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ON THE DECAY OF A REACTING SCALAR FIELD
IN TURBULENCE

by

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Use is made of the Direct Interaction Approximation, which has been developed by Kraichnan^(1,2) in his study of turbulence dynamics, to obtain a closed set of spectral equations in the case of an isotropically distributed scalar in isotropic turbulence. The scalar is assumed to be undergoing not only convective mixing and molecular diffusion, but also a reaction which is second order in the reactant concentration. To minimise other coexisting phenomena the effect of heat production due to the reaction is ignored as is any dynamic influence of the reactant or products of reaction on the carrier turbulence.

Such a system shares with the more complicated phenomenon of shear flow turbulence an interaction between a mean and a fluctuating component, and it promises to be interesting not only in its own right, since second order reactions are common, but also as a preliminary for the situation in shear flows in which one has a spatially dependent mean.

The pertinent approximation equations are presented and it is hoped that numerical solutions paralleling Kraichnan's⁽³⁾ velocity field computations can be achieved.

Although no further reference is made to the fact, it is obvious that equivalent equations for scalar mixing and for a scalar undergoing a first order reaction are easily obtainable by analogy from the second order reaction results presented here.

1. Introduction:

A promising approximation technique has been developed by Kraichnan in his work on turbulence dynamics and numerical calculations for the case of isotropically distributed and incompressible, decaying turbulence have been published⁽³⁾. There are several features of the results which give one grounds for confidence in the approximation in the low Reynold's number range to which it has been applied. In particular the shewness of the velocity field shows for all the cases examined an asymptotic value remarkably close to that obtained experimentally. The approximation has been shown to be energetically consistent and it has physical interpretations which appear reasonable. It seems to be worthwhile to pursue its predictions further.

Kraichnan⁽⁴⁾ has recently presented the formal extension of his approach to shear flow and thermally driven turbulence and is presumably seeking solutions of the system of equations so obtained. Another approach is to concentrate on scalar fields which are transported by the turbulence. These share much of the statistical complexity of the turbulence field and in fact have proven to be just as difficult a stochastic problem. In any situation in which the scalar is a particle-attached invariant interest is again focused only on the velocity field⁽⁵⁾ and its statistical properties. It is easy to show that if a first order reaction is also permitted to such a particle-attached scalar in a decaying field the problem can be transformed into the non-reacting case. The existence of a dependence of the scalar field evolution on its spatial gradients,

such as occurs when molecular diffusion becomes significant gives rise to scalar field complexities of the same order of difficulty as that presented by turbulence itself. However the formal similarities between the Navier-Stokes Equations and the scalar conservation relation are such that Kraichnan's approximation is immediately applicable. In fact the details of such a system with the additional difficulty of buoyancy induced motion have already been presented⁽⁴⁾.

Another system which introduces a further non-linearity but retains a simple relation between the velocity field dynamics and scalar field kinetics is that of a scalar being convected, diffused and simultaneously reacting to the second order in its concentration. Such reactions are not uncommon. For example collision controlled decompositions are frequently second order at low enough pressures. Generally, however, there is a thermal energy associated with the reaction which can produce buoyancy forces. If these are to be included in the analysis of, say, the decay of such a reaction the complications compound rapidly and the penalty of having two scalar fields, possibly interacting with each other and being carried simultaneously by a velocity field sensitive to both, is excessive numerical complexity and the suspicion that current solution techniques may be unequal to the task of obtaining significant results.

We therefore propose to retain dynamic passivity of the scalar undergoing the second order reaction with the hope of identifying important features of the reaction's nonlinearities.

The buoyancy term (Boussinesq approximation) retained in previous⁽⁴⁾ work gives an indication of the role of temperature induced density fluctuation within the direct interaction framework.

2. Scalar Field-velocity Field Equations

The simplest linkage between a turbulent field and a transported scalar is one in which the scalar exerts no influence on the dynamics of the turbulence. This kind of dynamic passivity has asymptotic validity in a linear scalar system for small fluctuations of the scalar and its use to amplify the velocity scalar interaction by no means emasculates the problem. It is, however, a less satisfactory assumption for the case under consideration here since if the second order chemical reacting term is to be an effective transporter of scalar "energy" in wave number space or an efficient destroyer of such "energy" the scalar must occur in greater than infinitesimal amounts. We have to assume therefore that despite the significance of the term which is second order in concentration the consequent generator of thermal energy is sufficiently low to not introduce significant buoyancy effects or to otherwise interfere dynamically with the carrier turbulence.

An immediate consequence of such a passivity assumption is that the velocity field description by the Navier Stokes equations can be considered in isolation. We will denote the Eulerian velocity field by $u_i(x, \tau)$ and assume that any of its statistical properties can be summoned at will. We specifically have in mind Kraichnan's computations for an array of initial conditions of the decay of an isotropic field of turbulence. Alternatively, for simplicity, one could postulate the impossible combination of isotropy

and nondecay of the velocity field. For our purposes, to derive the pertinent scalar equations, the only specification necessary is that the velocity field be isotropic.

There is another consequence of dynamic passivity which will be of fundamental importance in the following derivations. In Kraichnan's theory a crucial role is played by the modal response function which essentially describes how a perturbation of one mode in a dynamical system is transported through the coexisting modes. Obviously, by passivity, if the modes consist of scalar and velocity field Fourier elements the perturbation of the former will have no influence on the later and a wide group of responses can be ignored in the subsequent development. The reverse is evidently not true. Velocity field modes exert, through the inertia terms, a dominant influence over the scalar field they transport. Naturally there will also always be a response of the scalar field modes to a perturbation of one of their own number.

The complexity of the turbulence dynamics problem invariably forces on the investigator the necessity of postulating symmetries in the system under examination. Kraichnan has presented in a formal manner a set of equations in real space and time which make no such appeals. They are consequently extremely complex and presumably solution depends on simplifications which will in part be geometric. We will adopt the notion of isotropic distribution of both velocity and scalar fluctuating fields, a zero mean velocity and a spatially uniform mean concentration which will, of course, because of the nonlinear reaction be time dependent. The less restrictive assumption of homogeneity effects a great deal of simplification but for our purposes in the search for the most readily

solvable system we will in general also specify isotropy. An exception is made in an investigation of the final period of decay where classically homogeneity is sufficient for a solution.

We will take as self-evident the following equations which describe the phenomena of turbulent mixing of a scalar field which undergoes a second order reaction.

The incompressible Navier-Stokes equation (2.1),

$$\frac{\partial f(x,t)}{\partial t} = - \frac{\partial}{\partial x_i} (u_i(x,t) f(x,t)) + K \nabla^2 f(x,t) - C f^2(x,t) \quad (2.2),$$

where $f(x,t)$ is the concentration of the reacting species.

Since the Navier-Stokes equations consist of terms which are either linear or bilinear in the velocity field and the mass conservation law, equation (2.2), reveals a similar property for $f(x,t)$ we may expect that a matrix formulation will collapse these two equations into a single one. It is convenient to make use of the elaborate notational conventions of Kraichnan as used in reference (4), (hereafter referred to as K).

Thus we can write the single matrix equation in the same form as K Equation (6.8.)

$$\left(\frac{\partial}{\partial t} - \eta_{ij} \nabla^2 \right) U_j(x,t) = - \frac{1}{2} P_{ijm} (\nabla) \left[U_j(x,t) U_m(x,t) \right] - L_{ij} (\nabla) \left[U_j(x,t) \right] + F_i(x,t) \quad (2.3),$$

where the operator definitions are as given by K, Equations (6.7), (6.9), (6.10) and (6.11), with the following changes;

$$(a) \quad P_{0jm}(\nabla) = 0 \quad \text{except} \quad P_{000} = 2C, \quad P_{00i} = P_{0i0} = \frac{\partial}{\partial x_i}$$

(b) α the volume coefficient of thermal expansion is to be taken as zero.

$$\text{e.g.} \quad L_{i0}(\nabla) = 0 \quad i \neq 0$$

$$(c) \quad U_0(x, t) = f(x, t)$$

We may form the various statistical equations that can be generated from Equation(s.3) in the usual manner to obtain

$$\frac{d}{dt} \bar{U}_0(t) = -C \bar{U}_0^2(t) - C U_{00}(x, t; x, t) \quad (2.4),$$

$$\begin{aligned} \left[\frac{\partial}{\partial t} - K \nabla^2 \right] U_{00}(x, t; x', t') &= -2C \bar{U}_0(t) U_{00}(x, t; x', t') \\ &- C U_{000}(x, t; x, t; x', t') \\ &- \frac{1}{2} \frac{\partial}{\partial x_i} U_{0j0}(x, t; x, t; x', t') \\ &- \frac{1}{2} \frac{\partial}{\partial x_j} U_{i00}(x, t; x, t; x', t') \end{aligned}$$

(2.5),

and

$$\begin{aligned} \left[\frac{\partial}{\partial t} - \kappa \nabla^2 \right] G_{oo}(x, t; x', t') &= -2C \bar{U}_o(x, t; x', t') G_{oo}(x, t; x', t') \\ &\quad - 2C H_{ooo}(x, t; x, t; x', t') - \frac{\partial}{\partial x_j} H_{ojo}(x, t; x, t; x', t') \\ &\quad - \frac{\partial}{\partial x_j} H_{joo}(x, t; x, t; x', t') + \delta^3(x-x') \delta(t-t') \end{aligned}$$

(2.6),

where the simplifications of passivity and homogeneity have already been applied. That equations (2.4), (2.5) and (2.6), when coupled with the velocity field results from K, are sufficient follows from the result, which is not quite self-evident but is easy to show, that forms of the response function $G_{oi}(x, t; x', t')$; $i \neq 0$ play no role in the dynamics of the interaction under the assumptions of scalar and velocity field isotropy.

3. Final Period of Decay

A not quite trivial solution of turbulent velocity field dynamics has been obtained⁽⁶⁾ for the asymptotic regime in turbulent decay when effectively for any given wave number the inertia contribution becomes negligible compared to the diffusive effects. A similar solution exists for decay of a passive scalar field obeying a linear conservation law. For the present problem we can, by the classical argument, expect the last two terms in Equation (2.5) to become insignificant asymptotically.

If we can further ignore the other triple moment term, then the combined equations (2.4) and (2.5) form a closed set. A

sufficient condition for neglecting the second term on the right hand side of (2.5) compared to the first term on the same side would be the relationship*

$$\frac{U_{000}(x,t; x',t')}{\bar{U}_0(t) U_{00}(x,t; x',t')} = o(1) \text{ as } t, t' \rightarrow \infty \quad (3.1)$$

In the direct interaction approximation with which the subsequent sections of this report are concerned it will be seen that such a relationship becomes highly plausible, but for the moment we take it as an assumption and show that the consequences are consistent with the assumption. We find under these circumstances that mean and fluctuating scalar fields are related by

$$\frac{d\bar{U}_0(t)}{dt} = -C \bar{U}_0^2 - C U_{00}(x,t; x,t) \quad (3.2),$$

$$\left[\frac{\partial}{\partial t} - K \nabla_x^2 \right] U_{00}(x,t; x',t') = -2C \bar{U}_0(t) U_{00}(x,t; x',t') \quad (3.3).$$

A more convenient form of (3.3) which also makes explicit use of the homogeneity of $U_{00}(x,t; x',t')$ can easily be derived,

$$\left[\frac{\partial}{\partial t} - 2K \nabla_r^2 \right] U_{00}(r,t) = -4C \bar{U}_0(t) U_{00}(r,t) \quad (3.4),$$

* In the limit $x' = x; t' = t$, and assuming bounded skewness, (3.1) is equivalent to

$$\frac{[U_{00}(0,t)]^{1/2}}{\bar{U}_0(t)} = o(1) \text{ as } t \rightarrow \infty.$$

See Reference (7) Section (4.1).

where $\underline{r} = \underline{x} - \underline{x}'$ and hence (4.1) becomes

$$\frac{d}{dt} \bar{v}_0(t) = -c \bar{v}_0^2 - c v_{00}(0, t) \quad (3.5).$$

A spatial Fourier transform of the correlation function into, say, $\phi(\underline{k}, t)$ leads to an immediate formal solution of (3.4) in wave number space,

$$\phi(\underline{k}, t) = \phi(\underline{k}, t_0) e^{-2K k^2 (t - t_0) - 4c \int_{t_0}^t \bar{v}_0(t') dt'} \quad (3.6),$$

and the consequent form of the mean equation

$$\frac{d \bar{v}_0(t)}{dt} = -c \bar{v}_0^2 - c \int \phi(\underline{k}, t) d\underline{k} \quad (3.7).$$

For kinematic reasons⁽⁷⁾ the spherical shell mean of $\phi(\underline{k}, t)$ behaves as a quadratic in k near the wave number origin and since, from (3.5) for $t - t_0$ very large, only wave numbers near the origin contribute significantly to the total scalar "energy", (3.7) can be written

$$\frac{d \bar{v}_0(t)}{dt} = -c \bar{v}_0^2 - c \int \alpha k^2 e^{-2K k^2 (t - t_0) - 4c \int_{t_0}^t \bar{v}_0(t') dt'}$$

$$\alpha = \text{const}$$

from which a straight forward integration yields

$$\frac{d\bar{v}_0(\tau)}{d\tau} = -c\bar{v}_0^2(\tau) - \frac{\sqrt{\pi}}{4} \frac{d}{[2K(\tau-\tau_0)]^{3/2}} e^{-4c \int_{\tau_0}^{\tau} \bar{v}_0(\tau') d\tau'} \quad (3.8)$$

The analysis leading to (3.8) is only of asymptotic validity. Thus τ_0 is a virtual time origin about which we can have no information other than experimental and that does not appear to be available.

The final term in (3.8) is positive for all time and hence the rate of decay of $\bar{v}_0(\tau)$ can be no less rapid than inversely with time, which is the reaction-controlled behavior of a second order system.

Defining $\tau = c(t - \tau_0)$ and $\bar{v}_0(\frac{\tau}{c} + \tau_0) = \theta(\tau)$ we can differentiate (3.8) to obtain

$$\theta'' = -6\theta\theta' - 4\theta^2 - \frac{3}{2\tau}(\theta' + \theta^2) \quad (3.9)$$

Thus we can extract a particular solution

$$\theta = \frac{1}{\tau} \quad (3.10)$$

and know from (3.8) that no other solution can exhibit a less rapid decay.

Substitution of the result (3.10) into (3.8) yields two asymptotic statements;

as $t \rightarrow \infty$

$$\overline{U}_0(t) \sim \frac{1}{t - \tau_0} \quad (3.11),$$

$$\overline{U}_{00}(0,t) \sim \frac{1}{(t - \tau_0)^{1/2}} \quad (3.12),$$

We may note a comparison between (3.12) and the corresponding results for the turbulent energy decay in the final period⁽⁶⁾ and for the asymptotic rate of energy decay of a nonreacting scalar⁽⁷⁾

$$\overline{u_i u_i}(0,t) \sim \frac{1}{(t - \tau_0)^{5/2}} \quad (3.13),$$

For the nonreacting

$$\text{scalar} \quad \overline{U}_{00}(0,t) \sim \frac{1}{(t - \tau_0)^{3/2}} \quad (3.14),$$

If we now refer forward to equation (4.2) in the next section, the first term on the right hand side is the Direct Interaction approximation to the term containing $\overline{U}_{00}(x,t; x',t')$ in (2.5). Application of (3.11) and (3.12) to (4.2) indicates a consistency with the assumption (3.1). In fact the quadratic nature of the dependence of the first term on the right hand side of (4.2) on $\overline{U}_{00}(x,t; x',t')$ and the dependence of the spectrum $\phi(k,t)$ on $\overline{U}_0(t)$ as given by (3.6) strongly suggests that only the left hand side of (4.2) be retained in a final period analysis. This of course is what has been done in deriving (3.11) and (3.12).

* See also Reference (7), Equation 36.

4. The Direct Interaction Approximation

Application of Kraichnan's approximation to the incomplete set of equations (2.4), (2.5) and (2.6) is entirely straight forward and formally similar, at least in their space-time form, to the results he has quoted in K.

The approximation itself has been described in physical and mathematical detail by its author in several publications. It satisfies all of the elementary physical requirements such as realizability and consistency and appears to predict for isotropic decaying turbulence at low Reynold's Numbers several of the experimentally observed features of turbulence. It is hoped that it may prove equally as useful in predicting similar features in comparable Reynolds and Damkohler Number ranges for the problem under investigation here.

The algebraic details of the approximation as applied to the triple moment terms (those with three subscripts) in (2.5) and (2.6) can be omitted. Use is made of K equations (5.1) and (5.2) and the definitions following equation (2.3) above with the following results;

$$\frac{d \bar{U}_0(t)}{dt} = -c \bar{U}_0^2(t) - C U_{00}(\underline{x}, t; \underline{x}, t) \quad (4.1)$$

$$\left[\frac{\partial}{\partial t} - K \nabla^2 + 2c \bar{U}_0(t) \right] U_{00}(\underline{x}, t; \underline{x}', t') = -2C^2 \int_{\underline{y}}^{t'} G_{00}(\underline{x}', t'; \underline{y}, s) U_{00}^2(\underline{x}, t; \underline{y}, s) ds$$

$$- \int_{\underline{y}}^{t_0} G_{00}(\underline{x}', t'; \underline{y}, s) U_{yb}(\underline{x}, t; \underline{y}, s) \frac{\partial^2 U_{00}(\underline{x}, t; \underline{y}, s)}{\partial x_i \partial y_b} ds \quad (4.2),$$

$$\begin{aligned}
 \left[\frac{\partial}{\partial t} - \kappa \nabla^2 + 2C \bar{U}_0(t) \right] G_{00}(x, t; x', t') &= 4C^2 \int_{x'}^x \int_{t'}^t G_{00}(x, t; y, s) U_{00}(x, t; y, s) G_{00}(y, s; x', t') ds \\
 &+ \int_{x'}^x \int_{t'}^t U_{0b}(x, t; y, s) \frac{\partial}{\partial x_m} G_{00}(x, t; y, s) \frac{\partial}{\partial y_b} G_{00}(y, s; x', t') ds
 \end{aligned}
 \tag{4.3}$$

For the purposes of computation and dynamical interpretation it is convenient to transform the above set of equations into Fourier space.

A typical transformation can be demonstrated with the term

$$\int_{x'}^x \int_{t'}^t G_{00}(x, t; y, s) U_{00}^2(x, t; y, s) ds$$

which appears in (4.2)

Define

$$G_{00}(x, t; x', t') = \int_{\underline{p}} \theta(\underline{p}, t; t') e^{i \underline{p} \cdot (x - x')} d\underline{p},$$

$$U_{00}(x, t; x', t') = \int_{\underline{q}} \phi(\underline{q}, t; t') e^{i \underline{q} \cdot (x - x')} d\underline{q},$$

$$U_{0b}(x, t; x', t') = \int_{\underline{k}} \psi_{0b}(\underline{k}, t; t') e^{i \underline{k} \cdot (x - x')} d\underline{k},$$

and we obtain

$$\begin{aligned}
 & \int d\underline{y} \cdot \int_{t_0}^{t'} G_{00}(\underline{x}, t'; \underline{y}, s) U_{00}^2(\underline{x}, t; \underline{y}, s) ds \\
 &= \int d^3 \underline{y} \int_{t_0}^{t'} \int_{\underline{p}} \Theta(\underline{p}, t', s) e^{i \underline{p} \cdot (\underline{x}' - \underline{y})} d\underline{p} \cdot \int_{\underline{k}} e^{i \underline{k} \cdot (\underline{x} - \underline{y})} \int_{\underline{k}'} \phi(\underline{k}', t, s) \phi(\underline{k} - \underline{k}', t, s) d\underline{k}' \\
 &= \iiint_{\underline{p}, \underline{k}, \underline{k}', \underline{y}} d^3 \underline{y} e^{-i(\underline{k} + \underline{p}) \cdot \underline{y}} \int_{t_0}^t \Theta(\underline{p}, t', s) e^{i \underline{k} \cdot \underline{x}'} \phi(\underline{k}', t, s) \phi(\underline{k} - \underline{k}', t, s) \\
 & \quad e^{i \underline{p} \cdot \underline{x}} d\underline{k} d\underline{k}' d\underline{p} ds \\
 &= \iiint_{t_0}^{t'} \int_{\underline{k}, \underline{p}} S(\underline{k} + \underline{p}) \int_{\underline{k}'} \Theta(\underline{p}, t', s) \phi(\underline{k}', t, s) \phi(\underline{k} - \underline{k}', t, s) e^{i \underline{k} \cdot \underline{x} + i \underline{p} \cdot \underline{x}'} d\underline{k} d\underline{p} d\underline{k}' ds \\
 &= \int_{\underline{k}} e^{i \underline{k} \cdot (\underline{x} - \underline{x}')} \int_{t_0}^{t'} \int_{\underline{k}'} \Theta(-\underline{k}, t', s) \phi(\underline{k}', t, s) \phi(\underline{k} - \underline{k}', t, s) d\underline{k}' ds d\underline{k}
 \end{aligned} \tag{4.4}$$

Application of similar techniques to the other non-linear terms in the space time equations leads to the following reasonably compact description of the phenomena of isotropic turbulent mixing, diffusion and reactive decay of a reacting scalar.

$$\frac{d \bar{U}_0(t)}{dt} = -c \bar{U}_0(t) - c \int_{\underline{k}} \phi(\underline{k}, t, t) d\underline{k} \tag{4.5}$$

$$\left[\frac{\partial}{\partial t} + Kk^2 + 2L\bar{u}_0(t) \right] \phi(\underline{k}, t, t') = - \int_{t_0}^{t'} \int_{\underline{k}'} \left[\theta(-\underline{k}, t', s) \phi(\underline{k} - \underline{k}', t, s) \right. \\ \left. + [2c^2 \phi(\underline{k}', t, s) + k_j k_b \psi_{jb}(\underline{k}', t, s)] d\underline{k}' ds \right]$$

(4.6)

$$\left[\frac{\partial}{\partial t} + Kk^2 + 2L\bar{u}_0(t) \right] \theta(\underline{k}, t, t') = \int_{t'}^t \int_{\underline{k}'} \left[\theta(\underline{k}, s, t') \theta(\underline{k} - \underline{k}', t, s) \right. \\ \left. + [4c^2 \phi(\underline{k}', t, s) + k_j k_b \psi_{jb}(\underline{k}', t, s)] d\underline{k}' ds \right] \\ + \delta(t - t')$$

(4.7)

Isotropy of the various statistical functions involved in the above description implies that they are functions only of the magnitude of their vector arguments. Pursuit of this obvious simplification is postponed until an investigation of possible numerical solution techniques for the above set is undertaken. Initial conditions will be quite similar to those employed by Kraichnan⁽³⁾ in his investigation of isotropic turbulence dynamics and in fact a suitable form for the "known" velocity field function $\psi_{jb}(\underline{k}', t, s)$ could be to the numerical results reported by him.

5. Conclusion

An attempt is being made⁽⁸⁾ to apply the direct interaction approximation to the solution of the passive scalar mixing problem. Previous work⁽⁹⁾ has shown that for the freely decaying situation

the linear reaction problem could be extracted from such a solution. If it is possible to further solve the nonlinear scalar problem whose statement in Fourier space has been presented in the foregoing, then a direct comparison of spectral behavior for these two kinds of reaction kinetics will be able to be made. In view of the current lack of either theory or experiment for the freely decaying nonlinear case and the reasonable success of the Direct Interaction Approximation in predicting some specific features of low Reynolds number turbulence it appears to be well worth while to attempt to effect such a comparison.

In a recent paper Corrsin⁽¹⁰⁾ has deduced the shape, in various wave number subranges, of concentration spectra for turbulence with an isothermal, second-order reaction. Both fields are stationary and locally isotropic.

Another use to which the above formulation could be put is to attempt to predict some of the stationary states with which Corrsin has been concerned. An approximation suggestion by Kraichnan⁽¹¹⁾ for turbulent fields seems to be relevant to such an attempt.

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