeta(t_1)...\zeta(t_n)|a\rangle$ and an average over the dynamical variables $\langle..\rangle = \int_\Sigma \mathrm{d}a P(a) \dots$. Consequently, if we identify $\langle\zeta(t_1)...\zeta(t_n)|a\rangle$ with $\langle f(t_1)...f(t_n)|\boldsymbol{v}\rangle$, the generalized Langevin equation (4.24) shows exactly the structure of the familiar stochastic differential equation.

From the Markov approximation (5.7) together with the assumption of a Gaussian $\boldsymbol{f}(t)$, we obtain a Fokker–Planck equation for the Lagrangian p.d.f. $P_L(\boldsymbol{v}, \boldsymbol{r}, t|\boldsymbol{r}_0)$ by standard methods or from the evolution equation for $\delta(\boldsymbol{v} - \boldsymbol{v}(t))$ (Heppe 1997):

$$\partial_t P_L + v_m \partial_{r_m} P_L = -\partial_{v_i}\Big(\langle L v_{0i}|\boldsymbol{v}\rangle P_L\Big) + \partial_{v_i}\Big(P_E M_{ij}(\boldsymbol{v}) \partial_{v_j}\{P_L/P_E\}\Big); \tag{5.18}$$

or by assuming the memory to have no explicit dependence on $\boldsymbol{v}$:

$$\partial_t P_L + v_m \partial_{r_m} P_L = -\partial_{v_i}\Big(\{\langle L v_{0i}|\boldsymbol{v}\rangle + M_{ij}\partial_{v_j}\log P_E\} P_L\Big) + M_{ij}\partial_{v_i}\partial_{v_j} P_L. \tag{5.19}$$

The Fokker–Planck equation (5.19) is the one considered by Thomson (1990) with the generalization of a memory matrix possibly dependent on the particle separation. Note that the assumption that the memory is independent of velocity increments is required to avoid the Ito–Stratonovich ambiguity in the interpretation of the stochastic integrals in the Langevin equation (4.24).

6. Numerical simulations of the dispersion model

We specify a stochastic dispersion model in accordance with the theory of § 4 employing the approximations discussed in § 5 and solve it numerically for a large number of particle pairs (2×10^4). The conditional acceleration is given by its linear stochastic estimate $A_i = -\gamma_{ij} v_j$ (5.4). Using the Kolmogorov structure equation $\Gamma_{ij} \equiv \gamma_{ik} D_{kj} = -\frac{1}{2} \partial_{r_m} D_{mij}$ can be expressed in terms of the longitudinal second-order structure function

$$\Gamma_{NN}(r) = \tfrac{2}{3}\epsilon - \frac{v}{2r}\Big(r^2 D'''_{LL}(r) + 6r D''_{LL}(r) + 4D'_{LL}(r)\Big), \tag{6.1}$$

$$\Gamma_{LL}(r) = \tfrac{2}{3}\epsilon - \frac{v}{r}\Big(r D''_{LL}(r) + 4D'_{LL}(r)\Big), \tag{6.2}$$

where a prime denotes the derivative with respect to r. From the Batchelor interpolation (2.9) the normalized relaxation coefficients in the viscous subrange (VSR) read $\gamma_{LL} = \gamma_{NN} = \gamma_{\mathrm{VSR}}/\tau_\eta \equiv \frac{56}{3}(15 b_{LL})^{-3/2}/\tau_\eta$. Furthermore we can see from (6.1) and (6.2) that the leading contribution to Γ_* ($* = NN, LL$) in the inertial range is given by the energy dissipation rate (cf. GT82).

We approximate $D_{NN}(r)$ and $\Gamma_*(r)$ by interpolation functions analogous to (2.9) to simplify the numerical calculations. For scales $r \leqslant L_1$ we use

$$\tilde{\Gamma}_*(r) = \frac{2}{3}\frac{(r/\eta)^2}{r_{\Gamma *}^2 + (r/\eta)^2} v_\eta^2/\tau_\eta, \tag{6.3}$$

where $r_{\Gamma LL}^2 = 10/\gamma_{\mathrm{VSR}}$ and $r_{\Gamma NN}^2 = 5/\gamma_{\mathrm{VSR}}$ so that γ_{VSR} determines the crossover from viscous to integral scales. The relaxation $\boldsymbol{A}$ will be switched off on integral scales, where $\partial_{r_m} D_{mij}$ vanishes if one-point statistics are Gaussian which is usually a reasonable assumption in isotropic turbulence.

In accordance with K41 dimensional analysis we also employ $\tilde{\Gamma}_*$ for the memory:

$$M_{ij}(\boldsymbol{r}) = a_x \tilde{\Gamma}_{ij}(\boldsymbol{r}). \tag{6.4}$$

The a_x are free parameters, being piecewise constant in the viscous range, the inertial range, and on integral scales, denoted as a_{VSR}, a_{ISR}, and a_{INT}, respectively. This is due to the lack of knowledge about the time scale of the acceleration correlation τ_a in (5.11).

Since P_E is only partly known, we choose the following drift term:

$$a_i = -(1 + a_x)\tilde{\gamma}_{ij} v_j. \tag{6.5}$$

Note that the memory-induced part of the drift would only be exact if P_E was Gaussian. Nevertheless, (6.5) is consistent with the constraints (3.8) and (3.4) to second order. Therefore, on subintegral scales our model reads

$$\mathrm{d}v_i(t) = -(1 + a_x)\tilde{\gamma}_{ij} v_j \mathrm{d}t + (2a_x)^{1/2} \tilde{\Gamma}_{ij}^{1/2} \mathrm{d}W_j \tag{6.6}$$

and essentially coincides with the model of Pedrizzetti & Novikov (1994) in the inertial range, but with a different choice of model parameters, satisfying the first

two constraints of their moment hierarchy on all scales. The '1' is omitted on integral scales. On inertial and integral scales the force becomes the usual isotropic white noise $(2C_0\epsilon)^{1/2}\mathrm{d}W_i$, which fixes $a_{\mathrm{ISR}} = a_{\mathrm{INT}} = \frac{3}{2}C_0$ ((3.9), cf. § 7). Only the viscous-range parameter a_{VSR} will be varied subsequently. It determines the decorrelation of velocity differences on viscous scales where the Markov assumption is likely to fail. We hope that a suitable choice of a_{VSR} can partly compensate for the unphysical Markov assumption in the viscous range.

The solution of the stochastic model in the Markov approximation can be computed efficiently provided we know how to handle the multiplicative noise, i.e. the force depending on the stochastic process itself. In order to find a stochastic force with correlation $\boldsymbol{M}(\boldsymbol{r})$, we employ the decomposition property of isotropic matrices $\boldsymbol{M} = M_{NN}\boldsymbol{P} + M_{LL}\boldsymbol{N}$, where $P_{ij} = \delta_{ij} - n_i n_j$ and $N_{ij} = n_i n_j$. Because $\boldsymbol{PN} = \boldsymbol{NP} = 0$ holds, we can easily take the square root of $\boldsymbol{M}$ or for any power $\alpha = j/2$ where j is an integer $\boldsymbol{M}^{\alpha} = M_{NN}^{\alpha}\boldsymbol{P} + M_{LL}^{\alpha}\boldsymbol{M}$. A Gaussian white-noise with correlation $\langle \mathrm{d}f_i \mathrm{d}f_j \rangle = M_{ij}\mathrm{d}t$ can hence be generated as

$$\mathrm{d}f_i = \left(M_{NN}^{1/2}P_{ij} + M_{LL}^{1/2}N_{ij}\right)\mathrm{d}t^{1/2}\xi_j, \tag{6.7}$$

where the ξ_i are independent Gaussian random variables with variance one and mean zero.

The forward Euler method is used for numerical time stepping of the stochastic differential equations. Since we intend to integrate the model over several orders of magnitude in time, we choose a variable step size comparable to the one of Thomson (1990). Note that the Ito–Stratonovich dilemma does not enter here because the stochastic forces in the velocity equation depend only on the separation and not on the velocity vector itself. The so-called spurious drift vanishes.

We compare the statistics of our model to the two-particle DNS data of Yeung (1994) at the moderate Reynolds number of $Re_\lambda = 90$ by investigating similar dispersion statistics. In order to determine the model parameter a_{INT} we match the long-time behaviour of the Markov model to the well-known Taylor result for times much larger than the Lagrangian integral time scale $t \gg T_L$:

$$\langle (\boldsymbol{r}(t) - \boldsymbol{r}_0)^2 \rangle = 12u'^2 T_L t, \tag{6.8}$$

which yields $a_{\mathrm{INT}} = (3u'^2/v_\eta^2)/(T_L/\tau_\eta) = 8.0$, where we have used the DNS data $u'^2/v_\eta^2 = 23.2$ and $T_L/\tau_\eta = 8.7$. The integral length scale of the DNS is $L_1/\eta = 56.0$. All the statistics shown in the following are generated for $a_{\mathrm{ISR}} = a_{\mathrm{INT}} = 8.0$. As a first guess we choose $a_{\mathrm{VSR}} = 8$ as well, but we will see that larger values give better agreement with the velocity statistics of the DNS. Note that the Lagrangian integral time scale T_L depends on the choice of the external force employed in the DNS to drive the turbulence.

Figure 1 shows the growth of the root-mean-square (r.m.s.) separation $\langle (\boldsymbol{r}(t) - \boldsymbol{r}_0)^2 \rangle^{1/2}$ for different initial separations. Over sufficiently small times ($t \ll 10\tau_\eta$) the fluid particles move in straight lines, with no appreciable change in their relative velocity. This accounts for the linear increase in r.m.s. separation, regardless of the initial separation. The velocity variance determines the initial behaviour $\langle (\boldsymbol{r}(t) - \boldsymbol{r}_0)^2 \rangle = \langle \boldsymbol{v}^2(0) \rangle t^2$.

Note that we cannot satisfy the initial condition (3.3) accurately, since the exact form of $P_E(\boldsymbol{v}|\boldsymbol{r})$ is unknown. Here we simply give all particles the same initial separation $\boldsymbol{r}_0/3^{1/2}(1,1,1)^T$ and velocity $\boldsymbol{v}(\boldsymbol{r}_0, 0) = (D_{ii}(\boldsymbol{r}_0)/3)^{1/2}(1,1,1)^T$ which approximates the short-time behaviour of the DNS sufficiently well and indicates the good quality of

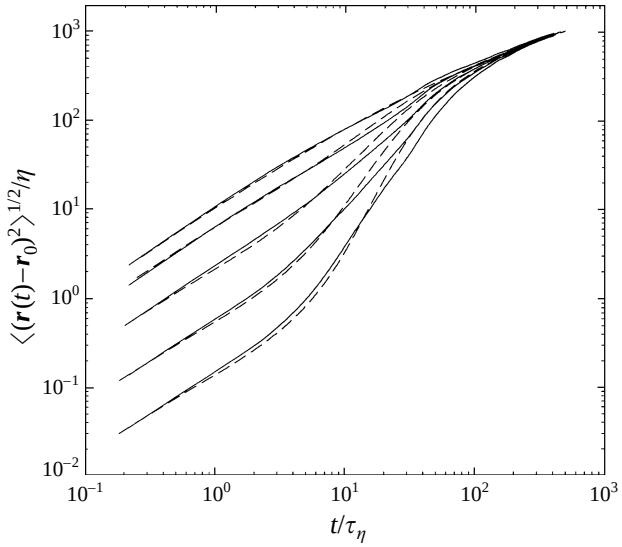

FIGURE 1. Root-mean-square separation $\langle(\boldsymbol{r}(t)-\boldsymbol{r}_0)^2\rangle^{1/2}/\eta$ versus time t/τ_η in Kolmogorov scaling for two particles initially separated by $\frac{1}{4}\eta$, η, 4η, 16η, 64η from bottom to top, respectively. Lines show DNS data, model results are dashed ($Re_\lambda = 90$, $a_{\mathrm{VSR}} = 8.0$).

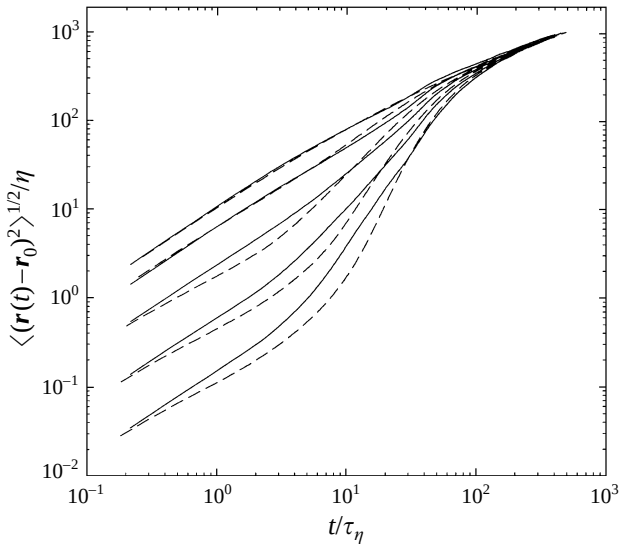

FIGURE 2. As figure 1 but for $a_{\mathrm{VSR}} = 20$.

the interpolation function (2.9). This simple choice of the initial velocities obviously violates (3.3). However, the stochastic force almost instantaneously randomizes the initial velocities such that the bias due to our unphysical initial condition decays rapidly during the first few time steps.

For small times $t \leqslant T_L$ the model gives a surprisingly accurate description of the dispersion, also as expected for very long times where the Taylor result applies. At small times one expects $\langle \boldsymbol{r}^2(t)\rangle$ to grow exponentially and accordingly $\langle(\boldsymbol{r}(t) - \boldsymbol{r}_0)^2\rangle$ should have an exponential transition region between viscous and inertial or integral scales (Batchelor 1952; Grossmann & Procaccia 1984). The model shows the

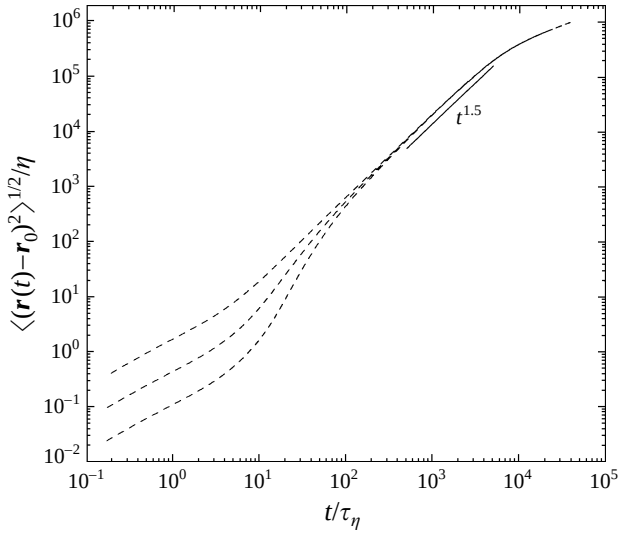

FIGURE 3. Root-mean-square separation versus time for particles initially separated by $\frac{1}{4}\eta$, η, 4η, from bottom to top respectively. Also shown is a line with slope 3/2 ($Re_\lambda = 10^4$, $a_{\mathrm{VSR}} = 8$).

a_{ISR}	G_Δ	sk(r)	fl(r)
8	0.44	1.10	4.7
12	0.31	1.20	5.0
20	0.14	1.35	5.8
30	0.07	1.45	6.3

TABLE 1. Dependence of the inertial-range values of the r.m.s. separation (i.e. the Richardson constant G_Δ), the skewness, and the flatness of the separation magnitude on the model parameter a_{ISR}.

exponential growth for small initial separations with a growth rate determined by the parameter a_{VSR}. Figure 2 presents the separation statistics for $a_{\mathrm{VSR}} = 20$: for small $\boldsymbol{r}_0$ the small-time separation decreases whereas the agreement with the DNS improves at times $t \geqslant 50\eta$. The deviations of the modelled dispersion from the DNS are mainly due to the lack of an appreciable exponential growth region in the DNS.

At intermediate times ($\tau_\eta \ll t \ll T_L$) and high Reynolds numbers one expects a Richardson law

$$\langle \boldsymbol{r}^2(t)\rangle = G_\Delta \epsilon t^3. \tag{6.9}$$

Note that there is no indication of a Richardson law at the moderate Re_λ of the DNS. The curves for small initial separations in figures 1 and 2 show a growth even stronger than $t^{3/2}$ linking viscous and integral scales. The Richardson scaling can be observed at the much higher Reynolds number $Re_\lambda = 10^4$ which is far beyond the reach of DNS and roughly of the order of atmospheric turbulence (figure 3). Remarkably, even at this very high Re_λ the $t^{3/2}$-law extends over less than two decades. The dependence of the Richardson constant G_Δ on inertial range parameter a_{ISR} is given in table 1. It varies in approximately the same range as the highly uncertain experimental value 0.06–0.45 (Tatarski 1960 gives 0.06 which seems to be the only available measurement; Malik 1991 gets a range of G_Δ values dependent on how ϵ is estimated from the

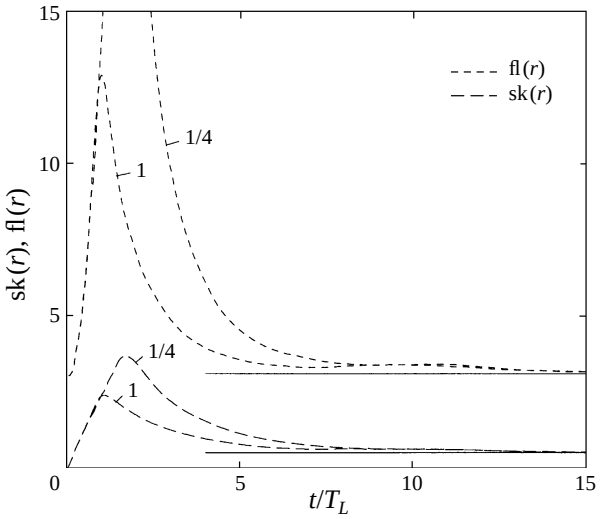

FIGURE 4. Skewness and flatness of the separation magnitude $r(t) = |\boldsymbol{r}(t)|$ versus time t/T_L in integral scaling for $r_0 \equiv |\boldsymbol{r}_0| = \frac{1}{4}\eta$ and η. For comparison the theoretical skewness (0.5) and flatness (3.1) of a χ^2-distribution are shown ($Re_\lambda = 90$, $a_{\mathrm{VSR}} = 20$).

Tatarski data). Kinematic simulations also predict a value $O(0.1)$, but so far do not include a viscous range and are computationally much more expensive than our Markovian random flight simulations (Fung *et al.* 1992; Fung & Vassilicos 1997; Elliott & Majda 1996). However, our G_Δ is smaller than that of two-point closure theories such as EDQNM ($G_\Delta \sim O(1)$). Grossmann & Procaccia (1984), who also apply the Mori–Zwanzig projector formalism but completely neglect memory effects, obtain a very large $G_\Delta = 13$. Comparison to stochastic models of the well-mixed type will be made in the next section.

As in the DNS, we find that the particle separation vectors at large times are Gaussian to a very good approximation, which is reflected by a chi-square distribution of the p.d.f. magnitude of the separation $r = |\boldsymbol{r}|$. The theoretical skewness and flatness factors equal 0.5 and 3.1, respectively and are also obtained by the model for $t \gg T_L$ (figure 4). The short-time behaviour (not shown in Yeung 1994) for small initial separations is highly non-Gaussian with the skewness as well as the flatness of r showing a large peak increasing with smaller initial separations. This high peak results from the fact that after the straight initial movement some particle pairs have drifted far apart and have large relative velocities, while most pairs are still close together having smaller relative velocities. The qualitative trend is well captured by our model; however, the peak values are very sensitive to the choice of a_{VSR} and for $a_{\mathrm{VSR}} \leqslant 20$ are somewhat underestimated. The DNS peak values for the skewness and flatness of r at $\boldsymbol{r}_0/\eta = 1/4$ are 5 and 40, whereas they are 3.6 and 25 for the model with $a_{\mathrm{VSR}} = 20$, respectively. In the inertial range the p.d.f. of r retains its non-Gaussian form, however less pronounced and skewness $\mathrm{sk}(r)$ and flatness $\mathrm{fl}(r)$ are constant and independent of the initial separation. Numerical values are listed in table 1.

The statistics of the relative velocity field determines the rate at which two fluid particles separate from each other depending on the Eulerian spatial correlation as a function of the particle separation. Yeung investigates the particle–pair velocity correlation $\rho(u_1^{(1)}, u_1^{(2)}) = \langle u_1^{(1)} u_1^{(2)} \rangle / u'^2$ where $\boldsymbol{u}^{(1)} = \boldsymbol{u}(t; \boldsymbol{x}_0)$ and $\boldsymbol{u}^{(2)} = \boldsymbol{u}(t; \boldsymbol{x}_0 + \boldsymbol{r}_0)$

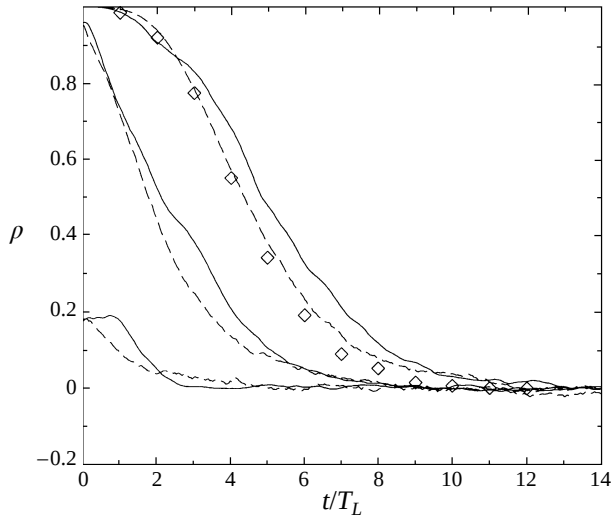

FIGURE 5. Two-particle velocity correlation $\rho \approx 1 - \langle \boldsymbol{v}^2(t) \rangle / 6u'^2$ versus time. The initial separations are $\frac{1}{4}\eta$, 4η, and 64η, from top to bottom respectively. DNS data at $Re_\lambda = 90$ are full lines, diamonds show DNS data for $r_0 = \frac{1}{4}\eta$ at $Re_\lambda = 140$, and model data are dashed ($a_{\mathrm{VSR}} = 20$).

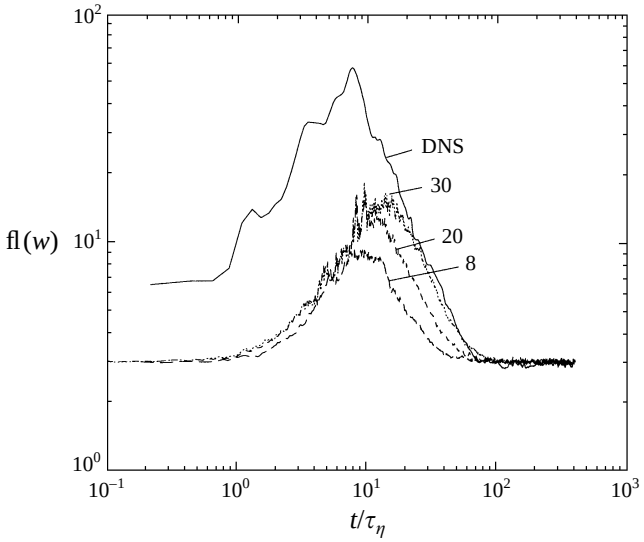

FIGURE 6. The flatness of one component w of the relative velocity versus time for $r_0 = \frac{1}{4}\eta$ and different viscous-range parameters ($a_{\mathrm{VSR}} = 8$, 20, and 30, $Re_\lambda = 90$).

(Yeung 1994). Owing to homogeneity the one-particle velocity variance equals u'^2 and is independent of the initial position and hence $\rho(u_1^{(1)}, u_1^{(2)}) = 1 - \langle \boldsymbol{v}^2(t) \rangle / 6u'^2$. Figure 5 shows the correlation coefficient for several initial separations. The modelled velocities de-correlate quicker than those of the DNS; however, especially compared to the DNS at the higher $Re_\lambda = 140$ (Yeung 1997) the overall agreement is satisfactory.

The evolution of the flatness of the relative velocity is shown in figure 6. At large times the flatness approaches the Gaussian value of 3, corresponding to the difference of two Gaussian-distributed-single particle velocities in homogeneous turbulence.

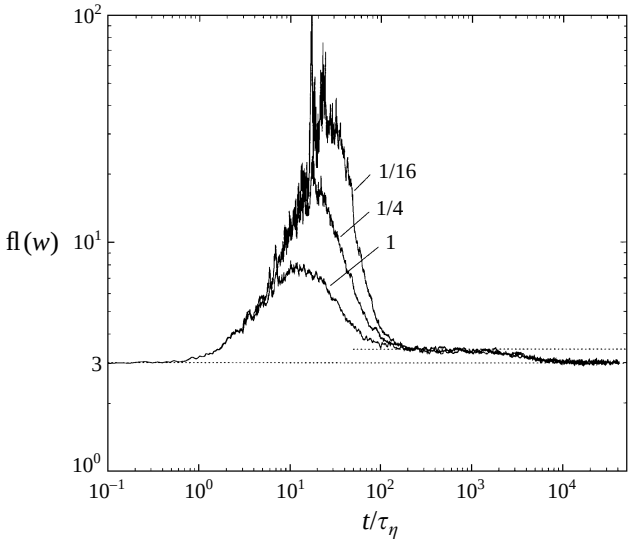

FIGURE 7. The flatness of the relative velocity component w versus time for particles initially separated by $\frac{1}{16}\eta$, $\frac{1}{4}\eta$, η. Lines indicate the Gaussian values 3 and 3.4 ($Re_\lambda = 10^4$, $a_{\mathrm{VSR}} = 20$).

a_{ISR}	fl(w)	sk(V)	fl(V)
8	3.40	0.35	3.35
12	3.45	0.30	3.45
20	3.50	0.25	3.50
30	3.50	0.20	3.50

TABLE 2. Dependence of the inertial-range values of the flatness of one velocity component w and the skewness and flatness of the longitudinal velocity difference V on a_{ISR}.

For very small times ($t \leqslant \tau_\eta$) and small initial separations the relative velocity is proportional to the velocity gradient which is intermittent and non-Gaussian. This is not reflected by our model which does not take intermittency into account and hence predicts the Gaussian value of 3 initially. It now becomes clear that our somewhat artificial delta-distribution of the initial velocities has little effect even on the short-time behaviour, which is determined by the distribution of the stochastic force. Note that there is considerable statistical uncertainty in the DNS data. At intermediate times (on a logarithmic scale) and for small $\boldsymbol{r}_0/\eta$, the flatness passes through a very high non-Gaussian peak corresponding to the one found for the separation flatness. In contrast to the flatness of the separation r, the large values of the velocity flatness (fl(w) $\geqslant$ 50 in the DNS at $r/\eta = 1/4$) do not depend crucially on a_{VSR} (figure 6). Flatness values of that order are obtained by our model only for much smaller initial separations ($\boldsymbol{r}_0/\eta = 1/16$, figure 7).

Clearly, the high flatness values are a property of the transition region between the viscous and the inertial range. The flatness in the inertial range (≈ 3.4) as predicted by the model is only slightly larger than the Gaussian value of 3 which again is obtained in the large-time limit (figure 7, table 2). Physical particles initially close to each other remain so until they are swept apart by different eddies of much larger size which rapidly increase their separation and highly distort the p.d.f. of relative

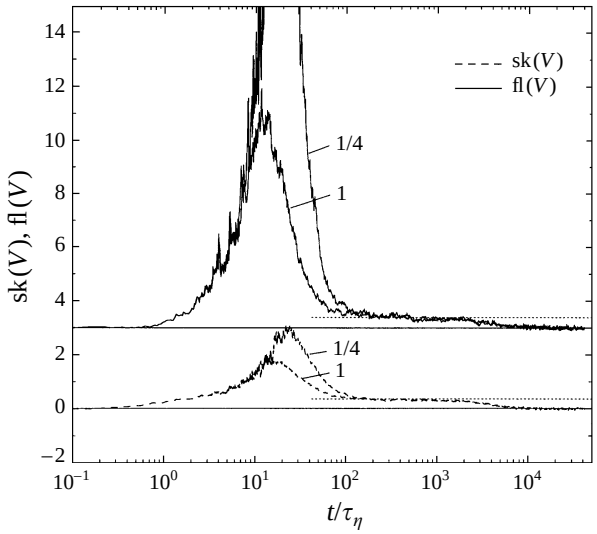

FIGURE 8. Skewness and flatness of longitudinal velocity differences $V \equiv \boldsymbol{v}\cdot\boldsymbol{r}/r$ versus time for particles initially separated by $\frac{1}{4}\eta$ and η. Lines indicate the numerical values 0, 0.35, 3, and 3.35, respectively ($Re_\lambda = 10^4$, $a_{\mathrm{VSR}} = 20$).

separations and velocities. In the inertial range, however, particles are separated by eddies of the size of their separation resulting in velocity p.d.f.s much closer to Gaussian.

We believe that non-Markovian effects of the velocity histories are responsible for the large deviation of the short-time dispersion behaviour from Gaussianity. Kinematic simulation which creates non-random trajectories gives a quantitatively better agreement for the Lagrangian velocity flatness (N. Malik and J. C. Vassilicos, private communication).

In table 2 we also give the inertial range values of the skewness and flatness of the longitudinal relative velocity $\mathrm{d}r/\mathrm{d}t = \boldsymbol{v}\cdot\boldsymbol{r}/r \equiv V$. We point out that the modelled skewness of V is always positive opposite to the Eulerian velocity differences, but in accordance to DNS data (P. K. Yeung, private communication, figure 8). Again, peak values are likely to be underestimated but the qualitative behaviour should be similar.

Figure 9 displays the variance $\langle \Delta v_i \Delta v_i \rangle$ of the relative velocity increments $\Delta\boldsymbol{v} = \boldsymbol{v}(t) - \boldsymbol{v}(0)$ which for $\boldsymbol{r}_0 \gg L_1$ equals the one-particle Lagrangian structure function $2\langle \Delta u_i \Delta u_i \rangle \equiv 6D(t)$. Whereas the long-time behaviour of the Markov model coincides with that of the DNS, the short-time behaviour of the Markov model proportional to t is qualitatively in error with the physical one of the DNS (proportional to t^2). We also show the non-Markovian one-particle model of Sawford (1991), which has the correct quadratic short-time behaviour (see the discussion in § 7).

Finally, we investigate the probability of alignment between the separation $\boldsymbol{r}(t)$ and velocity vector $\boldsymbol{v}(t)$, which determines whether particle pairs are instantaneously moving apart or coming closer to each other. An acute alignment angle θ means the particles are moving apart $\cos\theta = V/|\boldsymbol{v}| > 0$. The probability of acute alignment $P_\theta = P(0 < \theta < 90^\circ)$ for $\boldsymbol{r}_0 = \frac{1}{4}\eta$ is shown in figure 10. A preferred alignment begins to develop as the particles separate from each other, particularly strong at small times

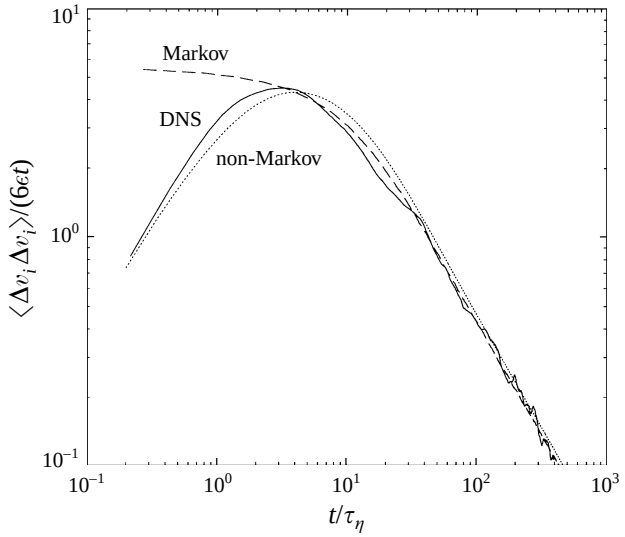

FIGURE 9. Variances of relative velocity increments $\langle \Delta v_i \Delta v_i \rangle / (6\epsilon t)$ versus t/τ_η for particles initially separated by $r_0/\eta = 64$ ($Re_\lambda = 90, a_{\mathrm{VSR}} = 8$) for the Markovian model, the non-Markovian stochastic model of Sawford (1991), and the DNS, respectively.

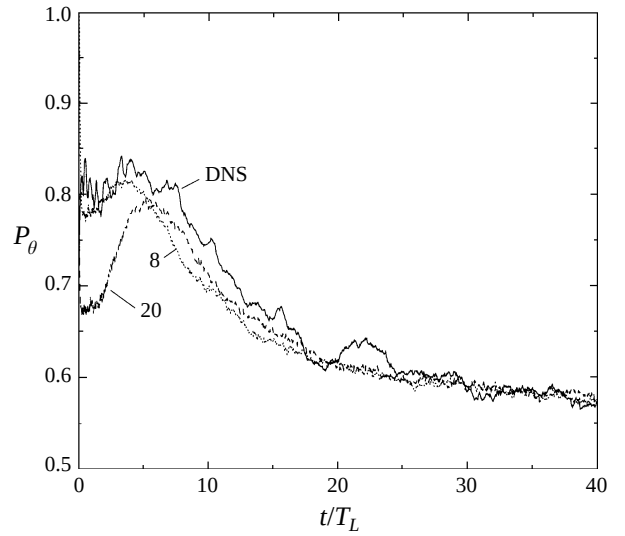

FIGURE 10. Probability of acute alignment between separation and relative velocity vectors $P_\theta = P(0 < \theta < 90^\circ)$ for particles initially separated by $r_0 = \frac{1}{4}\eta$ and viscous-range parameters $a_{\mathrm{VSR}} = 8$ and 20, respectively ($Re_\lambda = 90$).

($t \leqslant 10T_L$). The very slow decrease of alignment at large times is well captured by an Ornstein–Uhlenbeck process to which the model reduces at large times.

The rate of increase of r.m.s. separation is given by $\frac{1}{2}\mathrm{d}\langle \boldsymbol{r}^2 \rangle / \mathrm{d}t = \langle rV \rangle$. The correlation coefficient $\rho(r, V) = \langle rV \rangle / (\langle r \rangle \langle V \rangle) - 1$ is displayed in figure 11 at a high Re_λ. The peak value for short times and $r/\eta = 1/4$ is 2.8 close to the DNS value of 3.2. The long-time asymptotic value of 0.18 is within a few percent of that of the DNS, figure 11, see also table 3).

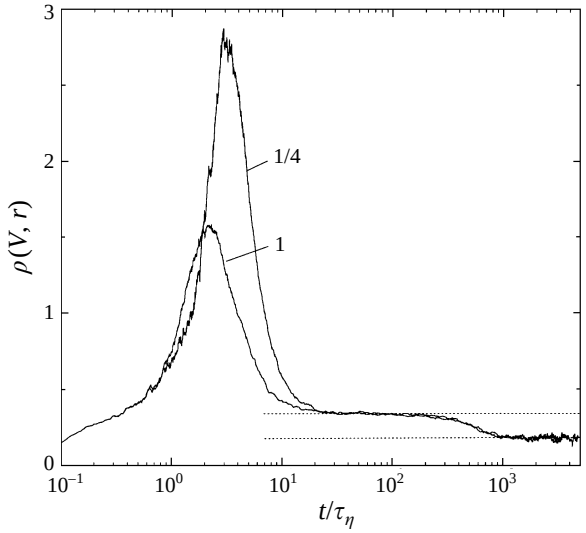

FIGURE 11. Evolution of the correlation coefficient $\rho(V,r)$ for initial separations $\frac{1}{4}\eta$ and η. Lines indicate the numerical values 0.18 and 0.34, respectively ($Re_\lambda = 10^4$, $a_{\mathrm{VSR}} = 20$).

a_{ISR}	P_θ	$\rho(V,r)$
8	0.80	0.34
12	0.75	0.37
20	0.68	0.43
30	0.63	0.49

TABLE 3. Dependence of the inertial-range values of the probability of acute alignment $P_\theta = P(0 < \theta < 90^\circ)$ and the correlation coefficient $\rho(V,r)$ on a_{ISR}.

7. Discussion and conclusions

Stochastic modelling of Lagrangian velocities has been successfully applied to dispersion studies (Thomson 1987, 1990) and more recently by Pedrizzetti & Novikov (1994) to modelling the Eulerian p.d.f. of velocity increments. There are two main results of this work. The first provides a formal background to these models in the form of an exact nonlinear generalized Langevin equation (4.24). The well-known Mori–Zwanzig projector formalism is adapted to the dynamics of Lagrangian velocity differences and applied to the Navier–Stokes equations. The inherent difficulty of the projector method in obtaining information about the random force makes it impossible to derive mathematical rigorous results. Nevertheless, we demonstrate the usefulness of our theoretical approach for the construction of stochastic dispersion models clarifying the assumptions underlying a stochastic description of turbulent dispersion. We have introduced the generalized Langevin equation as a suitable structure on which one can impose further approximations such as the Markov assumption and stochastic estimation of the model coefficients.

Secondly, we compare the modelled dispersion behaviour to DNS. For that to be feasible one has to include the viscous scales. We show that merely adjusting the second-order structure function does not adequately change the Lagrangian decorrelation time. Surprisingly, our stochastic model gives a robust qualitative picture

of small-scale moderate Reynolds number dispersion despite the obviously unphysical Markov assumption on viscous scales. In contrast to DNS, the model readily extends to high Reynolds number inertial-range dispersion where the Markov approximation is probably more reliable. We find further support for a Richardson constant of the order 0.1 previously found in experiments and kinematical simulations.

How does the inertial-range dispersion compare to the well-mixed models? In the long-time limit $t \gg T_L$ the two particles become statistically independent of each other and all models reduce to the well-known one-particle Langevin equation for one velocity component $U(t)$:

$$\mathrm{d}U(t) = -\frac{1}{T_L}U(t)\,\mathrm{d}t + \left(\frac{2u'^2}{T_L}\right)^{1/2}\mathrm{d}W(t). \tag{7.1}$$

If one accepts (7.1) as a high Reynolds number model, T_L is related to the Lagrangian Kolmogorov constant C_0, the inertial-range coefficient of the Lagrangian structure function $D(t) = C_0\epsilon t$ (see e.g. Pope 1994: $T_L^{-1} = C_0\epsilon/2u'^2$). There is great uncertainty about the numerical value of C_0. Estimates from DNS and experiments vary in the range $C_0 = 2$–7 possibly due to a strong Reynolds number dependence (Pope 1994; Du *et al.* 1995). Using similar inertial-range arguments the amplitude of the stochastic force of the well-mixed models is usually taken as $(2C_0\epsilon)^{1/2}$ which implies in our notation $a_{\mathrm{ISR}} = a_{\mathrm{INT}} = \frac{3}{2}C_0$. From our consideration in § 5 there is little *a priori* reason why the amplitude of the stochastic force should be the same in the inertial range and on integral scales, i.e. a_{ISR} might be different from a_{INT}, especially if there is an external force acting on integral scales only. Clearly, the larger C_0 or a_{ISR} the more G_Δ decreases and the smaller the relevance of the conditional acceleration becomes.

For the same C_0 our G_Δ is at the lower end of the Richardson coefficients predicted by different well-mixed models ($C_0 \approx 6$: $G_\Delta \approx 0.6$–1.2, Borgas & Sawford 1994). Note that stochastic models based on the moments approximation approach in general do not satisfy the well-mixed condition as discussed for the one-particle case by Du *et al.* (1994). If the well-mixed condition is violated a uniform Eulerian distribution is no longer guaranteed, i.e. $\int \mathrm{d}\boldsymbol{v} P_E(\boldsymbol{v}|\boldsymbol{r})$ might not equal unity, although this is difficult to check numerically. Furthermore, satisfying the second-moment constraint does not guarantee that P_E actually possesses a skewness given by the Kolmogorov structure equation. Errors in the higher moments can propagate down to the third or second order due to the coupling of moments of different orders in the hierarchy implied by the continuity equation (4.25), (D. J. Thomson, private communication). In principle, the models can be improved by taking intermittency and non-Markovian effects into account.

7.1. *Intermittency and conditional averages*

The generalized Langevin equation reformulates the entire Navier–Stokes dynamics and hence includes intermittency effects, too. However, here we do not allow for any intermittency corrections of Eulerian statistics appearing in the approximate model coefficients. This can be justified by noting that only low-order structure functions are included for which those intermittency corrections are known to be small (Frisch 1995). However, as mentioned above, small-scale velocity statistics strongly deviate from Gaussianity such that one can at best expect qualitative predictions from a simple 'non-intermittent' model. The inclusion of intermittency effects would necessarily introduce further arbitrariness to the model.

Obviously, a better knowledge of the Eulerian flow statistics, i.e. $P_E(\boldsymbol{v}|\boldsymbol{r})$, can

immediately be incorporated in the memory-induced part of the systematic drift term $M_{ij}\partial_{v_j}\log P_E$ in (5.19). On the other hand, it seems to be practically impossible to relax the Gaussian assumption on the distribution of the random force $\boldsymbol{f}(t)$.

Even for given stationary Eulerian statistics the separation of two fluid particles is a non-stationary process, since the particles tend to separate in time. As discussed by Thomson (1996), neither the well-mixed models together with the assumption of a Gaussian Eulerian p.d.f. nor the EDQNM closure is capable of accounting for the lack of time reversal asymmetry in turbulent dispersion. We find that the time reversal asymmetry is related to the acceleration conditionally averaged for fixed velocity increments $\boldsymbol{A}$. Extending ideas of GT82, stochastic estimation relates $\boldsymbol{A}$ to the skewness of velocity increments and hence to the energy dissipation in the inertial range via the Kolmogorov structure equation independent of any intermittency corrections.

Approximations of conditional averages in terms of low-order polynomials have received a great deal of attention recently, especially in the context of the turbulent mixing of a passive scalar (Pope & Ching 1993; Kraichnan, Yakhot & Chen 1995, Appendix B). Although the ansatz (5.1) has the form of a Taylor expansion about the given data (here $\boldsymbol{v}$), the linear approximation as the lowest non-trivial contribution (to the conditional scalar dissipation in the case of scalar mixing) gives surprisingly accurate results even for larger values of the given data (Pope & Ching 1993; Kraichnan *et al.* 1995). This is partly due to the fact that the linear mean-square estimate is exact if (in our case) $L\boldsymbol{v}$ and $\boldsymbol{v}$ are joint normally distributed (Adrian 1996). Moreover, the good quality of the linear approximation also results from the potential of the expansion in orthogonal polynomials of the delta-function in the conditional average to be *uniformly* convergent (Kraichnan 1970). Of course, $L\boldsymbol{v}$ and $\boldsymbol{v}$ considered here cannot be joint Gaussian since the skewness of $\boldsymbol{v}$ is non-vanishing and given by the Kolmogorov structure equation.

The extension of low-order stochastic estimation to the turbulent velocities suggested in this work requires further numerical and experimental investigations. It will be very interesting to see whether a simple polynomial expression such as the linear (5.4) or the quadratic (5.6) approximation is capable of describing the conditional acceleration.

7.2. *Non-Markovian effects*

The short-time dispersion behaviour of the model can be improved by allowing for non-Markovian effects. Whereas this seems to be difficult and computationally expensive for the two-particle case, it is instructive to look at non-Markovian corrections in the one-particle or large-separation limit $r \to \infty$.

The simplest generalization to a delta-correlated random force or memory is an exponentially decaying one

$$\langle f_i(t)f_j(0)|\boldsymbol{v}\rangle \approx K_{LL}(r)\mathrm{e}^{-\tilde{\gamma}_{LL}(r)t}n_in_j + K_{NN}(r)\mathrm{e}^{-\tilde{\gamma}_{NN}(r)t}(\delta_{ij}-n_in_j), \tag{7.2}$$

where $K_{ij}(\boldsymbol{r}) = \langle Lv_i(\boldsymbol{r})Lv_j(\boldsymbol{r})\rangle$ denotes the memory amplitude and $\tilde{\gamma}_*$ the memory damping. In the large-separation limit the drift $\langle L\boldsymbol{v}_0|\boldsymbol{v}(\boldsymbol{r})\rangle$ vanishes and all matrices become isotropic and r-independent, e.g. $K_{ij} \to ku'^2\delta_{ij}$, where $k = k_* = a'^2/u'^2$ and a'^2 is the acceleration variance $\frac{1}{3}(\langle(\nabla p)^2\rangle + \nu^2\langle(\nabla^2\boldsymbol{u})^2\rangle)$.

Considering only one velocity component and assuming the Eulerian p.d.f. to be

Gaussian, the generalized Langevin equation (4.24) reads

$$\frac{\mathrm{d}}{\mathrm{d}t}U(t) = -\int_0^t k\mathrm{e}^{-\tilde{\gamma}(t-\tau)}U(\tau)\,\mathrm{d}\tau + f(t), \tag{7.3}$$

where $f(t)$ is a Gaussian random force correlated as

$$\langle f(t)f(0)\rangle = ku'^2\mathrm{e}^{-\tilde{\gamma}t}. \tag{7.4}$$

It is readily shown (by Laplace transformation) that the non-Markovian model (7.3) corresponds to an asymptotically stationary white-noise model for the Lagrangian acceleration $A(t) = \dot{U}(t)$ which is exactly the second-order autoregressive stochastic model of Sawford (1991) (see also Pope 1994; Heinz 1997):

$$\mathrm{d}A(t) + \alpha_1 A(t)\,\mathrm{d}t + \alpha_2\int_0^t A(t')\,\mathrm{d}t'\,\mathrm{d}t = (2\alpha_1\alpha_2 u'^2)^{1/2}\,\mathrm{d}W(t), \tag{7.5}$$

where $\alpha_1 = \tilde{\gamma}$ and $\alpha_2 = k$. For an appropriately chosen memory damping $\tilde{\gamma}$ the model shows a remarkable agreement with the DNS of Yeung & Pope (1989). The physical inconsistency of modelling the acceleration as a Markov process can be removed by choosing a memory function with vanishing time derivatives at the origin.

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Appendix A

We derive the continuity equation of the Eulerian p.d.f. $P(\boldsymbol{v}|\boldsymbol{r}_0)$ (4.25) from

$$L\delta(\boldsymbol{v}-\boldsymbol{v}_0) = -\{Lv_{0j}\}\partial_{v_j}\delta(\boldsymbol{v}-\boldsymbol{v}_0) \tag{A 1}$$

and by using (4.12)

$$\begin{aligned} PL\delta(\boldsymbol{v}-\boldsymbol{v}_0) &= \int \mathrm{d}\boldsymbol{v}'\frac{\langle L\delta(\boldsymbol{v}-\boldsymbol{v}_0)\delta(\boldsymbol{v}'-\boldsymbol{v}_0)\rangle}{P_E(\boldsymbol{v}'|\boldsymbol{r}_0)}\delta(\boldsymbol{v}'-\boldsymbol{v}_0) \\ &= -\partial_{v_j}\Big(\langle Lv_{0j}|\boldsymbol{v}\rangle\delta(\boldsymbol{v}-\boldsymbol{v}_0)\Big). \end{aligned} \tag{A 2}$$

Applying the skewness formula (4.8) to (A 2) yields

$$\begin{aligned} &-\partial_{v_j}\Big(\langle Lv_{0j}|\boldsymbol{v}\rangle\delta(\boldsymbol{v}-\boldsymbol{v}_0)\Big) \\ &\quad= \int\Big(-\mathrm{d}\boldsymbol{v}'\frac{\langle\delta(\boldsymbol{v}-\boldsymbol{v}_0)L\delta(\boldsymbol{v}'-\boldsymbol{v}_0)\rangle}{P_E(\boldsymbol{v}'|\boldsymbol{r}_0)} + \frac{v_m\partial_{r_m}\langle\delta(\boldsymbol{v}-\boldsymbol{v}_0)\delta(\boldsymbol{v}'-\boldsymbol{v}_0)\rangle}{P_E(\boldsymbol{v}'|\boldsymbol{r}_0)}\Big)\delta(\boldsymbol{v}'-\boldsymbol{v}_0) \\ &\quad= -\langle\delta(\boldsymbol{v}-\boldsymbol{v}_0)Lv_{0j}\rangle\partial_{v_j}\frac{\delta(\boldsymbol{v}-\boldsymbol{v}_0)}{P_E(\boldsymbol{v}|\boldsymbol{r}_0)} + \frac{v_m\partial_{r_m}P_E(\boldsymbol{v}|\boldsymbol{r}_0)}{P_E(\boldsymbol{v}|\boldsymbol{r}_0)}\delta(\boldsymbol{v}-\boldsymbol{v}_0), \end{aligned} \tag{A 3}$$

where again we used (4.12), (A 1) and partial integration. Ensemble averaging then leads to the continuity equation for the Eulerian p.d.f. P_E (4.25).

The alternative formulation of the memory kernel (4.22) can be obtained from the

skewness formula (4.8) if the turbulence is stationary and homogeneous:

$$\begin{aligned}\langle L\boldsymbol{f}(\tau)|\boldsymbol{v}\rangle P_E(\boldsymbol{v}|\boldsymbol{r}_0) &= \langle L\boldsymbol{f}(\tau)\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle \\ &= -\langle \boldsymbol{f}(\tau)L\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle + v_m\partial_{r_m}\langle \boldsymbol{f}(\tau)\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle, \end{aligned} \tag{A 4}$$

where the second term vanishes because of $\boldsymbol{f}(\tau) = Q\boldsymbol{f}(\tau)$ and $Q\delta(\boldsymbol{v}-\boldsymbol{v}_0) = 0$. The first term in (A 4) yields (4.22) by means of (A 1):

$$\partial_{v_j}\langle \boldsymbol{f}(\tau)QL\boldsymbol{v}_0\delta(\boldsymbol{v}-\boldsymbol{v}_0)\rangle = \partial_{v_j}\Big(\langle \boldsymbol{f}(\tau)\boldsymbol{f}(0)|\boldsymbol{v}\rangle P_E(\boldsymbol{v}|\boldsymbol{r}_0)\Big). \tag{A 5}$$

Appendix B

Here we briefly sketch how to apply our new approach to stochastic modelling to the Lagrangian concentration differences between two particles. Regardless of any memory effects we find exactly the scalar probability density closure of Kraichnan (1994), which successfully compares to DNS and seems to give a reasonable account even of higher-order scalar structure functions showing anomalous (non-Kolmogorov) scaling (Kraichnan *et al.* 1995).

We consider the concentration difference between two fluid particles initially at time $t = 0$ separated by a distance $\boldsymbol{r}_0$ denoted as

$$\Delta\phi(t) \equiv \phi(\boldsymbol{X}(t;\boldsymbol{x}_0+\boldsymbol{r}_0)) - \phi(\boldsymbol{X}(t;\boldsymbol{x}_0)), \tag{B 1}$$

which evolves by the advection–diffusion equation written here for concentration differences:

$$\frac{\mathrm{d}}{\mathrm{d}t}\Delta\phi(t) = \kappa\nabla_x^2\Delta\phi(t), \tag{B 2}$$

where κ is the molecular diffusivity and $\nabla_x^2 = \partial_{x_m}\partial_{x_m}$.

In order to make analytical progress, we have to choose the simplest model for the convection of fluid particles, namely the Markov model of § 5 in the diffusion limit (Thomson 1987; Borgas & Sawford 1994). Note that this is only for mathematical convenience: for the implementation of a Monte Carlo solution algorithm the full Markov model might give more reliable results without much additional numerical effort.

In the diffusion limit we obtain the following stochastic mixing model:

$$\mathrm{d}r_i(t) = \tfrac{1}{2}\eta(\boldsymbol{r}(t))_{kj}^{1/2}\partial_{r_k}\eta(\boldsymbol{r}(t))_{ij}^{1/2}\,\mathrm{d}t + \eta(\boldsymbol{r}(t))_{ij}^{1/2}\,\mathrm{d}W_j(t), \tag{B 3}$$

$$\mathrm{d}\Delta\phi(t) = -\gamma_\phi(r(t))\Delta\phi(t)\,\mathrm{d}t, \tag{B 4}$$

where the memory of scalar differences has been neglected, an approximation which improves with increasing Péclet number. The eddy diffusivity $\eta_*(\boldsymbol{r}) \propto \gamma_*^{-1}(\boldsymbol{r})D_*(\boldsymbol{r})$ shows the expected behaviour $\eta_* \propto r^2$ in the VSR and $\eta_* \propto r^{4/3}$ in the ISR.

The relaxation coefficient $\gamma_\phi(r)$ in the passive scalar field equation (B 4) is the linear stochastic estimate of the conditionally averaged scalar diffusion $\langle\kappa\nabla_x^2\Delta\phi_0|\Delta\phi(r)\rangle$ and is given by

$$\gamma_\phi(r) = -\frac{\langle\Delta\phi(r)L_\phi\Delta\phi(r)\rangle}{\langle(\Delta\phi(r))^2\rangle} = -\kappa\frac{\nabla_r^2 S_2(r) - \nabla_r^2 S_2(0)}{S_2(r)}, \tag{B 5}$$

where $\nabla_r^2 F(r) = \partial_{r_m}\partial_{r_m}F = 1/r^2\partial_r(r^2\partial_r F)$ due to isotropy. This can be seen by using an explicit representation of the time evolution operator L_ϕ analogously to the procedure for the evaluation of $A_i^{(1)}$ in § 5. Then, by making use of the equivalent of

the Kolmogorov structure equation for the scalar (the Yaglom equation) one arrives at (B 5).

The Fokker–Planck equation according to the model (B 3) and (B 4) can now be derived for the Lagrangian p.d.f. $P_L(\Delta\phi, \boldsymbol{r}, t|\boldsymbol{r}_0)$ of two fluid particles having a concentration difference $\Delta\phi$ and separation $\boldsymbol{r}$ given that they were separated by $\boldsymbol{r}_0$ initially. By means of the fundamental relation (cf. (3.2))

$$P_E(\Delta\phi|\boldsymbol{r}, t) = \int P_L(\Delta\phi, \boldsymbol{r}, t|\boldsymbol{r}_0, t_0)\mathrm{d}\boldsymbol{r}_0 \tag{B 6}$$

this Fokker–Planck equation is also obeyed by the Eulerian p.d.f. $P_E(\Delta\phi|\boldsymbol{r}, t)$ of the concentration difference $\Delta\phi$ at time t between two fixed points at distance $\boldsymbol{r}$ and can be written in the stationary state as

$$-(1/r^2)\partial_r\left(r^2\eta_{LL}(r)\partial_r P_E\right) = \partial_{\Delta\phi}\left(\gamma_\phi\Delta\phi P_E\right). \tag{B 7}$$

Isotropy implies that P_E depends only on $r = |\boldsymbol{r}|$. Note that (B 7) holds even if memory effects of the scalar (but not of the velocity field) are taken into account. Equation (B 7) is a special case of the exact equation

$$\partial_{r_i}\left(\langle v_i|\Delta\phi(r)\rangle P_E\right) + \partial_{\Delta\phi}\left(\langle\kappa\nabla_x^2\Delta\phi|\Delta\phi(r)\rangle P_E\right) = 0. \tag{B 8}$$

Taking the even moments of (B 7), i.e. multiplying by $(\Delta\phi)^{2n}$ and integrating over $\Delta\phi$, gives a differential equation for the even-order scalar structure functions $S_{2n}(r) = \langle(\Delta\phi)^{2n}\rangle$ yielding anomalous inertial-range scaling $S_{2n}(r) \propto r^{\zeta_{2n}}$ with $\zeta_{2n} \propto n^{1/2}$. Closed equations for the scalar structure functions are possible because (B 3) is linear in $\Delta\phi$. This is exactly the Kraichnan p.d.f. closure derived here from a Lagrangian approach. At second order the well-known Batchelor (1959) phenomenology of the scalar structure function $S_2(r)$ is recovered.

From our point of view especially the linear estimation of the conditional average is important. It has received strong support from numerical simulations (Kraichnan *et al.* 1995) and experiments (Ching *et al.* 1996).

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