Lagrangian views on turbulent mixing of passive scalars

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The Lagrangian view of passive scalar turbulence has recently produced interesting results and interpretations. Innovations in theory, experiments, simulations and data analysis of Lagrangian turbulence are reviewed here in brief. Part of the review is closely related to the so-called Kraichnan model for the advection of the passive scalar in synthetic turbulence. Possible implications for a better understanding of the passive scalar mixing in Navier-Stokes turbulence are also discussed.

1. Introduction

Eulerian and Lagrangian points of view on the motion of fluids, and on the mixing of scalars, are equivalent (Prandtl & Tietjens 1934) but cannot be related to each other in analytically tractable ways except in a few special instances. The bridge between the two descriptions is formally obvious, at least for a conserved scalar. A batch of a massless tracer particle which is advected in an incompressible turbulent flow $\mathbf{u}$, subject to the diffusion κ, follows the Langevin equation (Gardiner 2004) given by

$$\dot{\mathbf{x}} = \mathbf{u}(\mathbf{x}(t), t) + \sqrt{2\kappa}\, \boldsymbol{\chi}(t)\,, \tag{1.1}$$

where $\boldsymbol{\chi}(t)$ is vectorial white noise that is statistically independent in each of its three components. This equation is complementary to a Fokker-Planck equation for $\theta(\mathbf{x}, t)$, the probability density function (PDF) of the tracer at time t at position $\mathbf{x}$. The equation

$$\frac{\partial \theta}{\partial t} + (\mathbf{u} \cdot \nabla)\theta = \kappa \nabla^2 \theta \tag{1.2}$$

is exactly the advection-diffusion equation for a scalar field in the Eulerian frame. The scalar diffusivity is denoted by κ. Its ratio to the kinematic viscosity ν of the fluid is the Schmidt number

$$Sc = \frac{\nu}{\kappa}\,, \tag{1.3}$$

or, for an advected temperature field, the Prandtl number, Pr.

The scalar mixing process at a given Reynolds number is significantly different in the Kolmogorov-Obukhov-Corrsin regime given by $Sc \leq 1$ (Kolmogorov 1941, Obukhov 1949, Corrsin 1951) from that in the Batchelor regime given by $Sc \gg 1$

(Batchelor 1959). While the scalar is advected by the turbulent inertial range in the former, it is mainly advected in the latter by smooth velocity structures of the viscous range of turbulence.

In turbulent mixing of passive scalars, and in turbulence in general, Eulerian and Lagrangian views have their specialized places. For example, the former appears better suited for studying problems such as nonpremixed combustion in reacting flows (Linãn & Williams 1993, Peters 2000). Indeed, the comparative ease of making Eulerian measurements renders them more appealing in a number of instances. If, on the other hand, one has to calculate the dispersion of θ arising from a few localized sources, the Lagrangian view is more appropriate (Sawford 2001). Examples of such applications include fumigation (Sawford *et al.* 1998), the spread of buoyant plumes (Heinz & van Dop 1999), and the surface motion of buoys over the sea (Maurizi *et al.* 2004). Lagrangian studies are being recognized as increasingly important in geophysics, e.g. for the mixing of plankton and other biomatter in the upper ocean (Seuront & Schmitt 2004) and for droplet dynamics in cumulus clouds (Vaillancourt *et al.* 2002).

In spite of their wide-spread utility, Lagrangian data are harder to obtain experimentally and are more expensive to compute. For example, it is unclear if we can ever measure with satisfactory accuracy high-order moments of the particle acceleration along a trajectory in a turbulent flow of moderately high Reynolds number (Yakhot & Sreenivasan 2005); see also discussion of Fig. 1 in Sec. 3. The relevant point here is that measuring (as an example) the fourth moment of the Lagrangian acceleration is equivalent to measuring the twelfth moment of Eulerian velocity differences (which has not been obtained so far with impeccable accuracy). The basis for this connection is the relation for the Lagrangian acceleration expressed in terms of Eulerian velocity increments,

$$a \approx \frac{(\delta_\eta u)^2}{\eta} \approx \frac{(\delta_\eta u)^3}{\nu} \, , \qquad (1.4)$$

where η is the fluctuating small scale around the Kolmogorov length η_K and the combination $\eta \times \delta_\eta u/\nu = 1$, relating the velocity increment $\delta_\eta u$ on the length scale η to η. The difficulties are compounded by the tendency of most conventional Lagrangian tracers to cluster near solid walls, which produces a large mismatch of information between fluid particles and tracers. Large shear will have similar effects. Thus, one should ask if the additional complexities associated with Lagrangian calculations and measurements justify the investment and effort, leaving aside their relative novelty for the present: When is the Lagrangian perspective the method of choice? What makes it advantageous? What have we learnt in the big picture?

New insights into these questions, especially on the scalar mixing problem, have become possible for the so-called Kraichnan model (Kraichnan 1968, 1994). For a review with an extensive list of references, see Falkovich *et al.* (2001). The Kraichnan model has been applied to diverse physical circumstances such as the mixing of passive scalars and their dynamics in compressible turbulence (Gawędzki & Vergassola 2000, Bec *et al.* 2004). In a further development, the claim has been made (with certain careful caveats) that the anomalous scaling of active scalars can be understood in terms of passive scalar fields (Ching *et al.* 2003).

All these developments deserve some comment. This is one of our purposes here. Secondly, we will comment on these theoretical achievements in the context

of numerical and experimental work on the mixing of the passive scalar in Navier-Stokes turbulence. In the process, we draw attention to a few open questions that should be addressed in the future. We pay particular attention to the Kraichnan model and its relation to homogeneous turbulence. Clearly, there are many other aspects related to the turbulent mixing which cannot be covered here. For studies of turbulent mixing in inhomogeneous shear layers, jets or the mixing by Rayleigh-Taylor instabilities, see, for example, the review by Dimotakis (2005) or a recent work by Cabot & Cook (2006).

The outline of the paper is as follows. We will briefly review the scalar advection in a white-in-time Kraichnan flow in the next section and consider basic ideas which are closely tied with the Lagrangian view. We then discuss numerical and experimental efforts connected to the Lagrangian picture of mixing in Navier-Stokes turbulence. In the last section, we relate these findings to passive scalar mixing in Navier-Stokes turbulence, particularly for high Schmidt numbers.

2. Scalar mixing in white-in-time turbulence

It is useful to begin with the anomalous scaling for the passive scalar obeying the Kraichnan model, in large part because some exact results are available. The crucial element of the model is that it uses for the velocity in the advection-diffusion equation a stochastic Gaussian field with a time correlation that decays infinitely rapidly (or is "white in time") and a spatial correlation that has a power law structure with a prescribed scaling exponent, $0 < \zeta < 2$. That is,

$$\langle u_i(\mathbf{x}, t) u_j(\mathbf{y}, t') \rangle = D_{ij}(\mathbf{x} - \mathbf{y}) \delta(t - t'), \tag{2.1}$$

with

$$D_{ij}(\mathbf{r}) = D_0 \delta_{ij} - d_{ij}(\mathbf{r}) = D_0 \delta_{ij} - D_1 \left((2 + \zeta) \delta_{ij} - \zeta \frac{r_i r_j}{r^2} \right) r^\zeta, \tag{2.2}$$

where $\mathbf{r} = \mathbf{x} - \mathbf{y}$, $r = |\mathbf{r}|$ and $i, j = 1, 2, 3$, and $\langle . \rangle$ denotes a suitable average; D_{ij} is a diffusivity with the dimension of $L^2 T^{-1}$, and D_0 and D_1 are constants. The case $\zeta = 0$ stands for advection in a very rough flow and $\zeta = 2$ for transport in a smooth flow (see Bernard (2000) for a compact introduction to the subject). This power-law scaling is similar to the Navier-Stokes case (though the scaling exponent ζ here assumes an arbitrary value between 0 and 2), but the temporal scaling is qualitatively different. For statistical stationarity, a random forcing $f_\theta(\mathbf{x}, t)$ has to be added to the right hand side of (1.2), with the property that

$$\langle f_\theta(\mathbf{x}, t) f_\theta(\mathbf{y}, t') \rangle = C(r/L) \delta(t - t'). \tag{2.3}$$

The function $C(r/L)$ varies only on the large scale L and decays rapidly to zero for smaller scales. Kraichnan's insight was that this model possesses the essential elements of the scalar mixing while retaining analytical tractability. Indeed, it has been possible to establish anomalous scaling (Falkovich *et al.* 2001) for this model even though the idealized advecting flow itself does not exhibit such anomaly. In other words, the scaling exponents for the Eulerian structure functions $S_n(r) = \langle (\theta(\mathbf{x} + \mathbf{r}) - \theta(\mathbf{x}))^n \rangle \sim r^{\xi_n}$ of the scalar increments differ from the classical Kolmogorov-Obukhov form (which is $\xi_n = (2 - \zeta)n/2$ for this model) and vary nonlinearly with the order of the moment.

Let us write down the evolution equation for the n-point correlator of a statistically stationary passive scalar field in the frame of Kraichnan's model. The equation is

$$M_n \langle \theta(\mathbf{x}_1) \cdots \theta(\mathbf{x}_j) \cdots \theta(\mathbf{x}_k) \cdots \theta(\mathbf{x}_n) \rangle = \frac{1}{2} \sum_{j,k=1}^{n} C(r_{jk}/L) \times$$

$$\langle \theta(\mathbf{x}_1) \cdots \theta(\mathbf{x}_{j-1}) \theta(\mathbf{x}_{j+1}) \cdots \theta(\mathbf{x}_{k-1}) \theta(\mathbf{x}_{k+1}) \cdots \theta(\mathbf{x}_n) \rangle. \quad (2.4)$$

The white-in-time character of the advecting flow yields no dependence of the n-th order scalar moment on mixed velocity-scalar moments of order $n+1$, which would be a manifestation of the well-known closure problem characteristic of mixing in Navier-Stokes turbulence. The operator M_n contains, besides the Laplacian diffusion, an additional turbulent diffusion part, d_{ij}, and can be written as

$$M_n = -\kappa \sum_{l=1}^{n} \nabla_{\mathbf{x}_l}^2 + \frac{1}{2} \sum_{l,m=1}^{n} d_{ij}(\mathbf{x}_l - \mathbf{x}_m) \nabla_{\mathbf{x}_l}^i \nabla_{\mathbf{x}_m}^j. \quad (2.5)$$

The so-called zero modes in the Kraichnan model are solutions of the homogeneous subproblem in Eq. (2.4), i.e. when the right hand side is set to zero. The universality of the scaling of $S_n(r)$ with respect to r is caused by the scaling dominance of the zero modes in comparison to particular solutions of the inhomogeneous problem (2.4). An important new insight obtained from Kraichnan's model is indeed that zero modes are the reason for the deviations from the classical scaling of passive scalar structure functions in the inertial range. We wish to stress, however, that no explicit expression for the zero modes has been calculated, and that only expansions in limiting cases for ζ and for $\kappa \to 0$ have been found (e.g., Bernard 2000, Shraiman & Siggia 2000, Arad *et al.* 2001). However, accurate numerical solutions have been obtained for the general case (Frisch *et al.* 1998, Gat *et al.* 1998, Chen & Kraichnan 1998).

The basic physical picture is this: To understand the scaling exponent ξ_3 of a third-order quantity, say, it is obvious that one needs to study the properties of objects generated from the scalar variable at three different positions in space. At any point in time, the three tracer particles at the three positions form a triangle. The triangle is described by the lengthscale R—which, for specificity, can be taken as the geometric mean of the lengths of the sides of the triangle—and two of the three angles of the triangle, say ψ and ϕ (Pumir *et al.* 2000, Celani & Vergassola 2001). As the three particles advect, the triangles change in shape and size. If we rescale the triangles to the same size at each time step, the dynamics reduces to the evolution of shapes of triangles, or to a suitable function $f(\psi, \phi)$ of the two angles ψ and ϕ. The important result obtained for the Kraichnan model is that the three-point statistics are governed by those trajectories for which the change in the length scale R is compensated by the change in shape of the triangles such that the product $R^{\xi_3} f(\psi, \phi)$ is a constant. As particles move in the Kraichnan flow, an n-particle cloud grows in size but fluctuations in the cloud shape decrease in magnitude. The latter happens because the correlation between particles—which arises because they are contained within the integral scale of the velocity field— weakens with the separation distance. Therefore, as mentioned above, one looks for suitable functions of size *and* shape that have the property of being conserved via the balance between the growth in size and the decrease of shape fluctuations.

The important qualitative lesson from this work is that certain types of Lagrangian characteristics, conserved only on the average, determine the statistical scaling of Eulerian structure functions (Falkovich & Sreenivasan 2006). This is a clear instance where the Lagrangian point of view has been essential to understanding better the Eulerian quantities in turbulence—a conclusion that may have broad validity in systems with strong fluctuations. There is therefore enough justification for the enthusiasm about the progress made.

However, there are several differences between the predictions of the Kraichnan model and the behavior of a passive scalar in Navier-Stokes turbulence. A partial list now follows:

(i) Before we compare experiments and simulations in Navier-Stokes turbulence with the Kraichnan model, we should consider the meaning of the Schmidt number Sc in the model. This is not obvious because the advecting flow has no timescale for comparison with the diffusion time.

In Navier-Stokes turbulence the smallest length is commonly thought to be the Kolmogorov scale η_K and the corresponding timescale to be $\tau_\eta = \eta_K^2/\nu$. For $Sc \gg 1$, the passive scalar contains scales smaller than η_K, which are acted upon by the strain-field in the sub-Kolmogorov range, which is the same as that imposed at η_K. Thus, the diffusive time scale $\tau_D = \eta_B^2/\kappa$, defined by means of the Batchelor diffusion length η_B, is the same as τ_η, leading to the formula $Sc = \eta_K^2/\eta_B^2$. Formally, one can define a "Kolmogorov scale", $\tilde{\eta}_K = (\nu/D_1)^{1/\zeta}$, and a "Batchelor scale", $\tilde{\eta}_B = (\kappa/D_1)^{1/\zeta}$, such that

$$\tilde{Sc} = \left(\frac{\tilde{\eta}_K}{\tilde{\eta}_B}\right)^\zeta, \tag{2.6}$$

can be regarded as the generalized Schmidt number (E & vanden-Eijnden 2001). Here, D_1 is the constant in (2.2). This definition coincides with the classical definition for $\zeta = 2$. We recall, however, that the Kraichnan model discusses the advection of a scalar of fixed diffusivity in a prescribed synthetic flow. For the latter, no knowledge of viscosity is necessary. Therefore, despite the ingenuity of the above argument, a proper Schmidt number does not arise as a physical dimensionless parameter in this setting, in contrast to the Navier-Stokes case.

(ii) For the case of decaying turbulence behind heated grids (for which the velocity and scalar fields are both nearly homogeneous and isotropic, and $Sc = O(1)$), it is almost certainly true that the decaying scalar assumes a self-similar form (i.e., the PDF reaches a self-similar state). It is almost certainly Gaussian (e.g., Sreenivasan *et al.* 1980), whereas all indications are that they attain exponential tails for the Kraichnan model (Balkovsky & Fouxon 1999).

(iii) The PDF of the scalar field in a statistically stationary Navier-Stokes flow field with stochastic scalar driving is almost certainly Gaussian or sub-Gaussian (Mydlarski & Warhaft 1998, Watanabe & Gotoh 2004). A mean scalar gradient driving yields nearly Gaussian PDFs (Overholt & Pope 1996, Ferchichi & Tavoularis 2002, Schumacher & Sreenivasan 2005) and, in some experiments, to exponential tails (Jayesh & Warhaft 1991, Warhaft 2000). If the Gaussian result is correct, it would be in disagreement with the Kraichnan model, for which the tails of the PDF are always super-Gaussian or exponential (Shraiman & Siggia 1994, Balkovsky & Fouxon 1999).

At present, we do not fully understand the circumstances under which the passive scalar mixed by Navier-Stokes turbulence assumes a Gaussian or exponential PDF. The shape of the PDF in inhomogeneous shear flows is understood even less well. For example, if one measures it on the centreline of the wake of a heated cylinder, the PDF in the far-field has an exponential shape for the cold part (coming from the entrainment of the ambient cold fluid) but is Gaussian for the hot part (coming from upstream all the way from the heated cylinder). See Kailasnath *et al.* (1993).

(iv) Consider scalar gradient statistics for the Kraichnan model. An analytical result for the tails of the PDF of scalar dissipation $\epsilon_\theta = \kappa(\nabla\theta)^2$ exists for a smooth white-in-time flow. Using the Lagrangian approach, Chertkov *et al.* (1998) and Gamba & Kolokolov (1999) deduced the behavior to be

$$p(\epsilon_\theta) \sim \frac{1}{\sqrt{\epsilon_\theta}} \exp\left(-\epsilon_\theta^{1/3}\right) \quad \text{for} \quad \epsilon_\theta \gg \langle\epsilon_\theta\rangle. \tag{2.7}$$

The numerical data of Schumacher *et al.* (2005) from very finely resolved simulations of high-Sc mixing converge to this formula from below.

(v) It is still unclear in the Kraichnan model as to which qualitative and quantitative difference arise from the finite-time correlation of the advecting flow. This question has been addressed at least partially in Boffetta *et al.* (2004). It is shown there that finite-time correlations of the velocity field in a free-slip surface are important for the clustering tendency of Lagrangian tracers, and the resemblance to the case of infinitesimally small correlation times is only qualitative. It needs to be shown that an instantaneously reshuffled flow can cause the same ramp-cliff structures which are observed for advection in Navier-Stokes turbulence. The simulations by Chen and Kraichnan (1998) suggest that ramp-cliff features are possible even in the Kraichnan model, even if it may appear counter-intuitive *a priori*.

(vi) The relation between active and passive scalars remains unclear as a general principle. Celani *et al.* (2004) review the work on a number of passive and active scalar fields, and conclude that the scaling of these various fields are, in fact, non-universal. The differences are attributed to the correlation between the input to the scalar field and the particle trajectories, again invoking Lagrangian interpretation.

Finally, one may speculate that the research on the Kraichnan model has some implications for Navier-Stokes turbulence as well. It is worth recalling that Kolmogorov formulated his original 1941 theory in what is now called "quasi-Lagrangian frame" (Belinicher & L'vov 1987); it may thus be said that the importance of the Lagrangian nature of turbulent energy cascade was thus implicit in Kolmogorov's work. This issue was recognized also by Kraichnan (1964), who then reformulated the DIA accordingly (Kraichnan 1965). There is still a chasm that needs to be bridged between the work on the Kraichnan model for the scalar and the calculation of anomalous exponents for the hydrodynamic field (see, e.g., Chen *et al.* 2005), but some progress is being made (e.g., Angheluta *et al.* 2006). The nonlinearity of the Navier-Stokes equations leads to a strong coupling of the equations of the correlations of different order, which makes it more difficult to calculate scaling exponents from the infinite set of equations (L'vov & Procaccia 1998).

Table 1. *Calculated and estimated values of the length and time scale ratios in the DNS data. N is the number of grid points on a linear side of the computational box. Some of the numbers have been taken from the simulations by Yeung (2002) and Yeung et al. (2005).*

N	R_λ	L/η	T/t_η
128	89	56	8
256	140	100	13
512	230	190	20
1024	390	300	40
2048	700	600	65
4096	1200	1000	120
132,072	8900	11,000	900

3. Lagrangian simulations and experiments in Navier-Stokes turbulence

As explained earlier, following a cloud of Lagrangian particles has given new insights, and it is no surprise that numerical schemes based on Lagrangian particles have been done for computing the scaling exponents accurately (Frisch *et al.* 1998, Gat *et al.* 1998). These calculations are not meant to be efficient for computing all aspects of Lagrangian hydrodynamics. Indeed, for such purposes, the preferred method is still the Direct Numerical Simulation (DNS) of the Eulerian equations followed by smooth interpolations of velocities at the positions of the advected tracers (Yeung 2002, Toschi & Bodenschatz 2009). The Lagrangian data are thus more expensive. From Table I, one can infer the computational work (which is the product of the grid points and the number of integration time steps) needed for computing homogeneous and isotropic turbulence in a periodic box of N grid points on the side. It is typically of the order of N^3; see (3.1). Here, R_λ is the Taylor microscale Reynolds number, L/η_K is the ratio of the large scale of the velocity to the Kolmogorov dissipation scale, and T/t_η is the ratio of the characteristic time of the large scale L to that of the dissipation scale η_K.

In Table I, except for the last row,† the data have been deduced from existing DNS data, but those for the Lagrangian timescale ratio have been obtained by extrapolating existing experience at substantially lower Reynolds numbers.

In Eulerian turbulence, the inertial scaling range is roughly about 1/100th of L/η_K, as was discussed for example by Sreenivasan & Dhruva (1998) for the atmospheric boundary layer data at $R_\lambda = 10,000$-$20,000$. (Actually, this fraction appears to depend weakly on the Reynolds number, but we shall not discuss this detail here.) If the same factor holds for time scales as well, we may find a decade of scaling only for R_λ of the order of 10,000. Even this is not certain because the Lagrangian events are distributed with stronger tails than Eulerian events (La Porta *et al.* 2001, Mordant *et al.* 2001) and hence the incursion of the nonuniversal features of the large scale may be stronger for Lagrangian properties; in any case, it is clear

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† The last row is an estimate of the upper bound of what is computable in principle. The physical size of a computer cluster has been assumed to be that of the present Earth Simulator, the size of the computing element has been replaced by atomic dimensions (since it cannot get any smaller!), and different parts of the computer are assumed to communicate at the speed of light (since it cannot get any faster!). While these upper bound estimates can be improved in detail, they provide a reasonable order of magnitude.

that far higher Reynolds numbers are needed to observe universality in Lagrangian data. It is thus no surprise that even Richardson's law of dispersion (Richardson 1926) is yet to be observed directly over a decent range of scales in simulations and experiments (Ott & Mann 2000, Boffetta & Sokolov 2002, Biferale *et al.* 2005, Bougoin *et al.* 2006, Schumacher 2008). Recall also that Richardson's own compilation consisted of data from several disparate sources and, on hindsight, leaves room for improvement. Further difficulties in observing a Richardson scaling are related to the sensitive dependence on the initial conditions as discussed by Bourgoin *et al.* (2006) and Sawford *et al.* (2008).‡

We believe that the estimates presented in Table I may have to be revised unfavorably because of small-scale intermittency. According to the well-known estimate, the computational work needed for the DNS of turbulence varies as the third power (e.g., Orszag 1973) of the large scale Reynolds number, Re:

$$N^3 \times N_T \sim \left(\frac{L}{\eta_K}\right)^4 = Re^3\,. \tag{3.1}$$

Here, N_T is the number of time steps for one large-scale eddy turnover time. It has been argued recently that intermittency renders the smallest spatial scales smaller than the Kolmogorov scale, η_K (Sreenivasan 2004, Yakhot & Sreenivasan 2005, Schumacher *et al.* 2007, Schumacher 2007). Thus, in the DNS of turbulence that computes the smallest scale, the Eulerian computational work would increase as the fourth power of the large scale Reynolds number (Yakhot & Sreenivasan 2005, Schumacher *et al.* 2007). Pragmatically, one may be able to work with an intermediate value between the third and the fourth power of the Reynolds number by sacrificing a little on the very smallest scales, but the net effect is that one can only achieve, for a given computational box size, lower Reynolds numbers than the above estimates suggest.

In Lagrangian simulations, the conventional estimate for the computational work is of the order of $Re^3 \ln(Re)$, the logarithmic factor arising from interpolations of the Eulerian data. This multiplying factor is nontrivial if Re is large. All indications are that the Lagrangian data are more intermittent in character. If so, what is the corresponding estimate of the computational cost, in analogy to the fourth power dependence on Reynolds number, compared with the traditional third power in Eulerian simulations? For a further discussion of these points, see also Yakhot (2008b).

In connection with Lagrangian intermittency (Novikov 1989), the precise set of measurements that one should make is somewhat unclear. In principle, the statistics of pairs of particles that maintain a fixed separation distance will be different from those of fixed separation along a single particle trajectory. Commonly evaluated quantities are the moments of velocity differences of a single Lagrangian particle taken at two times separated by a chosen delay. The frequently used assumption of translating the intermittency of Eulerian spatial increments to that of Lagrangian temporal increments has been questioned (Homann *et al.* 2007, Yakhot 2008b). Strong Lagrangian accelerations (i.e., strong Lagrangian intermittency events) in-

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‡ In the Kraichnan model, a Richardson scaling of the mean-square distance with respect to time follows for a scaling exponent $\zeta = 4/3$ instead of the classical Kolmogorov-like exponent $\zeta = 2/3$.
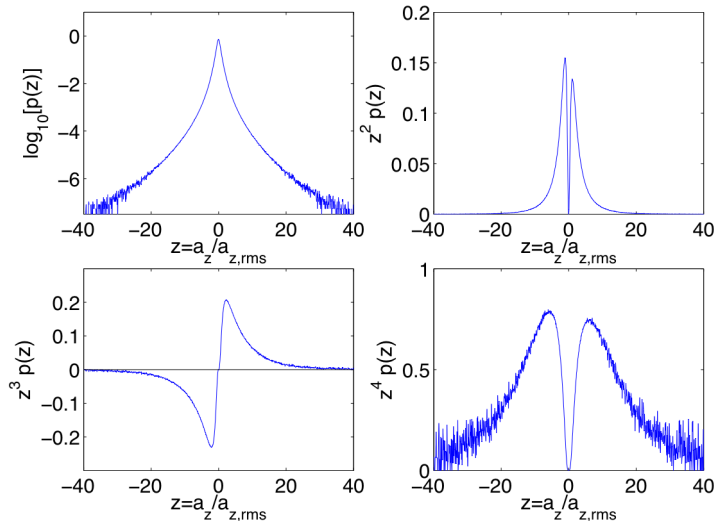
Figure 1. Convergence of the statistics of the vertical acceleration component a_z in turbulent Rayleigh-Bénard convection (Schumacher 2008, 2009). The results is gathered for 4.4×10^8 events in a Cartesian slab of aspect ratio 4 which is resolved by $2048 \times 2048 \times 513$ grid points. The Rayleigh number is $Ra = 1.2 \times 10^8$ and the Prandtl number $Pr = 0.7$. The bulk Reynolds number is proportional to the square-root of the Rayleigh number.

deed appear frequently at the edge of a vortex sheet or in the plane perpendicular to a quasi-one-dimensional vortex tube, these being atypical in the Eulerian case.

In fact, the following interesting, albeit rough, facts can be deduced from measurements and simulations (La Porta *et al.* 2001, Mordant *et al.* 2001, Biferale *et al.* 2004, Reynolds *et al.* 2005, Homann *et al.* 2007) of Lagrangian turbulence. Lagrangian accelerations of the order of 5 times the standard deviation occur with the frequency of one in a thousand, those with 30 times the standard deviation occur with a frequency of one in a million, and those of magnitude 100 times the standard deviation occur with a non-negligible frequency of once every billion times. It is hard to see how the non-universal effects can be avoided altogether under the circumstances. In the experimental data of Crawford *et al.* (2005), one can see that the conditional accelerations depend quite strongly on the magnitude of velocity fluctuations. Figure 1 demonstrates that the convergence of the Lagrangian statistics requires significant efforts. The vertical acceleration (which is less intermittent than the lateral acceleration) in convective turbulence is shown. Even for a record of more than 4×10^8 data points, the statistical convergence of the fourth-order moment remains problematic.

Two consequences of these observations are worth noting. First, it becomes more difficult to obtain converged statistics of high-order Lagrangian moments. Second, because the tails are highly extended, rare events contribute more strongly to high-order moments. Since the tails are also affected by the (non-universal) large scale features, it is not clear that one can seek universal characteristics of high-order moments with the same level of confidence as for Eulerian quantities. Only recently, first successful attempts to this problem in case of Lagrangian structure
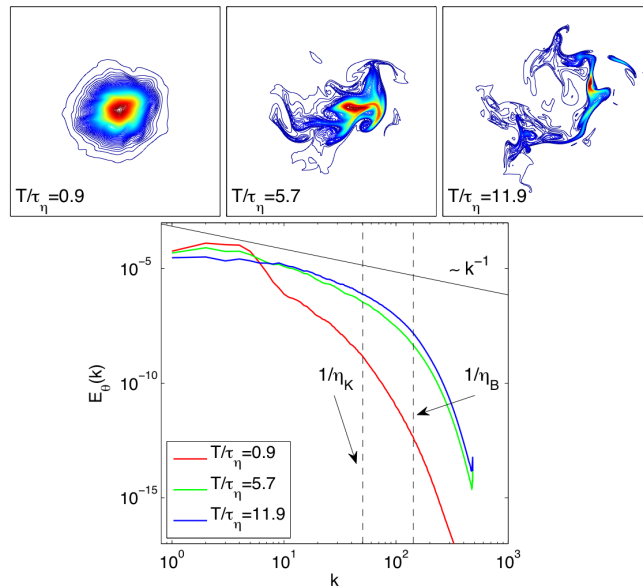
Figure 2. Mixing of an initially Gaussian (and decaying) scalar blob in a homogeneous isotropic statistically stationary turbulent Navier-Stokes flow at a Taylor microscale Reynolds number of $R_\lambda = 64$ and a Schmidt number $Sc = 8$. The turbulence is resolved in a periodic box of side length 2π with 1024^3 grid points. The top row shows the evolution of concentration contours in x-z plane cuts at $y = \pi$ for progressing time. The bottom panel shows the corresponding evolution of the scalar variance spectrum. The classical Batchelor scaling, $E_\theta(k) \sim k^{-1}$, the Kolmogorov dissipation length η_K and the Batchelor diffusion length, η_B are indicated as well.

functions were reported. Theoretical predictions for the scaling exponents in the limit $Re \to \infty$ (Zybin *et al.* 2008) were found to be in remarkable agreement with experiments (Xu *et al.* 2006). Certainly, more experiments are necessary in this area. As a measure of the sensitivity of Lagrangian statistical quantities to the tails of the distribution, one may cite the work of Sawford *et al.* (2005) which shows that backward relative diffusion is much faster than forward diffusion even for stationary and isotropic turbulence.

4. Implications for scalar mixing at high Schmidt numbers

Let us wrap up the issues of the last two sections by discussing the implications of the Kraichnan model for the particularly important case of scalar mixing at high Schmidt numbers. We will consider three aspects.

The argument can be made that the Kraichnan model for the smooth limit of the velocity field, $\zeta \to 2$, comes close to the high-Sc mixing regime in a Navier-Stokes flow with very large Reynolds numbers. The latter would yield a very short Kolmogorov timescale τ_η and an extended viscous-convective range. However, one fundamental difference remains. The finite time over which a local flow pattern with

compressional strain persists is a necessity to steepen the frequently observed sheet-like scalar gradient patterns against the action of molecular diffusion (Sreenivasan & Prasad 1989, Dahm *et al.* 1991, Buch & Dahm 1996, Villermaux & Meunier 2003, Schumacher *et al.* 2005). Therefore, the permanent re-shuffling of the local flow patterns in the Kraichnan case will destroy the well-known stretch-twist-fold scenario, and sheet-like structure of the scalar gradient fields may well be absent. However, statistically, the Kraichnan model does produce very strong gradients (as given by (2.7) for the tails), which seem to form a limit to the mixing in Navier-Stokes turbulence. All current numerical studies on high-Schmidt-number mixing in Navier-Stokes turbulence suffer from the small Reynolds numbers that can be obtained when the largest gradient fluctuations are resolved (Schumacher *et al.* 2005). Therefore, the existence of the so-called mixing transition, which postulates a weaker Reynolds number dependence of scalar mixing at large Reynolds numbers (Dimotakis 2005, Yakhot 2008a), is still an open issue. Progress on the structural differences and their relation to the statistics for both mixing schemes will be quite useful.

Although the closure problem is removed in the Kraichnan model (see (2.4)), analytical predictions on the anomalous scaling of scalar structure functions, $S_n(r)$, are possible only for limiting cases. For example, the Kraichnan model gives a scaling exponent $\xi_3 = 3 - 7\zeta/5$ when the scalar fluctuations are sustained by a constant mean scalar gradient (Pumir 1996). This scaling could be checked, e.g., by advecting the scalar in Navier-Stokes turbulence first and determining ξ_3. The time correlations of the advecting flow could be destroyed as in Boffetta *et al.* (2004) in order to obtain a Kraichnan flow and the third-order moment analysis can be repeated subsequently. Here, a direct link to the Lagrangian viewpoint is also possible by solving the (stochastic) equation (1.1) for triplets or quadruplets of tracers and studying shapes in the spirit of Pumir *et al.* (2000) and Celani & Vergassola (2001).

Kraichnan's (1968) original motivation for his model was to demonstrate that the opposite extreme to Batchelor's (1959) quasistatic straining motion can result in the same scalar variance spectrum $E_\theta(k) \sim k^{-1}$. This can be seen to be so from (2.5) which becomes, for the second order and $\zeta = 2$,

$$-[2\kappa\nabla_{\mathbf{r}}^2 + d_{ij}(\mathbf{r})\nabla_{\mathbf{r}}^i\nabla_{\mathbf{r}}^j]\langle\theta(\mathbf{x})\theta(\mathbf{x}+\mathbf{r})\rangle = C(r/L)\,. \qquad (4.1)$$

The second term on the left hand side corresponds to the scalar variance transfer term, $T_\theta(k)$, for homogeneous isotropic turbulence and reads

$$T_\theta(k) = 2D_1 \frac{\partial}{\partial k}\left[k^4 \frac{\partial}{\partial k}\left(\frac{E_\theta(k)}{k^2}\right)\right]\,, \qquad (4.2)$$

after Fourier transformation. Indeed, $E_\theta(k) \sim k^{-1}$ yields a k-independent transfer rate $\int_k^\infty T_\theta(p)\,\mathrm{d}p$ in the viscous-convective range. The experiments which demonstrate such spectral roll-off are quite rare; see, e.g., Villermaux *et al.* (2001), who note that the k^{-1} power requires both a large range of scalar scales and a sufficiently large Reynolds number.† It is furthermore unclear how the large-scale forcing of the passive scalar will affect the spectrum. Figure 2 illustrates a simulation which seems

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† A corresponding logarithmic scaling of the second order structure function was, however, observed by Borgas *et al.* (2004).

to be the simplest and therefore most transparent case for verifying the concepts of both Batchelor (1959) and Kraichnan (1968). A scalar concentration blob is advected in a statistically stationary turbulent flow for $Sc = 8$. The data correspond only to moderate Schmidt numbers because of the current numerical limitations, but perhaps display a slow approach to the k^{-1} spectrum.

5. Concluding remarks

The recent Lagrangian work has ushered in a breath of fresh air in turbulence, but it has not been sufficiently integrated with prior Lagrangian perspectives. Among others, most of the recent work has focused exclusively on material particles, but there are many other Lagrangian aspects to the turbulence problem. For example, classical works of Taylor & Green (1937) and Taylor (1938) discuss vortex line-stretching as the basic Lagrangian mechanism of turbulence. The modern work has little to offer towards understanding that problem. The general problems raised by Batchelor (1952)—such as the extension of material lines and surfaces and fluxes across surfaces—have yet to be understood well, even for the Kraichnan model; even basic issues such as the existence of material lines and surfaces in the infinite Reynolds number limit have yet to be addressed. The list of unanswered questions might also include the fractal nature of isosurfaces (e.g., Sreenivasan 1991). It would be useful to answer questions such as: what are fractal dimensions of isoscalar surfaces and other material objects in the Kraichnan model? There is extensive numerical work on the problem (see San Gil 2001), but no theoretical answers. This particular question is important for the mixing of reactive scalars or turbulence in clouds. Scalar isolevel sets in Navier-Stokes case do not display monofractal behaviour for low-Reynolds-number flows (Frederiksen *et al.* 1997, Schumacher & Sreenivasan 2005), though it appears that larger Reynolds numbers of the advecting flow will change this behavior. Other approaches such as the dissipation element analysis which reduces the mixing to a permanent reshuffling of the separatrix lines between zeros of the scalar gradient might also provide complementary insight on the nature of high-Sc mixing (Wang & Peters 2006).

It is our belief that the Lagrangian perspective on turbulence is really essential and that it has not been pushed far enough. It is also our belief that the intrinsic mechanisms of turbulence are essentially Lagrangian.

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