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ON THE DECAY OF A REACTING SCALAR FIELD IN TURBULENCE

Part II

SPECTRAL BEHAVIOR AT HIGH WAVE NUMBERS

by

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1. Introduction

In part I of this paper[1] the direct interaction approximation was invoked to obtain a determinate set of equations describing the behavior of an isotropic scalar field under two circumstances. Either the scalar is undergoing an isothermal first order reaction or it is undergoing an isothermal second order reaction. It is the purpose of this part of the paper to investigate the high wave number regions of the scalar spectrum and to do so we examine stationary states of both the velocity and scalar fields in the hope that under certain circumstances the structure of the high wave number regions may differ inappreciably for stationary and freely decaying fields. From Kolmogoroff[2] and Kraichnan[3] one can expect that a very high Reynolds number would be one necessary condition and in fact we will presume the existence of an inertial subrange and a dissipation range for the velocity field and assume the energy spectrum results of Kraichnan in these high wave number ranges. The localness of energy spectral transfer, which has been demonstrated[3], will also be appealed to in the following.

The addition of reaction, whether it be first or second order, gives rise to other time scales distinct from those of diffusion and of convection. For approximate stationarity in the high wave number regions it is evident that these scales

will have to be restricted in value. To examine this more closely stationarity is assumed and the subsequent equations are examined for the roles which reaction rate plays.

In particular the asymptotic form of the response function will provide time scale information

2. Scalar Response and Time Correlation Functions at High Wave Number

In its stationary form the direct interaction equation for the response function for a 2nd order reaction, which was derived in (1) appears as

$$\left\{ \frac{\partial}{\partial t} + \kappa k^2 + 2C\bar{v}_0 \right\} g_0(k,\tau)$$

$$= -\pi k \iint_{\Delta} (1-\omega^2)\, pq\, dp\, dq \int_0^{\tau} g_0(k,\tau-s)\, g_0(q,s)\, U(p,s)\, ds$$

$$- 8\pi \frac{C^2}{k} \iint_{\Delta} pq\, dp\, dq \int_0^{\tau} g_0(k,\tau-s)\, g_0(q,s)\, \phi(p,s)\, ds$$

(1.1).

For a first order reaction the last term on the right hand side of (1.1) is nonexistant and $\bar{v}_0$ would be dropped from the last term on the left hand side.

Stationarity also permits the replacement of $U(p,s)$ by $(2\pi p^2)^{-1} E(p)\, \Gamma(p,s)$ and $\phi(p,s)$ by $(4\pi p^2)^{-1} E_s(p)\, \Gamma_s(p,s)$, where $E_s(p)$ and $\Gamma_s(p,s)$ are the scalar equivalents of the energy spectrum

function and the velocity field time correlation functions investigated by Kraichnan.

Thus we have for the right hand side of (1.1)

$$-\frac{k}{2}\iint_{\Delta} E(p)(1-\omega^2)\frac{q}{p}\,dp\,dq\int_0^{\tau} g_0(k,\tau-s)\,g_0(q,s)\,r(p,s)\,ds$$

$$-2\frac{c^2}{k}\iint_{\Delta} E_s(p)\frac{q}{p}\,dp\,dq\int_0^{\tau} g_0(k,\tau-s)\,g_0(q,s)\,r_s(p,s)\,ds$$

Following Kraichnan we may, at high wave numbers, assume that the main contributions of $E(p)$ and $E_s(p)$ are for $p \ll k$ which, since $\underset{\sim}{p}+\underset{\sim}{q}+\underset{\sim}{k}=0$, implies $q \approx k$. Also, since time correlations decrease with wave number, we may estimate the time integrals in the above expression by assuming

$$g_0(k,\tau) \approx g_0(q,\tau)$$

$$r_s(p,s) \approx 1 \approx r(p,s)$$

With these approximations the time and space integrals become independent and the spatial ones are immediately integrable.

We find that (1.1) becomes

$$\left\{\frac{\partial}{\partial t}+Kk^2+2c\bar{v}_0\right\}g_0(k,\tau) = -\left(k^2u'^2+4c^2v'^2\right)\int_0^{\tau} g_0(k,\tau-s)\,g_0 \qquad (1.2)$$

where u' is the root mean square velocity in any direction and f' is the root mean square concentration. The solution of (1.2) with the initial condition $g(k, 0^+) = 1$ is

$$g(k,\tau) = e^{-(Kk^2 + 2C\bar{v}_0)\tau} \frac{J_1[2(ku' + 2Cf')\tau]}{(ku' + 2Cf')\tau} \qquad (1.3)$$

Equation (1.3) shares the advantages and disadvantages of its velocity field analogue discussed in (3). It is pertinent here to notice four time scales: viscous decay $(Kk^2)^{-1}$; reactive decay $(2C\bar{v}_0)^{-1}$; spectral transfer by convection $(ku')^{-1}$; spectral transfer by reaction $(2Cf')^{-1}$. For a first order reaction the second of the above becomes just $(2C)^{-1}$ and the fourth time scale drops altogether.

Thus in the wave number range $k >> \frac{2Cf'}{u'}$ first and second order reactions have similar response functions and perhaps similar spectral transfer behaviors. To affirm the latter consider the time correlation function. In the stationary state the equation describing this function becomes, after some algebra similar to that in (3),

$$\left\{\frac{\partial}{\partial t} + Kk^2 + 2C\bar{v}_0\right\}\Gamma_0(k,\tau) = (k^2u'^2 + 2Cf'^2)\int_\theta^\infty g_0(k,\tau-\theta)\Gamma_0(k,\tau)\,d\tau$$
$$-(k^2u'^2 - 4C^2f'^2)\int_0^\infty g_0(k,\tau)\,\Gamma_0(k,\tau-\theta)\,d\tau \qquad (1.4)$$

It does not appear possible to construct a solution as was done for the velocity field in the inertial subrange. We merely remark that the time scales are those which occur in (1.3) and that, in particular, the wave number range

$k >> \left(\frac{2c\tau'}{u'}\right)$ (1.5) is indeed one in which the spectral transfer due to reaction can be ignored. If such a condition holds in the convection dominated spectral regions the solution of (1.4) obtained by Lee[4] in the nonreactive case is pertinent.

Corrsin[5] has estimated a reactive time which depends on the local spectrum value so that the equivalent condition for neglecting reactive spectral transport is for him

$$k >> c\left(\frac{E_s(k)}{E(k)}\right)^{\frac{1}{2}}$$

The two conditions are likely to be at variance only for low Schmidt number flows in the inertial-diffusive range. In such cases the essential localness of cascading affirmed by Kolmogoroff and Kraichnan would seem to make the latter a more reasonable measure. The prediction by the Direct Interaction hypothesis of dependence of a regime of negligable reactive transport on mean square properties rather than local spectral content is analogous to the retention of mean square velocity in the Direct Interaction prediction of the turbulence spectrum in the inertial subrange. Kraichnan[6] has concluded that the latter is a feature of his approximation which is not realistic and he has proposed a modification to correct this defect.

3. Spectral Function

It is convenient to rewrite the scalar spectrum covariance function in a form in which the time variables are simultaneous. The consequences of doing so in the stationary case is the equation:

$$\{2\kappa k^2 + 4C\bar{\vartheta}_0\} E_\rho(k) = k \iint_\Delta (1-\omega^2)(pq)^{-1} E(p)\, dp\, dq \{k^2 E_\rho(q)\Theta(p,k,q) - q^2 E_\rho(k)\Theta(p,q,k)\}$$

$$+ 2kC^2 \iint_\Delta (pq)^{-1} \{E_\rho(q) E_\rho(p) \Theta_1(p,k,q) + 2E_\rho(p) E_\rho(k) \Theta_1(p,q,k)\}\, dp\, dq \qquad (1.6).$$

$$\Theta(p,k,q) = \int_0^\infty r(p,s)\, g_\rho(k,s)\, r_\rho(q,s)\, ds$$

and

$$\Theta_1(p,k,q) = \int_0^\infty r_\rho(p,s)\, g_\rho(k,s)\, r_\rho(q,s)\, ds$$

are time scales dominated by the shortest of those presented after (1.3) and possibly the vorticity decay time $(\nu k^2)^{-1}$.

The property characterizing the reaction independent terms on the right hand side of (1.6) is that their total contribution over all wave numbers is zero. That is, they

have the classical convective transfer function property and have no direct contribution to the decay of total scalar content. The same is not true of the reactive term which is always positive since each term in it is positive.

Thus in terms of the Direct Interaction hypothesis second order reactions will produce a positive contribution to the spectral "energy" growth at all wave numbers. The curious quadratic occurrence of reaction rate which is evident in (1.6), and indeed in the original Direct Interaction equations, implies that this positive spectral contribution by second order reaction is independent of the sign of C and thus for a negative reaction rate (increasing scalar content) the fluctuating field also feeds the spectrum.

It does not necessarily follow that the predictions of the Direct Interaction hypothesis are unphysical. In fact if one studies exact solutions[7] to the nonlinear problem of second order reaction with no convective or diffusive terms and with an initial statistical scalar field description which is multivariate Gaussian one finds that those terms which contribute to the growth of the scalar spectrum, and which are also independent of the mean field, are all dependent on even powers of the reaction rate. Likewise other approximations, such as the cumulant discard variety, which produce a closed set of equations exhibit the same behavior under Gaussian assumption for the initial field. However in such cases the mean field-fluctuating field interaction is much more complicated in the ensuing equations than the direct interaction predicts and it will be a pertinent matter to

investigate the applicability of the direct interaction hypothesis to this nonconservative field. In the following we find that high wave number, stationary spectral prediction based on this hypothesis are plausible and in qualitative agreement with other existing predictions. Numerical integration then will probably be the only feasible test of the soundness of its application to nonlinear reaction.

Scalar Spectral Shape

Equation (1.6) describes the scalar spectral function behavior in a stationary, locally isotropic turbulence provided that the necessary input to establish stationarity is excluded from the high wave number regime under investigation. In reality we ask at least that the time scales associated with decay of the mean concentration and with decay of the low wave number region be very long compared to that of eddy decay in the high wave number region. Thus to the well established requirement for an inertial subrange in the fluid and an equivalent convective subrange for the scalar we must add the restriction that the reaction rate be sufficiently low that the mean concentration is essentially constant over the lifetime of a high wave number eddy.

For example a necessary condition would be

$$c \ll \left[k^3 E(k)\right]^{1/2}$$

for all k in the inertial subrange.

Thus, in view of the result (1.5) that reactive scalar transport becomes negligible for high enough k and of the stationarity requirement on reaction rate mentioned above, it will be assumed, except in the following paragraph, that stationarity exists and that $k \gg \frac{2C_\theta'}{u'}$ or its local equivalent $k \gg C\left(\frac{E_\theta(k)}{E(k)}\right)^{1/2}$. The consequence of these assumptions is that first and second order reactions have the same spectral shapes and differ only in the effective reaction rate.

In the event that the rate of reaction dominates both the total decay rate and spectral transport in the high wave number region preceeding viscous dissipation, and if one could arrange a stationary situation for the scalar field under such circumstances, the appropriate, if rather academic, form of spectrum function equation would be from (1.6)

$$4C\bar{U}_0 E_\theta(k) = 2kC^2 \iint_\Delta (pq)^{-1} \left\{ E_0(q) E_\theta(p) \Theta_1(p,k,q) + 2E_\theta(k) \Theta_1(p,q,k) E_\theta(p) \right\} dp\,dq$$

Since $\Theta_1 \sim C^{-1}$ dimensional analysis shows a spectral function which behaves as k^{-1}.

Unfortunately there appears to be no simple way to analyse (1.6) for the more interesting situation in which reactive and convective spectral transport are comparable but with the restrictions mentioned earlier, which effectively remove the reactive transport, some progress is possible under the broad assumption of localness of transport. This localness has already been examined for the velocity field[3] and in view of

the close similarity between the direct interaction equation of scalar transport and those of turbulence it appears to be a natural assumption here. We ask what kinds of spectra does the direct interaction hypothesis predict given such extended ranges of wave number that local spectral properties dominate the local spectral behavior.

Inertial Range

In the inertial subrange Kraichnan has obtained from the direct interaction approximation the form $E(k) = A(\epsilon\nu')^{\frac{1}{2}} k^{-3/2}$ where A is a numerical factor and ϵ is the rate of energy decay. As previously mentioned, later work has apparently convinced him that this conclusion is only qualitatively correct but for consistency the above result will be retained here with the expectation also that only qualitative agreement with reality can be expected since the same kinds of complex triad interactions determine the scalar spectrum as determine the energy spectrum.

The scalar transport power function $\Pi_\rho(k)$ can be defined by

$$-\frac{\partial \Pi_\rho(k)}{\partial k} = k \iint_\Delta (1-\omega^2)(pq)^{-1} E(p)\, dp\, dq \left\{ k^2 E_\rho(q)\Theta(p,k,q) - q^2 E_\rho(k)\Theta(p,q,k) \right\} \qquad (2.1).$$

$\Pi_\rho(k)$ represents the mean scalar quantity input to all modes of wave number higher than k from all modes lower than k and it evidently has, by the assumption of localness, the dimensional form

$$[\Pi_\rho(k)] \sim [\Theta][k]^4[E(k)][E_\rho(k)]$$

where the dominant contribution to Θ would normally be convective. Thus $\Theta \sim (u'k)^{-1}$.

Equation (1.5) becomes

$$\left\{ 2Kk^2 + 4C\bar{U}_0 \right\} E_\rho(k) = -\frac{\partial}{\partial k}\Pi_\rho(k)$$

which suggests, if $\Pi(k)$ is determined essential by local spectral values, that

$$\left\{ 2Kk^2 + 4C\bar{U}_0 \right\} E_\rho(k) = -\beta\left(\frac{\epsilon}{u'}\right)^{\frac{1}{2}} \frac{\partial}{\partial k}\left\{ k^{3/2} E_\rho(k) \right\}$$

where β is a constant of order unity. Consequently, it follows

$$E_\rho(k) \sim k^{-3/2} \exp\left\{ \frac{8}{\beta} C\bar{U}_0 \left(\frac{u'}{\epsilon}\right)^{\frac{1}{2}} k^{-\frac{1}{2}} - \frac{4}{3}\frac{K}{\beta}\left(\frac{u'}{\epsilon}\right)^{1/2} k^{3/2} \right\} \qquad (2.2)$$

The non-power-law result (2.2) would seem to invalidate the above argument. Indeed it is evident that (2.2) can not satisfy (2.1) since on integration over wave number triads reaction rate and diffusivity will appear in the denominator. However an expansion of the exponential in a power series reveals that at least the first few terms do satisfy the original integral equation (2.1). For k sufficiently small that the index of the exponential is essentially zero the result is that of pure mixing already described by Lee[4]. Thus it appears

that (2.2) is a valid solution in the spectral region from pure mixing to that in which the index of the exponential is somewhat less than unity, provided such a region is extensive enough.

In the inertial-diffusive-reactive range, Corrsin[5] by adapting and generalizing Onsager's spectral transport analysis, has obtained the following result for first order isothermal reactions which should be compared to (2.2) above

$$E_\rho(k) \sim k^{-5/3} \exp\left\{3C_1 \epsilon^{-1/3} k^{-2/3} - \tfrac{3}{2} K \epsilon^{-1/3} k^{4/3}\right\}$$

where C_1 is a first order reaction rate.

The similarity as far as dependence on k, ϵ, K and reaction rate are concerned is striking and if one can interpret (2.2) and (2.3) as at least predicting the spectral trend near the wave numbers where reaction or diffusion become significant such trends are qualitatively similar. The appearance of r.m.s. velocity in (2.2) derives from its appearance in the inertial subrange spectrum predicted by the direct interaction hypothesis, a feature which is probably not correct in the velocity spectrum[6] and therefore unlikely to be a real effect for the scalar spectrum.

At very small Schmidt numbers it is possible that there could exist a subrange in which θ is dominated by the time scale $(K k^2)^{-1}$ and a dimensional argument as before would lead to

$$2Kk^2 E_\rho(k) = -\frac{\beta}{K}\left(\frac{\epsilon}{u'}\right)^{\frac{1}{2}} \frac{\partial}{\partial k}\left(k^{\frac{1}{2}} E_\rho(k)\right)$$

$$\text{or} \quad E_\rho(k) \sim k^{-\frac{1}{2}} \exp\left\{-\frac{4}{5}\frac{K^2}{\beta}\left(\frac{u'}{\epsilon}\right)^{\frac{1}{2}} k^{5/2}\right\} \qquad (2.3)$$

which may indeed be valid for $k < \left[\frac{4}{5}\frac{K^2}{\beta}\left(\frac{u'}{\epsilon}\right)^{\frac{1}{2}}\right]^{-3/5}$.

It exhibits a considerable less steep slope than for pure mixing in the inertial subrange but such a situation is presumably only obtainable, if at all, with liquid metals at very high Reynolds numbers.

Viscous Range

For turbulence of sufficiently high Reynolds number Kraichnan[3] has demonstrated that the velocity spectrum function $E(k)$ in the wave number region where viscous decay is a direct effect, has the form

$$E(k) \sim \frac{k^2}{k_d^2} e^{-\alpha \frac{k}{k_d}} \tag{2.4}$$

where α is some numerical factor and k_d is the wave number marking the beginning of the viscosity dominated region of the spectrum.

The pertinent scalar spectrum function equation, from (1.5) is

$$\left\{2\kappa k^2 + 4C\bar{v}_0\right\} E_\rho(k) = k\iint_{\Delta} (1-\omega^2)(pq)^{-1} E(p)\, dp\, dq \left\{k^2 E_\rho(q)\Theta(p,k,q) - q^2 E_\rho(k)\Theta(p,q,k)\right\} \tag{2.5}$$

which, in view of the exponential nature of $E(k)$, immediately suggests the following form for $E_\rho(k)$:-

$$E_\rho(k) \sim k^n e^{-\alpha \frac{k}{k_d}}$$

For the case of Schmidt number of order 1, since reaction

rate effects have already been discarded in spectral transfer it is consistent to limit its value on the left hand side to be such that dissipation dominates there. Except for the possibility of rapid reaction, high mean and very small fluctuations at high Reynolds numbers such a limitation appears to be valid.

Then we have

$$2Kk^2E_\rho(k) = k\iint_{\Delta}(1-\omega^2)(pq)^{-1}E(p)\,dp\,dq\left\{k^2E_\rho(q)\Theta(p,k,q) - q^2E_\rho(k)\Theta(p,q,k)\right\} \tag{2.6}$$

Following Kraichnan we note that (2.4) and (2.6) restrict contributing triads to those that form very flat triangles with k and in particular the exponential drop off for $k>>k_d$ implies $E_\rho(k)<<E_\rho(q)$ for almost all q. Thus it follows

$$2Kk^2E_\rho(k) \approx k\iint_{\Delta}(1-\omega^2)(pq)^{-1}E(p)\,k^2E_\rho(q)\Theta(p,k,q)\,dp\,dq \tag{2.7}$$

for which the asymptotic solution exists

$$E_\rho(k) \sim \left(\frac{k}{k_d}\right)^2 e^{-\alpha\frac{k}{k_d}} \tag{2.8}$$

For Schmidt numbers different from unity the solution of (2.5) appears to be vastly complicated. One suspects that for $k>>k_d$ and for $k>>k_K$ equation (2.7) may be asymptotically valid. k_K is the wave number at which scalar diffusivity becomes effective in removing scalar content[8].

The general form of (2.5) suggests that even if reaction rate is effective as a loss mechanism at high wave number, that is if $c\bar{v}_0 > \frac{1}{2}Kh^2$, an exponential type of asymptotic solution is inevitable if $E(k)$ has the exponentially decaying asymptote predicted by direct interaction. Thus the existence of mean square derivatives of all orders of the scalar field appears to be guaranteed by this hypothesis no matter what the particular combination of parameters is in a given flow.

Conclusion:

The scalar field behavior at high Reynolds numbers which has been examined, admittedly non-rigorously, in the foregoing indicates a general qualitative agreement of the consequences of direct interaction with features known or predicted by other proceedures. For the reasons developed in the previous sections the role of reactions, first or second order, is not as significant at the high wave number regimes to which we are necessarily restricted here as it must be for the scalar energy containing region and a more discriminating investigation of the differences between no reaction, first order reaction and second order reaction must depend on numerical calculations in that region. Similarly it is in this region of the spectrum that the acceptability of the direct interaction hypothesis itself must be tested. For example the guarantees of energetic consistency provided by Kraichnan's model equation for the velocity field and for the scalar mixing problem have not been extended to the nonlinear reactive case. In view of the complicated scalar "energy" decay and

growth processes which reaction has been shown to produce regions of negative spectral energy may be the consequence for certain choices of turbulence level, mean concentration and initial spectral "energy" distribution. Part III of this paper will be concerned with the dynamics of decay in the low wave number regimes as obtained by numerical integration of the scalar direct interaction equation.

Note Added in Proof

It is possible to prove realizability of the reacting scalar field by a model system very similar to the one devised by Kraichnan[9] for shear and thermally driven turbulence. Although the model allows volume-integrated exchange of reactant among individual systems the average such exchange among the systems vanishes. In this sense it is analogous to energy and entropy in the above mentioned paper.

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