# Anomalous mixing times in turbulent binary mixtures at high Prandtl number

Ricardo Ruiz

Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

David R. Nelson

Department of Physics, Harvard University, Cambridge, Massachusetts 02138 (Received 27 May 1981)

A theory of turbulent binary fluid mixtures is applied to situations where the diffusive Prandtl number, P = v/D, far exceeds the Reynolds number R. This regime is accessible just above the consolute point in binary mixtures, where Prandtl numbers in excess of 10<sup>6</sup> have been observed. We find that sizable large-scale inhomogeneities are mixed to uniformity quite slowly, in a time of order  $\tau = t_0 P/R$ , where  $t_0$  is a characteristic stirring time. The spectrum of concentration fluctuations rapidly acquires a peak at wave vector  $k^* \sim k_0 R$ , where  $k_0^{-1}$  is the scale of the initial inhomogeneity. Mixing times of ten minutes or more are possible.

#### I. INTRODUCTION

Turbulence is known to have a striking effect on the efficiency of mixing. Despite the low diffusivities  $(D \sim 10^{-5} \text{ cm}^2/\text{sec})$  in miscible binary mixtures, one can easily mix a 1-cm inhomogeneity to uniformity in a few seconds. This striking enhancement of an effective transport coefficient is a common feature of turbulence.<sup>1</sup>

It is interesting to consider the reduction of mixing efficiency in the extreme limit of exceedingly small diffusivities. This limit is experimentally accessible in binary mixtures just above the consolute temperature  $T_c$ .<sup>2</sup> Indeed, the diffusivity actually vanishes like a power law,

$$D(T) \approx D_0 \left(\frac{T - T_c}{T_c}\right)^{\mu}, \qquad (1.1)$$

where the exponent  $\mu$  is in the range<sup>2</sup>

$$\mu \approx 0.63 - 0.74. \tag{1.2}$$

By holding the system a few millidegrees above  $T_c$ , diffusivities of order  $10^{-8}$  cm<sup>2</sup>/sec are possible. A dimensionless measure of the reduction in D is the Prandtl number

$$P = \nu / D , \qquad (1.3)$$

where  $\nu$  is the kinematic viscosity. Since the viscosity is only weakly singular<sup>2,3</sup> near the consolute point  $[\nu(T) \approx \text{const} \sim 10^{-2} \text{ cm}^2/\text{sec}]$ , Prandtl numbers as large as  $10^6$  can be obtained.

Questions of mixing efficiency focus on the relaxation of a concentration variable

$$\psi(\mathbf{\bar{r}},t) = [\rho_{\mathbf{A}}(\mathbf{\bar{r}},t) - \rho_{\mathbf{B}}(\mathbf{\bar{r}},t)]/\rho_0, \qquad (1.4)$$

where  $\rho_A(\bar{\mathbf{r}}, t)$  and  $\rho_B(\bar{\mathbf{r}}, t)$  are the mass densities of two species A and B, and  $\rho_0$  is the mean mass density. We will, for simplicity, consider a "symmetric" binary mixture where the consolute point occurs at 50-50 concentrations, and where

$$\langle \psi(\mathbf{\tilde{r}},t) \rangle = 0 \tag{1.5}$$

in equilibrium. Standard theories of turbulent mixing<sup>4-8</sup> assume that  $\psi(\bar{\mathbf{T}}, t)$  is passively advected by a turbulent velocity field  $\bar{\mathbf{v}}(\bar{\mathbf{T}}, t)$ ,

$$\frac{\partial \psi}{\partial t} + (\vec{\nabla} \cdot \vec{\nabla}) \psi = D \nabla^2 \psi .$$
 (1.6)

Suppose a large-scale inhomogeneity with  $\psi$  of order 1 is stirred with a characteristic period  $t_0$ . For Prandtl numbers of order unity or less, the inhomogeneity would be dissipated in a time of order  $t_0$ . For large  $P[P \gg \exp(R^{1/2})]$  a passively advected inhomogeneity would be mixed in a longer time (see Sec. II),

$$\tau_{\text{passive}} \approx t_0 \frac{\ln P}{2R^{1/2}} , \qquad (1.7)$$

where R is the Reynolds number. The mixing process can be significantly slowed down, but only in the limit of unrealistically large Prandtl numbers. A modest Reynolds number of, for example,  $10^3$ , would require Prandtl numbers in excess of  $10^{13}$  for an observable effect.

The concentration variable  $\psi(\mathbf{f}, t)$  is, however, not simply advected passively by the velocity field in 50-50 binary mixtures. Concentration gradients react back on the velocity field to produce flow. A theory of binary mixture turbulence capable of dealing with this effect was worked out in Ref. 9, henceforth referred to as I. Here, we observe that the theory predicts anomalously long mixing times in these "active" binary mixtures. In particular, we find that the mixing time in the limit  $P \gg R$  is

2727

© 1981 The American Physical Society

$$au_{\text{active}} \approx au_0 \frac{P}{R}$$
, (1.8)

in contrast to Eq. (1.7). For Prandtl numbers of order  $10^6$  and Reynolds numbers of order  $10^3$ , large-scale inhomogeneities can take ten minutes or more to mix.

Also of interest is the spectrum of concentration fluctuations

$$S(\mathbf{k},t) = \langle |\psi_{\mathbf{k}}(t)|^2 \rangle, \qquad (1.9)$$

which can be measured directly via light scattering.<sup>10</sup> Here  $\psi_{\vec{k}}(t)$  is the Fourier transform of  $\psi(\vec{r},t)$ . In turbulence theory, one traditionally works with the spectral function

$$C(k, t) \equiv 2\pi k^2 S(k, t)$$
. (1.10)

Figure 1 shows the time evolution of C(k, t), obtained from closure equations derived in I (see Sec. III). Fluctuations are concentrated initially around wave vector  $k_0$ , with  $R = 10^3$  and  $P = 10^5$ . The spectrum is rapidly transferred to large wave vectors, where it peaks up around a wave vector  $k^* \gg k_0$ . The peak will persist for a time given by Eq. (1.8). We find that  $k^*$  is determined only by  $k_0$  and the Reynolds number

$$\boldsymbol{k}^* \approx \boldsymbol{R} \boldsymbol{k}_0. \tag{1.11}$$

Equations (1.8) and (1.11), which are the principal results of this paper, apply when the variation of  $\psi(r, t=0)$  across the sample is of order unity. The modifications necessary when this variation is small are discussed in Sec. III. Of course, Eq. (1.7), (1.8), and (1.11) are all subject to dimensionless multiplicative corrections of order unity.

We note finally that all wave vectors discussed



FIG. 1. Time evolution of C(k,t) ( $\alpha = \nu^2$ ). The stirring force is sharply peaked around  $k \sim k_0$ , and the time is measured in units of the circulation time at  $k_0$ . The parameters are  $P = 10^5$ ,  $R \simeq 10^3$ , and  $C_{\text{tot}}(t=0) = 2^{-4}$ , corresponding to  $k_d \simeq 2^7$  and  $k^* \simeq 2^9$ .

above must be smaller than the inverse correlation length  $\xi^{-1}$ . Indeed, length scales smaller than  $\xi$  must be in equilibrium, in order that D(T)be given by Eq. (1.1). This does not seem to be a severe limitation, even at temperatures quite close to  $T_c$ .

In Sec. II, we briefly review the hydrodynamic equations appropriate to turbulent binary mixtures, as well as the predictions for *passively* advected mixtures. Section III summarizes the results for active binary mixtures at high Prandtl number. Although high Prandtl number mixtures were discussed briefly and qualitatively in I, both the numerical results and emphasis of this section are new. Several technical points are discussed in the Appendices.

#### II. HYDRODYNAMICS AND RESULTS FOR PASSIVELY ADVECTED MIXTURES

To describe turbulent binary mixtures, we supplement Eq. (1.6) with modified Navier-Stokes equations for the velocity field  $\bar{\mathbf{v}}(\mathbf{\dot{r}},t)$ , namely,<sup>3,9</sup>

$$(\partial_{t} + \vec{\nabla} \cdot \vec{\nabla}) \vec{\nabla} = \frac{-1}{\rho_{0}} \vec{\nabla} p' + \nu \nabla^{2} \vec{\nabla} - \alpha \nabla^{2} \psi \vec{\nabla} \psi + \vec{f},$$
(2.1a)

$$\vec{\nabla} \cdot \vec{\nabla} = 0 \tag{2.1b}$$

The quantity  $\mathbf{f}(\mathbf{\tilde{r}}, t)$  is a Gaussian stochastic force designed to simulate stirring at long wavelengths. Several longitudinal terms involving  $\psi$  have been absorbed into a modified pressure p'. The reversible coupling  $\alpha$  has the dimensions of a transport coefficient squared and is in order of magnitude,<sup>9</sup>

$$\alpha \sim \nu^2 \,. \tag{2.2}$$

The model of binary mixture hydrodynamics defined by Eqs. (1.6) and (2.1) has been studied both in<sup>3</sup> and out<sup>9</sup> of equilibrium.

In I, integral equations for the Fourier transformed spectra

$$E_{v}(k,t) = 2\pi k^{2} \int d^{3}r \ e^{i\vec{k}\cdot\vec{r}} \langle \vec{\nabla}(\vec{\mathbf{r}},t)\cdot\vec{\nabla}(\vec{\mathbf{0}},t) \rangle , \quad (2.3a)$$
$$C(k,t) = 2\pi k^{2} \int d^{3}r \ e^{i\vec{k}\cdot\vec{r}} \langle \psi(\vec{\mathbf{r}},t)\psi(\vec{\mathbf{0}},t) \rangle , \quad (2.3b)$$

were obtained using an eddy-damped quasinormal Markovian closure<sup>1</sup> of the hierarchy of correlation function equations associated with (1.6) and (2.1). These integral equations take the form

$$(\partial_t + 2Dk^2)C(k,t) = T_c(k,t),$$
 (2.4a)

$$(\partial_t + 2\nu k^2) E_v(k,t) = T_v(k,t) + F(k),$$
 (2.4b)

where F(k) is the spectrum associated with the Gaussian stochastic force  $\tilde{f}$ . The "transfer functions"  $T_c$  and  $T_v$  involve convolutions of the instan-

2728

10 <sup>2</sup>

taneous spectra  $E_v$  on C over all wave vectors (see Appendix A). The numerical results of I were restricted to unit Prandtl number.

In this section, we illustrate the predictions of standard turbulence theories<sup>4-8</sup> for passively advected mixtures ( $\alpha = 0$ ), with a numerical solution of Eq. (2.4) at high Prandtl number. We imagine that a large-scale concentration inhomogeneity is mixed toward uniformity by the usual Kolmogorov cascade of velocity fluctuations,<sup>1</sup>

$$E_{n}(k) \approx \epsilon^{2/3} k^{-5/3}$$
. (2.5)

The quantity  $\epsilon$  is the injection rate of kinetic energy. Assuming the fluid is stirred at a scale  $I_0 = k_0^{-1}$ (which we also take as the scale of the initial concentration inhomogeneity), the spectrum (2.5) holds over an "inertial range" of wave vectors

$$k_0 \leq k \leq k_d \,. \tag{2.6}$$

Velocity fluctuations vanish rapidly beyond a viscous cutoff.<sup>1</sup>

$$k_{s} = k_{0} (\epsilon / \nu^{3})^{1/4} \,. \tag{2.7}$$

Concentration fluctuations are carried quickly down to length scales of order  $k_d^{-1}$  by the kinetic turbulence. This happens in a time of order  $t_0$ , where  $t_0$  characterizes the stirring of large scales. The concentration spectrum rapidly acquires the form<sup>4-8</sup>

$$C(k) \sim k^{-5/3}$$
 (2.8)

with a time-dependent amplitude.

At high Prandtl number, D is much less than  $\nu$ , and concentration fluctuations cannot be dissipated at the scale at which the kinetic cascade terminates. The concentration continues to cascade towards small scales, but with a different powerlaw falloff of the spectrum<sup>4-8</sup>

$$C(k) \sim k^{-1}$$
. (2.9)

The characteristic time for a fluctuation to travel one octave in k space is a typical time for the velocity at  $k_a$ ,<sup>4-8</sup>

$$t_a = 1/\nu k_a^2 = \sqrt{\nu/\epsilon} \quad . \tag{2.10}$$

The quantity  $t_d^{-1}$  is like an effectively uniform strain rate which stretches small-scale concentration fluctuations.<sup>4-8</sup> The  $k^{-1}$  "viscous-convective" cascade of concentration fluctuations is terminated when diffusion finally becomes important, at wave vector<sup>4-8</sup>

$$k'_{d} = k_{d} \sqrt{\nu/D}$$
 (2.11)

The physical picture outlined above is illustrated in Fig. 2, which is the result of numerically integrating the closure equations (2.4).<sup>11</sup> Regimes with power-law spectra given by (2.8) and (2.9) are clearly indicated. Technical remarks about the numerical integration procedure necessary at high Prandtl number are given in Appendix A. The closure equations reduce to a particularly simple form when  $k \gg k_d$ . Mixing in the viscous-convective range of passive mixtures can be studied analytically in this limit. The results are given in Appendix B.

The above theory predicts a mixing time for large-scale concentration inhomogeneities consisting of three contributions,

$$\tau_{\text{pass}} \approx t_0 + t_d \ln(k'_d/k_d) + \frac{1}{D(k'_d)^2}$$
 (2.12)

The first term is the time for a fluctuation to reach  $k_d$ , the second is the time to cascade from  $k_d$  to  $k'_d$ , and the third is the dissipation time actually necessary at  $k'_d$ . (Corrections of order unity should multiply all three terms.) Making use of (2.10), (2.11), and the relation,<sup>1</sup>

$$t_d = t_0 / R^{1/2} , \qquad (2.13)$$

one can rewrite Eq. (2.12) as

$$\tau_{\text{pass}} = t_0 \left( 1 + \frac{\ln P}{2R^{1/2}} + R^{-1/2} \right) \,. \tag{2.14}$$

Evidently, the mixing time  $\tau_{\text{pass}}$  for large R is of order  $t_0$  unless  $\ln P \gg R^{1/2}$ , in which case Eq. (1.7) holds. As discussed in the introduction, unphysically large Prandtl numbers would be required to obtain mixing times significantly longer than  $t_0$ .

## **III. MIXING IN ACTIVE BINARY MIXTURES**

As fluctuations cascade toward the ultraviolet, concentration gradients are enhanced dramatically. One might expect that the  $\alpha$  term in Eq. (2.1a), which allows concentration gradients to react back on the velocity, eventually becomes important. As was discussed in I, two quadratic conservation laws play a role. In addition to the squared integral of  $\psi$ ,

$$C_{\rm tot} = \frac{1}{2} \int d^3 r \, \psi^2(\bar{\mathbf{T}}, t) \,, \qquad (3.1)$$

the quantity

$$E_{\text{tot}} = \frac{1}{2} \int d^{3} \gamma [|\bar{\mathbf{v}}(\bar{\mathbf{r}},t)|^{2} + \alpha |\bar{\mathbf{v}}\psi(\bar{\mathbf{r}},t)|^{2}] \qquad (3.2)$$

is conserved in the absence of dissipation and forcing. The dynamical coupling between  $\vec{\nabla}\psi$  and  $\vec{\nabla}$  tends to produce equipartition between the kinetic and gradient terms of (3.2). For large  $\alpha$ , the system actually supports wavelike excitations which produce this equipartition.<sup>9</sup> For more realistic values of  $\alpha$ , however, these waves are always overdamped.

In I, it was argued that the  $\alpha$  coupling has no



FIG. 2. Time evolution of C(k,t) for passive mixtures ( $\alpha = 0$ ). The stirring force and the time units are as in Fig. 1. Here one has  $P = 9 \times 10^4$ ,  $R \simeq 10^3$ , and  $C_{\text{tot}}(t=0)=1$ . The inset displays  $\pi_c(k) = \int_0^k T_c(q) dq$ . It would be flat if the Kolmogorov ideas about local cascades were perfectly obeyed.

effect until k exceeds  $k_d$ . At this point, the  $k^{-1}$  viscous-convective concentration cascade begins. Since C(k) is continually transferred toward large wave vectors, the root-mean-square fluctuations in the concentration gradients,

$$\left|\vec{\nabla}\psi\right|_{\rm rms}^2 = \int_0^\infty dk \, k^2 C(k) \,. \tag{3.3}$$

must increase. When  $|\vec{\nabla}\psi|_{\rm rms}$  becomes sufficiently large, concentration gradients are converted into velocity fluctuations and immediately dissipated by viscosity. Since  $|\vec{\nabla}\psi|_{\rm rms}$  can no longer grow, the ultraviolet cascade of concentration flucutations is terminated prematurely.

The characteristic rate at which concentration gradients are converted into kinetic energy and dissipated is<sup>9</sup>

$$\omega_{-} \approx \alpha B^{2}(k) / \nu , \qquad (3.4)$$

where

$$B^{2}(k) = \int_{0}^{k} dq \ q^{2}C(q) \ . \tag{3.5}$$

In I we derived the rate (3.4) from a linearization of the equations (1.6) and (2.1) around a configuration in which there was a uniform concentration gradient imposed on the system. The relaxation rate (3.4) can also be obtained from the closure equations (A3) and (A4) by using a nonlocal expansion without the need for any linearization.<sup>12</sup> In I it was shown, modulo possible logarithmic corrections,  $that^{13}$ 

$$B(k) \approx B_0 \left(\frac{k_d}{k_0}\right)^{2/3} \left(\frac{k}{k_d}\right), \qquad (3.6)$$

where  $B_0$  is the root-mean-square concentration gradient at time t=0:

$$B_0 = [\langle | \vec{\nabla} \psi(\vec{\mathbf{r}}, t=0|^2 \rangle]^{1/2}$$
$$= k_0 [\psi^2(\vec{\mathbf{r}}, t=0)]^{1/2}. \qquad (3.7)$$

We have used (3.7) to define the characteristic wave vector  $k_0$  of the initial large-scale unhomogeneity. The first multiplicative factor in (3.6) accounts for stretching of concentration gradients during the inertial range cascade, while the second takes into account stretching in the viscousconvective range. The cascade terminates when  $\omega_{-}$  just equals the strain rate  $\nu k_d^2 = \sqrt{\epsilon/\nu}$  at which concentration fluctuations are transferred from one octave to the next. Using (3.4), (3.6), and (3.7), we find

$$k^{*} = \left(\frac{\nu^{2}}{\alpha \psi_{0}^{2}}\right)^{1/2} \left(\frac{k_{d}}{k_{0}}\right)^{1/3} k_{d}, \qquad (3.8)$$

where

$$\psi_0^2 = \langle \psi^2(\tilde{\mathbf{r}}, t=0) \rangle = C_{\text{tot}}(0). \qquad (3.9)$$

For large Prandtl numbers  $P \gg R$ , the  $k^{-1}$  cascade never reaches the diffusive cutoff  $k'_d$ .

Although concentration gradients are dissipated at  $k^*$ , the concentration itself persists. Indeed, an immediate consequence of (1.6) is that

$$\frac{dC_{\rm tot}}{dt} = -2D \int_0^\infty dk \, k^2 C(k) \,. \tag{3.10}$$

If D is small, and concentration gradients dissipate before they can grow significantly, concentration fluctuations will get hung up at  $k^*$ . In contrast to the mixing time estimate (2.12) for passive mixtures, we find that (provided  $k^* < k'_d$ )

$$\tau_{\rm act} = t_0 + t_d \ln(k^*/k_d) + \frac{1}{D(k^*)^2} . \tag{3.11}$$

The third term dominates in the limit  $P \gg R$ , provided  $\psi_0$  is not too small,

$$\tau_{\rm act} \approx \frac{1}{D(k^*)^2} = t_0 \left(\frac{\alpha \psi_0^2}{\nu^2}\right) \frac{P}{R} .$$
 (3.12)

Eliminating  $k_d$  from (3.8), we can reexpress the other principal result of this paper,

$$k^{*} = \left(\frac{\nu^{2}}{\alpha\psi_{0}^{2}}\right)^{1/2} Rk_{0}. \qquad (3.13)$$

If  $\psi_0$  is of order unity, and the estimate (2.2) holds, the factors of  $\alpha \psi_0 / \nu^2$  appearing in (3.12) and (3.13) are themselves of order unity. We recall from I that  $\alpha$  is expressible in terms of measurable quantities,

$$\alpha = \frac{\xi^2}{m\chi},\tag{3.14}$$

where  $\xi$  is the thermal correlation length,  $\chi$  is the susceptibility per particle, and *m* is the particle mass. The relation (2.2) for  $\alpha$  is only a crude estimate, whose accuracy may vary from system

to system. Equation (3.14) allows  $\alpha$  to be obtained from experimentally measured quantities, however.

The result of numerically integrating the closure equations (2.1) at high Prandtl number, with  $\alpha = \nu^2$ , is displayed in Fig. 1. The spectrum rapidly acquires a peak at  $k^*$ , which should persist for a time given by (3.13). Details of the numerical integration required to produce Fig. 1 are discussed in Appendix A.

Our analysis neglects the "intermittent" or spotty dissipation fluctuations observed in real turbulence; such effects are usually rather small.<sup>1</sup> The closure approximation used here constitutes a kind of "mean-field theory" of turbulence. Arguments suggesting an intermittent probability distribution for  $(\overline{\nabla}\psi)^2$  in the viscous-convective range have been developed by Kraichnan.<sup>8</sup> It would be interesting to see in detail how this intermittency alters the estimates presented in this paper.

#### ACKNOWLEDGMENTS

It is a pleasure to thank U. Frisch and J. Toner for useful conversations, and R. H. Kraichnan for emphasizing the possible role of intermittency in the viscous-convective range. This work was supported by the National Science Foundation under Grant No. DMR77-09595. R. R. gratefully acknowledges financial support from the Fundacion I. T. P., Madrid, Spain, and the hospitality of Harvard University. One of us (D.R.N.) acknowledges the receipt of a grant from the Alfred P. Sloan Foundation.

## APPENDIX A: NUMERICAL INTEGRATION OF THE EDDY-DAMPED QUASINORMAL MARKOVIAN CLOSURE

In I we used the EDQNM closure<sup>1</sup> to derive the following set of equations for the velocity spectra  $E_v(k)$  and the concentration spectrum C(k) in three spatial dimensions:

$$(\partial_t + 2Dk^2)C(k) = T_c(k) , \qquad (A1)$$

$$(\partial_t + 2\nu k^2) E_v(k) = T_v(k) + F(k)$$
, (A2)

with

$$T_{v}(k) = \int_{\Delta_{k}} \int dp \ dq \ \frac{1}{kpq} \left[ \theta_{kpq}^{(2)} b_{kpq} E_{v}(q) [k^{2} E_{v}(p) - p^{2} E_{v}(k)] k^{2} + \theta_{kpq}^{(3)} \left( \frac{\sin\beta}{k} \right)^{2} p^{2} q^{2} (p^{2} - q^{2}) [\alpha^{2} (p^{2} - q^{2}) k^{2} C(p) C(q) - \alpha p^{2} C(q) E_{v}(k)] \right]$$
(A3)

and

$$T_{c}(k) = \int_{\Delta_{k}} \int dp \ dq \left(\frac{\sin\beta}{k}\right)^{2} \frac{kq}{p} \theta_{kpq}^{(1)} \left\{ E_{v}(p) \left[k^{2}C(q) - q^{2}C(k)\right] - 2(k^{2} - q^{2})p^{2}C(k)C(q) \right\}$$
(A4)

The double integrals are restricted by momentum conservation to momenta of magnitudes p and q such that the triad (k, p, q) forms a triangle. F(k) is the power spectrum of the forcing, assumed to be Gaussian white noise for convenience, and  $b_{kpq}$  is the usual kinematical coefficient<sup>14,15</sup>

$$b_{kpq} = \frac{1}{2k^2} \left(\frac{\sin\beta}{k}\right)^2 \left[k^2 p^2 + (p^2 - q^2)(k^2 - q^2)\right], \quad (A5)$$

where  $\beta$  is the angle opposite to k in the triangle formed by k, p, and q.

The triad relaxation times  $\theta_{kpq}^{(i)}(t)$ , (i=1,2,3) are taken to be<sup>14,15</sup>

$$\theta_{kpq}^{(i)}(t) = (1 - e^{-t \, \mu_{kpq}^{(i)}}) / \mu k p q^{(i)} , \qquad (A6)$$

where

$$\begin{aligned}
\mu_{kpq}^{(1)} &= \mu_{k}^{c} + \mu_{p}^{v} + \mu_{q}^{c}, \\
\mu_{kpq}^{(2)} &= \mu_{k}^{v} + \mu_{p}^{v} + \mu_{q}^{v}, \\
\mu_{kpq}^{(3)} &= \mu_{k}^{v} + \mu_{p}^{c} + \mu_{q}^{c},
\end{aligned} \tag{A7}$$

and

$$\mu_{k}^{c} - Dk^{2} = \mu_{k}^{v} - \nu k^{2} = C_{E} \left( \int_{0}^{k} dq \, q^{2} E(q) \right)^{1/2} + C_{w} \, k \left( \int_{0}^{k} dq \, \alpha q^{2} C(q) \right)^{1/2}.$$
 (A8)

 $C_{B}$  and  $C_{w}$  are both adjustable constants that we set to  $C_{B} = 0.360^{6}$  and  $C_{w} = 1.^{9}$  It was shown in I that (A3) and (A4) were compatible with the conservation laws (3.1) and (3.2) and their associated absolute equilibrium spectra.

Some comments on the technique used to integrate the closure equations numerically may be helpful. It is well known<sup>14,15</sup> that the usual logarithmic wave number discretization involved in evaluating (A3) and (A4) has the effect of eliminating from the transfer integral the contribution of all the nonisosceles triads whose elongation factor satisfies  $1/A(F) < 2^{1/F} - 1$ , F being the number of k points retained in each octave. Because the integrand in (2.3) vanishes for k=q, we have to use the results of the nonlocal expansion discussed in Appendix B to get any transfer of concentration fluctuations deep in the viscous regime. Since the validity of the cascade picture is deeply rooted in the conservation law associated with (3.1), we have added explicitly to the local transfer (A4) a contribution

$$\{T_{c}(k)\}_{n1} = \frac{2}{15} \frac{\partial}{\partial k} \left[ \int_{0}^{k/A(F)} dq \ q^{2} E_{v}(q) \theta_{kqk}^{(1)} \times \left(k^{2} \frac{\partial C}{\partial k} - 2kC(k)\right) \right].$$
(A9)

The effect of (A9) is dominant for  $k \gg k_d$  when the

fluid velocity reaches its steady state, and irrelevant for  $k < k_d$ , where the transfer is predominantly local.

We conclude this section with a remark concerning the numerical procedure used to obtain results for  $\alpha \neq 0$ . The nonlocal contribution (A9) has been added to the discretized version of the transfer integral (A4) as in the passive case. However, because it represents creation of smallscale concentration fluctuations, it violates, if taken alone, the conservation of the total kinetic and gradient energy (3.2). To restore that conservation law we must carry out the nonlocal expansion for the E(k)C(q) contribution to the energy transfer (A3) as well. The relevant triads are those of Fig. 3(b), and the result of the expansion is

$$T_{v}(k) = -\frac{2}{15} \left\{ \int_{kA(F)}^{\infty} dp \ \theta_{kpp}^{(3)} p^{2} \frac{\partial}{\partial p} \times \left[ p^{2} \frac{\partial}{\partial p} \left( \frac{C(p)}{p^{2}} \right) \right] \right\} k^{2} E_{v}(k) \cdot (A10)$$

It can be checked that (A9) and (A10) conserve energy up to a term, which is numerically insignificant when compared to the total kinetic energy. The cascade process is not affected by this small degree of nonconservation, but it is nevertheless distressing that we have not been able to derive expressions for  $T_v$  and  $T_c$  from the nonlocal expansions that satisfy exactly both conservation laws, even though the original transfer integrals (A3) and (A4) comply with exact detailed conservations of both these quantities.<sup>1</sup>



FIG. 3. Types of interacting triads contributing to (a) (A9) and (b) (A10).

## APPENDIX B: MIXING IN THE VISCOUS-CONVECTIVE RANGE

One can gain some analytical insight into the mechanism of mixing at  $k \gg k_d$ , with  $\alpha = 0$ , by considering the contribution to the transfer integral in (A4) of wave number triads like the one in Fig. 3(a). By definition, the shape parameter

$$A = \frac{\max(k, p, q)}{\min(k, p, q)}$$
(B1)

is large for the interactions represented by this type of triads that are responsible for the process of mixing in the viscous scales. Using the method of Ref. 16, we can expand (A4) in powers of (1/A) and get<sup>6</sup>

$$[T_{c}(k)]_{1s} = \frac{2}{15} \int_{0}^{k_{d}} dp \; \theta_{k}^{(1)} p^{2} E_{v}(p) \\ \times \left(k^{2} \frac{\partial^{2} C(k)}{\partial k^{2}} - 2C(k)\right).$$
(B2)

One sees easily that both the spectrum (2.9) and the absolute equilibrium spectrum  $C(k) \simeq k^2$  [corresponding to equipartition of C(k) over all wave vectors k] cause the transfer integral in (B2) to vanish exactly.

We can use (B2) further to analyze the actual evolutions towards the inertial range spectra. If we introduce the change of variables<sup>17</sup>

$$x = \ln(k/k_d)$$
,  $C(x) = C(k(x))$ , (B3)

we get, setting D = 0,

$$\partial_t \mathfrak{E}(x) = \Omega \left( \frac{\partial^2 \mathfrak{E}}{\partial x^2} - \frac{\partial \mathfrak{E}}{\partial x} - 2 \mathfrak{E} \right).$$
 (B4)

Here the quantity  $\Omega$  is defined by

$$\Omega = \int_{0}^{k_{d}} dp \; \theta_{kpk}^{(1)} p^{2} E_{p} \; . \tag{B5}$$

This characteristics rate becomes time independent and of order  $\sqrt{\epsilon/\nu}$  after the energy spectrum reaches its steady state.

Equation (B4) is a diffusion equation in the x variable, and can be exactly solved by a simple Fourier transform. To simulate the influx of concentration variance through  $k_d$  from the inertial ranges, we add a source term to the right-hand side of (B4),

$$\partial_t \mathcal{C}(x,t) = \Omega \left( \frac{\partial^2 \mathcal{C}}{\partial x^2} - \frac{\partial \mathcal{C}}{\partial x} - 2 \right) + \frac{\chi}{k_d} \delta(x)$$
 (B6)

and treat, for simplicity,  $\chi$  as a time-independent constant. The solution of (B6) is

$$e(x,t) = \frac{e^{-2\Omega t}}{(4\pi\Omega t)^{1/2}} \int dy \, e(y,0) \exp\left(-\frac{(\Omega t - x + y)^2}{4\Omega t}\right) \\ -\chi \int_0^t dt' \, \frac{e^{-2\Omega t'}}{(4\pi\Omega t')^{1/2}} \exp\left(-\frac{(\Omega t' - x)^2}{4\Omega t} + \chi/3\Omega\right) \left(e^{-x}H(x) + e^{2x}H(-x)\right), \tag{B7}$$

where

$$H(x) = \begin{cases} 1, & \text{if } x > 0 \\ 0, & \text{if } x < 0 \end{cases}$$
(B8)

is the Heaviside function.

In the limit  $t \rightarrow \infty$ , (B7) gives a steady state of absolute equilibrium at  $k < k_d$ , and the spectrum (2.9) when  $k > k_d$ . In reality, of course, the absolute equilibrium part of the solution is preempted by the inertial range behavior (2.8).

We can use (B7) further to understand how a concentration fluctuation, initially at some wave vector  $k = k_d e^y$ , gets transferred to the smaller scales. The first term of (B7) describes this fluctuation as centered at time t around a wave number k' such that

. .. . .

$$\ln k' = \ln k + \Omega t , \qquad (B9)$$

and Gaussianly distributed around it with a width  $\sqrt{\Omega t}$ . For fully developed turbulence this width is of the order  $\sqrt{t/t_d}$ , and thus extremely large at the times when the inertial ranges are achieved. This result is sharply in contrast with the one obtained in I from the continuum version of a simple "shell model." There, the fluctuations also followed the characteristic lines (B9), but with no spread at all. No spread occurs because the shell model does not incorporate the absolute equilibrium solution, and yields a first-order differential equation in the continuum limit.

24

- <sup>1</sup>S. A. Orszag, in *Fluid Dynamics*, 1973 Les Houches Lectures, edited by R. Balian and P. L. Peube (Gordon and Breach, New York, 1977); H. A. Rose and P.-L. Sulem, J. Phys. (Paris) 39, 44 (1978).
- <sup>2</sup>For a review of theory and experiment, see H. L. Swinney and D. L. Henry, Phys. Rev. A <u>8</u>, 2586 (1973).
- <sup>3</sup>P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. 49, 43 (1977) and references therein.
- <sup>4</sup>S. Corrsin, J. Appl. Phys. <u>22</u>, 469 (1951).
- <sup>5</sup>G. K. Batchelor, J. Fluid Mech. 5, 113 (1959).
- <sup>6</sup>R. H. Kraichnan, Phys. Fluids <u>11</u>, 945 (1968).
- <sup>7</sup>R. H. Kraichnan, Phys. Fluids 13, 22 (1970).
- <sup>8</sup>R. H. Kraichnan, J. Fluid Mech. <u>64</u>, 737 (1974).
- <sup>9</sup>R. Ruiz and D. R. Nelson, Phys. Rev. A <u>23</u>, 3224 (1981).
- <sup>10</sup>See, e.g., W. I. Goldburg and J. S. Huang, in *Fluctua*tions, Instabilities, and Phase Transitions, edited by

T. Riste (Plenum, New York, 1975). <sup>11</sup>The decay of passive scalars convected by homogeneous

turbulence has been investigated previously with

various closure approximations; see M. Larcheveque, J. P. Chollet, J. R. Herring, M. Lesieur, G. R. Newman, and D. Schertzer, in *Turbulent Shear Flows 2*, edited by J. S. Bradbury, F. Durst, B. E. Launder, F. W. Schmidt, and J. W. Whitelaw (Springer, Heidel-

berg, 1980) and references therein.

<sup>12</sup>R. Ruiz, Ph.D. thesis (unpublished).

<sup>13</sup>Equation (5.7) of I contains two misprints. This equation should read

$$B(k) = B(k_d) \left(\frac{k}{k_d}\right) = B(k_0) \left(\frac{k_d}{k_0}\right)^{2/3} \left(\frac{k}{k_d}\right)$$

- <sup>14</sup>A. Pouquet, M. Lesieur, J. C. André, and C. Basdevant, J. Fluid Mech. <u>72</u>, 305 (1975).
- <sup>15</sup>J. C. André and M. Lesieur, J. Fluid Mech. <u>81</u>, 187 (1977).
- <sup>16</sup>M. Lesieur and D. Schertzer, J. Mcc. (Paris) <u>17</u>, 610 (1978).
- <sup>17</sup>We are indebted to U. Frisch for this useful suggestion.