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LAGRANGIAN HISTORY DIRECT INTERACTION EQUATIONS
FOR ISOTROPIC TURBULENT MIXING UNDER A
SECOND ORDER CHEMICAL REACTION

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ABSTRACT

The Lagrangian History Direct Interaction approximation is applied to isotropic turbulent mixing of a second order chemical reaction and the resulting closed sets of equations are presented. An abridgement of them is carried out and the equations which apply to a specific stationary turbulence field and a statistically specified initial scalar field are displayed. It is shown that in the limit of a stochastically distributed second order reaction the equations reduce to those of Direct Interaction.
1. INTRODUCTION

It is the purpose of this paper to derive a closed set of equations describing the behaviour of a passive reactant when its concentration is decaying due to an isothermal reaction of second order and when it is simultaneously advected by a turbulent fluid. We propose to make use of the Lagrangian History Direct Interaction (L.H.D.I.) closure which was devised by Kraichnan\(^1\) for his studies of the dynamics of turbulence and turbulent convection. The approximation has been applied to other statistical problems in which there exist non-zero means\(^2\) and it appears to be the most promising of existing closures for dealing with this class of phenomena. In a previous work\(^3\) another closure scheme called the Direct Interaction (D.I.) approximation, which is also due to Kraichnan, has been employed to obtain a closed set of equations for this same reactive situation. It is our ultimate purpose to compare the performance of each closure under certain quantitative tests that can be devised, but the immediate objective to which this report is limited is to present the equations which result from applying the L.H.D.I. approximation and also to derive an abridged form of them.
2. THE DIRECT INTERACTION EQUATIONS

L.H.D.I. equations have been obtained for the non-reactive passive scalar field in isotropic turbulence\(^1\). Subsequent reference to this paper will be by the letter K and, where appropriate, an equation number taken from that paper. For example, the non-reacting scalar field equations using a generalized field is given by

\[
\frac{3}{\partial t} - D \nabla^2 \phi(x, t) = -u(x, t|t) \cdot \nabla \phi(x, t) \quad \text{K}(2.9),
\]

\[
\phi(x, t|t) = \phi(x, t) \quad \text{K}(2.10),
\]

\[
\frac{\partial \phi}{\partial t}(x, t|t) = -u(x, t|t) \cdot \nabla \phi(x, t|t) \quad \text{K}(2.11),
\]

where \(D\) is the kinematic diffusivity, and \(u(x, t|t)\) is the generalized velocity field which may be defined as the velocity measured at time \(t\) within the fluid element which passes through the point \(x\) at time \(t\). Similarly, \(\phi(x, t|t)\) is the value of the scalar field (e.g. concentration) measured at time \(t\) in the fluid element which passes through the point \(x\) at time \(t\).

The appropriate equations for the generalized concentration field \(\phi\) in the case of a second order reaction are \(\text{K}(2.10), \text{K}(2.11)\) and the following replacement for \(\text{K}(2.9)\)

\[
\frac{3}{\partial t} - D \nabla^2 \phi(x, t) = -u(x, t|t) \cdot \nabla \phi(x, t) - c \phi^2(x, t) \quad (2.1),
\]

It is pertinent to decompose the field \(\phi(x, t|t)\) into a mean and a fluctuating component

\[
\phi(x, t|t) = \bar{\phi}(t) + \gamma(x, t|t) \quad (2.2),
\]

where statistical homogeneity has been assumed.
The propagation from $(x, t)$ to $(x', t')$ is described by

\[ (\xi - x) \frac{\partial}{\partial \xi} (\xi - x) \frac{\partial}{\partial \xi} = \left( \frac{\partial}{\partial x} \right) \left( \frac{\partial}{\partial x} \right) \]

\[ = \left( \frac{\partial}{\partial x} \right) \left( \frac{\partial}{\partial x} \right) \]

According to the equation,

\[ \left( \frac{\partial}{\partial x} \right) \left( \frac{\partial}{\partial x} \right) = \left( \frac{\partial}{\partial x} \right) \left( \frac{\partial}{\partial x} \right) \]

resulting that the perturbation properties from $(x, t)$ to $(x, t)$ are given by the solution to

where the symbol $\delta$ denotes a functional derivative. Hence we obtain the

\[ \frac{\partial}{\partial \xi} (\xi - x) \frac{\partial}{\partial \xi} = \left( \frac{\partial}{\partial x} \right) \left( \frac{\partial}{\partial x} \right) \]

As defined by analogy with $(\xi, t)$ and $\xi(t)$ of the generalized Green's function for the interaction of perturbations at the scalar field is non-linear and the following equations describing the

In contrast to the non-linear case (60), the equation (7.4) for

\[ \frac{\partial}{\partial \xi} (\xi - x) \frac{\partial}{\partial \xi} = \left( \frac{\partial}{\partial x} \right) \left( \frac{\partial}{\partial x} \right) \]

\[ = \left( \frac{\partial}{\partial x} \right) \left( \frac{\partial}{\partial x} \right) \]

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The following set of equations:

The substitution of (2.2) into (2.1), (K.2.10) and (K.2.11) produces

3.
and, finally, it propagates from \((r,r)\) to \((t,r)\) according to

\[
\frac{\partial \hat{G}}{\partial t} (x,t|\tau;x',t'|r') = -u(x,t|\tau) \cdot \nabla_x \hat{G}(x,t|\tau;x',t'|r') \tag{2.10}
\]

Similarly, it is easy to deduce from (2.4) and (2.5) that the covariance \(\psi(x,t|\tau;x',t'|r')\), which is defined by

\[
\psi(x,t|\tau;x',t'|r') = \langle \gamma(x,t|\tau) \gamma(x',t'|r') \rangle,
\]
obeys the following equations:

\[
\left( \frac{\partial}{\partial t} - D \nabla_x^2 + 2c\Gamma(t) \right) \psi(x,t|\tau;x',t'|r') = -\langle u(x,t|\tau) \cdot \nabla_x \gamma(x,t|\tau) \gamma(x',t'|r') \rangle
\]

\[
-\langle \gamma(x,t|\tau) \gamma(x',t'|r') \rangle \tag{2.11}
\]

\[
\frac{\partial}{\partial t} \psi(x,t|\tau;x',t'|r') = -\langle u(x,t|\tau) \cdot \nabla_x \gamma(x,t|\tau) \gamma(x',t'|r') \rangle \tag{2.12}
\]

where \(\langle \rangle\) denotes an ensemble average.

It is also of use to have available the complimentary equations of (2.9) and (2.10). These follow precisely as in K (appendix B). The propagation from \((r',r')\) to \((r,r)\) obeys the equation

\[
\left( \frac{\partial}{\partial t} + D \nabla_x^2 \right) \hat{G}(x,t|\tau;x',t'|r') + 2c\Gamma(t) \hat{G}(x,t|\tau;x',t'|r')
\]

\[
= -u(x',t'|\tau) \nabla_x \hat{G}(x,t|\tau;x',t'|t')
\]

\[
-2c\gamma(x',t'|\tau) \hat{G}(x,t|\tau;x',t'|t') \tag{2.13}
\]

whereas, for example, from \((r,r)\) to \((t,r)\) the perturbation is described by

\[
\frac{\partial}{\partial t} \hat{G}(x,t|\tau;x',t'|r') = -u(x',t'|\tau) \cdot \nabla_x \hat{G}(x,t|\tau;x',t'|r') \tag{2.14}
\]

An ensemble-averages Green's function is defined by
\[ G(x, t | r; x', t' | r') = \langle \hat{G}(x, t | r; x', t' | r') \rangle \]

and a useful summary of the previous equations (2.6) to (2.14) is the following:

\[
\left( \frac{\partial}{\partial t} - D \frac{\partial^2}{\partial x^2} + 2c \rho(t) \right) \psi(x, t | r; x', t' | r') = S(x, t | r; x', t' | r') + R(x, t | r; x', t' | r') \tag{2.15},
\]

\[
\frac{\partial}{\partial t} \psi(x, t | r; x', t' | r') = S(x, t | r; x', t' | r') \tag{2.16},
\]

\[
\left( \frac{\partial}{\partial t} - D \frac{\partial^2}{\partial x^2} + 2c \rho(t) \right) G(x, t | r; x', t' | r') = \tilde{H}(x, t | r; x', t' | r') + Q(x, t | r; x', t' | r') \tag{2.17},
\]

\[
\frac{\partial}{\partial t} \psi(x, t | r; x', t' | r') = \tilde{H}(x, t | r; x', t' | r') \quad \text{if } t \geq r' \tag{2.18},
\]

\[
\left( \frac{\partial}{\partial t} + D \frac{\partial^2}{\partial x^2} + 2c \rho(t) \right) G(x, t | r; x', t' | t') = \tilde{H}^+(x, t | r; x', t' | t') + Q^+(x, t | r; x', t' | t') \tag{2.19},
\]

\[
\frac{\partial}{\partial t} \psi(x, t | r; x', t' | r') = \tilde{H}^+(x, t | r; x', t' | r') \quad \text{if } r \geq r' \tag{2.20}.
\]

S, \tilde{H} and \tilde{H}^+ are as defined in K (5.5), (5.6a) and (5.6b).

That is,

\[
S(x, t | r; x', t' | r') = - \langle u(x, t | t) \cdot \nabla_x \psi(x, t | r) \psi(x', t' | r') \rangle,
\]

\[
\tilde{H} (x, t | r; x', t' | r') = - \langle u(x, t | t) \cdot \nabla_x G(x, t | r; x', t' | r') \rangle,
\]

\[
\tilde{H}^+(x, t | r; x', t' | r') = - \langle u(x', t' | t') \cdot \nabla_x \tilde{G}(x, t | r; x', t' | r') \rangle.
\]

Also

\[
R(x, t | t; x', t' | r') = - c \langle u(x, t | t) \cdot \nabla_x \gamma(x, t | r) \gamma(x', t' | r') \rangle
\]

\[
Q(x, t | r; x', t' | r') = - 2c \langle u(x, t | t) \gamma(x, t | r) G(x, t | r; x', t' | r') \rangle
\]

\[
Q^+(x, t | r; x', t' | r') = - 2c \langle u(x', t' | t') \tilde{G}(x, t | r; x', t' | r') \rangle
\]
The direct Interaction approximations for $S$, $H$ and $H^+$ are fully displayed in $K(5.9),(5.10)$ and will not be repeated here. The same approximation can be applied immediately to $R$, $Q$ and $Q^+$ using the above definitions and Kraichnan's construction. One finds

$$R(x, t|r;x', t'|r') = 4c^2/fd^3yds(x, t|r;y,s)sG(x, t|r;y,s)sG(x', t'|r';y,s)s$$

$$+ 2c^2/fd^3yds(x, t|r;y,s)s\psi(x, t|r;y,s)sG(x', t'|r', y,s)s$$

$$G(x, t|r;x', t'|r') = 4c^2/fd^3yds(x, t|r;y,s)sG(x, t|r;y,s)sG(y,s)s$$

$$Q(x, t|r;x', t'|r') = 4c^2/fd^3yds(x, t|r;y,s)sG(x, t|r;y,s)sG(y,s)s$$

(2.21)

(2.22)

$Q^+(x, t|r;x', t'|r')$ is obtained from $Q(x, t|r;x', t'|r')$ by a formal substitution into (2.22) to obtain $Q(x', t'|r', x, t|r)$ and then change every $G$ function in the resulting equation according to the rule

$$G(z_1, t_1|r_1; z_2, t_2|r_2) + G(z_2, t_2|r_2; z_1, t_1|r_1).$$

To the above set of equations it is necessary to append that which describes the behaviour of the mean concentration. From (2.3) we have

$$\frac{d}{dt} \Gamma(t) = -c\Gamma^2(t) - c\psi(x, t|t; x, t).$$

(2.23)

A significant formal simplification of the previous set of equations occurs when we invoke isotropy for both the velocity and scalar fields. It is also very convenient to introduce Fourier transforms of the dependent variables $\psi, G, S, R, H, H^+, Q$ and $Q^+$. To avoid a proliferation of symbols the transformed variable will be denoted by an explicit display of the wave number $k$ as an independent variable replacing $x - x'$. For example, by invoking isotropy the Fourier transform of $\psi(x, t|r;x', t'|r')$ is written $\psi(k, t|x, t'|r').$
Equations (2.15) – (2.23) appear as follows:

\[
\frac{\partial}{\partial t} \psi(k,t|t,t'|r') = S(k,t|t,t'|r') + R(k,t|t,t'|r')
\]  
\(2.24\),

\[
\frac{\partial}{\partial t} \psi(k,t|t,t'|r') = S(k,t|t,t'|r')
\]  
\(2.25\),

\[
\frac{\partial}{\partial t} \Gamma(k,t|t,t'|r') = H(k,t|t,t'|r') + Q(k,t|t,t'|r')
\]  
\(2.26\),

\[
\frac{\partial}{\partial t} G(k,t|t,t'|r') = H^+(k,t|t,t'|t') + Q^+(k,t|t,t'|t'), r > t'
\]  
\(2.27\),

\[
\frac{\partial}{\partial t} G(k,t|t,t'|r') = H^+(k,t|t,t'|t') + Q^+(k,t|t,t'|t'), r > t'
\]  
\(2.28\),

\[
\frac{\partial}{\partial t} G(k,t|t,t'|r') = H^+(k,t|t,t'|t') + Q^+(k,t|t,t'|t'), r > t'
\]  
\(2.29\),

where

\[
S(k,t|t,t'|r') = \pi \int_{t_0}^{t'} ds_1 \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,t',s)
\]

\[
+ \pi \int_{t_0}^{t'} ds_1 \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,t',s)
\]

\[
- \pi \int_{t_0}^{r} ds_1 \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,s|s)
\]

\[
- \pi \int_{t_0}^{r} ds_1 \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,s|s)
\]

\[
R(k,t|t,t'|r') = 4\pi c^2 \int_{t_0}^{r'} ds \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,t',s)
\]

\[
+ 8\pi c^2 \int_{t_0}^{r} ds \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,s|s)
\]

\[
H(k,t|t,t'|r') = -\pi \int_{t_0}^{r'} ds \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,t',s)
\]

\[
- \pi \int_{t_0}^{r} ds \frac{1}{(1-w^2)U(p,t|t,s|s)} \psi(q,t|t,t',r') \psi(p,t|t,s|s)
\]
\[
\begin{align*}
\text{\textit{I}}&= -\pi k \int ds \int G(k,s|x,t|r') G(q,t|s|s) (1-\nu^2) U(p,t|s|s) \, dq \, dp \\
\end{align*}
\]  
(2.32)

\[
Q(k,t|r,t'|r') = 8\pi c^2 k^{-1} \int ds \int G(K,s|s,t'|r') G(q,t|r,s|s) S(p,t|r,s|s) \, dq \, dp \\
\]  
(2.33)

and the symbol \(\int\) means integration over all triangles \(p + q = \zeta\).

\(H^+(k,t|r,t'|r')\) is obtained by forming \(H(k,t'|r',t|r)\) through formal substitution into (2.32) and then, in the result, changing every \(G\) function according to the rule

\[
G(k,t_1|r_1, t_2|r_2) \rightarrow G(k,t_2|r_2, t_1|r_1).
\]

The same sequence of operations applies to forming \(Q^+(k,t'|r',t|r)\) from \(Q(k,t|r,t'|r')\).

We also have

\[
\frac{d}{dt} \bar{\rho}(t) = -c^2 \bar{\rho}(t) - 4\pi c^2 \int_0^\infty \psi(k,t,t,t) \, dk.
\]

(2.34)

In equations (2.30) and (2.32) we have written the Fourier transform of the velocity covariance \(<u_i(x,t|t)u_j(x',s|s)>\) in terms of the scalar \(U(k,t|t,s|s)\) to which it is related by

\[
U_{ij}(k,t|t,s|s) = \frac{1}{2} \left( \delta_{ij} - \frac{k_i k_j}{k^2} \right) U(k,t|t,s|s)
\]

It should be noted that in so doing we have assumed that \(U_{ij}\) is solenoidal in both indices \(i\) and \(j\). This is not necessarily true as Kraichnan has carefully pointed out. However, in transforming these equations to the L.H.D.I. approximation, part of the prescription for the transformation is that \(U_{ij}\) be solenoidal in both indices and we assume it here prematurely simply for convenience.
3. THE LAGRANGIAN HISTORY DIRECT INTERACTION EQUATIONS

The L.H.D.I. equations can be written down immediately from those of Direct Interaction (2.24)-(2.34) by making use of Kraichnan's prescription for such a transformation. As we mentioned at the end of Section 2, part of the prescription has already been applied to the derivation of (2.30) and (2.32). The remainder of it requires that in (2.30) to (2.33) every labelling time $s$ should be changed to $t$ if it arises from the perturbation expansion of a factor in (2.15)-(2.20) with a labelling time $t$ or to $t'$ if it arises from expansion of a factor with labelling time $t'$ (the labelling times are the time arguments which precede the vertical bars).

The consequences of these operations are that (2.30)-(2.33)

transform to

$$S(k,t|t,t'|r) = \pi k \int_{t'}^{r'} ds \int dq \int dp \, (1-\omega^2)U(p,q,t,t'|s)G(k,t'|r,t'|s)U(q,t,r,t'|s)$$

$$+ \pi k \int_{t'}^{r'} ds \int dq \int dp \, (1-\omega^2)U(p,q,t,t'|s)\psi(q,t,r,t'|r')$$

$$- \pi k \int_{t'}^{r'} ds \int dq \int dp \, (1-\omega^2)U(p,q,t,t|s)G(q,t,r,t|s)\psi(k,t'|r',t|s)$$

$$- \pi k \int_{t'}^{r'} ds \int dq \int dp \, (1-\omega^2)U(p,q,t,t|s)G(q,t,t|s) \phi(k,t'|r',t|r)$$

(3.1),

$$R(k,t|t,t'|r') = 8\pi c^2 k^{-1} \int_{t'}^{r'} ds \int dq \int dp \, \psi(p,t|t,t'|s)\psi(k,t'|r',t|s)G(q,t,r,t|s)$$

$$+ 4\pi c^2 k^{-1} \int_{t'}^{r'} ds \int dq \int dp \, \psi(p,t|t,t'|s)\psi(q,t,r,t'|s)G(k,t'|r',t'|s)$$

(3.2),

$$H(k,t|t,t'|r') = -\pi k \int_{t'}^{r'} ds \int G(k,t|t',t'|r')G(q,t|r,t'|r') (1-\omega^2)U(p,q,t,t|s)$$

$$- \pi k \int_{t'}^{r'} ds \int G(k,t|t',t'|r')G(q,t|r,t|s) (1-\omega^2)U(p,q,t,t|s)$$

(3.3).
\[ -\pi k \int ds \int_{pq} dp \, dq \, G(k, t | r, t' | r') (1 - \nu^2) U(p, t | t, t | s) \]  
\[ \phi^+(k, t | r, t' | r') = -\pi k \int ds \int_{pq} dp \, dq \, G(k, t | r, t' | r') G(q, t | r, t' | r') (1 - \nu^2) U(p, t' | t', s) \]  
\[ -\pi k \int ds \int_{pq} dp \, dq \, G(q, t | s, t' | r') (1 - \nu^2) U(r, t | t, t | s) \]  
\[ -\pi k \int ds \int_{pq} dp \, dq \, G(k, t | r, t' | r') (1 - \nu^2) U(p, t' | t', t' | s) \]  
\[ Q(k, t | t, t' | r') = 8 \pi c^2 k^{-1} \int_{pq} dp \, dq \, G(k, t | s, t' | r') G(q, t | r, t | s) \phi(p, t | r, t | s) \]  
\[ Q^+(k, t | r, t' | r') = 8 \pi c^2 k^{-1} \int_{pq} dp \, dq \, G(k, t | r, t' | s) G(q, t' | s, t' | r') \phi(p, t' | r', t' | s) \]  

It is also necessary to prescribe how the integration of (2.24)-(2.29) and (2.34) should proceed. It is easy to show that the prescription given in I (appendix C) is still pertinent for these extended equations. We will not pursue the matter further in this paper as we wish now to derive an abridged form of the L.H.D.I. equations and to present them in a form suitable for the calculation of the behaviour of a specific initial scalar spectrum in a specified stationary isotropic turbulent field.
4. THE ABRIDGED LAGRANGIAN HISTORY DIRECT INTERACTION EQUATIONS

We first obtain equations for the pure Lagrangian functions which, by definition, are those in which all labelling times are equal. From (2.24) to (2.29) and (2.34) we have,

\[ \frac{d}{dt} \chi(t) = -\epsilon \chi^2 - 4\pi \epsilon^2 \int_0^\infty k \psi(k, t|t, t|t) \, dk \]  
(4.1)

\[ \left( \frac{\partial}{\partial t} + 2Dk^2 + 4c\chi(t) \right) \psi(k, t|t, t|t) = 2S(k, t|t, t|t) + 2R(k, t|t, t|t) \]  
(4.2)

\[ \left( \frac{\partial}{\partial t} + Dk^2 + 2c\chi(t) \right) \psi(k, t|t, t|r) = S(k, t|t, t|r) + S(k, t|t, t|r) + R(k, t|t, t|r) \]  
(4.3)

\[ \frac{\partial}{\partial t} \psi(k, t|x, t'|r') = S(k, t|x, t'|r') + S(k, t'|r', t|r) \]  
(4.4)

\[ G(k, t|r, t|r) \equiv 1 \]  
(4.5)

\[ \left( \frac{\partial}{\partial t} + Dk^2 + 2c\chi(t) \right) G(k, t|t, t|r) = H(k, t|t, t|r) + H^+(k, t|t, t|r) + Q(k, t|t, t|r) \]  
(4.6)

\[ \frac{\partial}{\partial t} G(k, t|r, t'|r') = H(k, t|r, t'|r') + H^+(k, t|r, t'|r') \]  
(4.7)

The abridgement is realised by employing the following approximations or transformations.

\[ \psi(k, t|t, t|t_2) \rightarrow \psi(k, r_1|t_1, t|t_2) \quad r_1 > r_2 \]
\[ \psi(k, r_2|t_2, t|t_1) \quad r_2 > r_1 \]
\[ G(k, t|r_1, t|t_2) \rightarrow G(k, r_1|t_1, t_2|t) \]
\[ \psi(k, t|r_1, t|t_2) \rightarrow \psi(k, r_1|t_1, r_1|t_2) \quad r_1 > r_2 \]
\[ \psi(k, r_2|t_2, r_1|t_1) \quad r_2 > r_1 \]
Before displaying the consequences of this approximation, it is useful to introduce some notational shorthand.

\[ U(k, t| r) = U(k, t| t, t| r) \quad t > r \]
\[ = U(k, r| r, t| t) \quad t < r \]
\[ G(k, t| r) = G(k, t| t, t| r) \]
\[ \psi(k, t| r) = \psi(k, t| t, t| r) \quad t > r \]
\[ \psi(k, r| r, t| t) \quad t < r \]

We finally have

\[ \frac{d}{dt} \overline{G}(t) = -c^2 \overline{G}(t) - 4\pi \int_0^\infty k^2 \psi(k, t| t) dk \quad (4.8), \]
\[ \frac{\partial}{\partial t} + 2Dk^2 + 4c \overline{G}(t) \psi(k, t| t) = 2S(k, t| t) + 2\Psi(k, t| t) \quad (4.9), \]
\[ \frac{\partial}{\partial t} + Dk^2 + 2c \overline{G}(t) \psi(k, t| r) = S(k, t| r) + S(k, r| t) + \Psi(k, t| r) \quad t > r \quad (4.10), \]
\[ \frac{\partial}{\partial t} + Dk^2 + 2c \overline{G}(t) G(k, t| r) = \Psi(k, t| r) + \Psi^+(k, t| r) + Q(k, t| r) \quad t > r \quad (4.11), \]

\[ G(k, t| t) \equiv 1 \quad (4.12), \]

where

\[ S(k, t| r) = -\pi \int_0^t d\xi \int_0^\infty (1-w^2) U(p, t| s) G(q, t| s) \psi(k, t| s) \quad pq \quad dp \quad dq \]
\[ +\pi \int_0^t d\xi \int_0^\infty (1-w^2) U(p, r| s) G(q, r| s) \psi(q, t| s) \quad pq \quad dp \quad dq \]
\[ +\pi \int_0^t d\xi \int_0^\infty (1-w^2) U(p, t| r) \psi(q, t| r) \quad pq \quad dp \quad dq \quad (4.13), \]

\[ S(k, r| t) = -\pi \int_0^r d\xi \int_0^\infty (1-w^2) U(p, t| s) G(q, r| s) \psi(k, t| s) \quad pq \quad dp \quad dq \]
\[ +\pi \int_0^t d\xi \int_0^\infty (1-w^2) U(p, t| s) G(k, t| s) \psi(q, r| s) \quad pq \quad dp \quad dq \]
\[ +\pi \int_0^t d\xi \int_0^\infty (1-w^2) U(p, t| r) \psi(q, t| r) \quad pq \quad dp \quad dq \]
The set of equations (4.8)-(4.18) form a complete set which enables pure Lagrangian quantities to be calculated when $U(k|s), \psi(k,t|t_o)$ and $\bar{u}(t_o)$ are specified.

The Direct Interaction equations of this same problem for purely Eulerian quantities have been presented elsewhere by Lee. It is important to notice that in the case of zero turbulence the problem reduces to that of a stochastically distributed second order reactant and the A.L.H.D.I. equations and the D.I. equations reduce to identical statements. This is of course proper since in the absence of motion Eulerian and Lagrangian quantities are indistinguishable. It also means that previous investigations of the stochastically distributed reactant case remain valid in the A.L.H.D.I. approximation. Finally, it is easy to see by retracing the derivations of
of section 3 and 4 that in the same limit of zero turbulence the L.H.D.I. approximation coincides identically with the A.L.H.D.I. approximation. Perhaps the easiest way to indicate why this occurs is to notice that 
labelling times play only a parametric role in the expressions for R and Q.

5. A SPECIFIC EXAMPLE OF THE A.L.H.D.I. EQUATIONS

In this section we present the A.L.H.D.I. equations as they apply to a particular class of stationary isotropic velocity fields and an initial scalar spectrum. It is our purpose ultimately, but not in this manuscript, to compute numerically the spectral evolution for just this situation. We will be particularly interested in the limit of no molecular diffusion (D = 0) as a very discriminating test of the approximation seems to be possible there and the following definitions and choices of dimensional groups are made with these requirements in mind.

Let \( \hat{k} = \frac{k}{k_0} \), \( \hat{t} = \Phi(t)ct \), \( N_{da} = \frac{c\Phi(o)}{\frac{Dk^2}{k_0}} \), \( N = \frac{u}{c\Phi(o)} \),

\[ \hat{\psi}(k,t|t) = \frac{k_0^3}{\gamma^2(o)}\psi(k,t|t), \hat{\bar{\psi}}(t) = \frac{\Phi(t)}{\Phi(o)}, \alpha = \frac{\Phi(o)}{\gamma(o)}k, \]

\[ U(k,t|r) = \frac{2u^2}{\pi^{2/2}k_o^5}k^2 \exp \left\{ -\left(\frac{k}{k_c}\right)^2 - \frac{1}{2}\left(\frac{u^2k(t-r)}{k_0}\right)^2 \right\}, \]

\[ \hat{\psi}(k,o|o) = (2\pi)^{-3/2} \exp \left\{ -\frac{1}{2}\left(\frac{k}{k_o}\right)^2 \right\} \] and \( \Phi(o) \equiv 1 \).

where \( u^2 \) is the mean square kinetic energy of the turbulence in any component direction. A detailed discussion of these spectra can be found in the literature, although it must be remembered that the velocity function \( U(k,t|r) \) is now Lagrangian rather than Eulerian and the stationarity evident in its
definition above is a Lagrangian stationarity. That any real turbulent field would exhibit approximate Lagrangian stationarity is highly unlikely, but it is not our purpose here to use the equations to examine a real situation. There are some general properties that the equations should exhibit for any kinematically allowable velocity field and it is these properties for which we will be testing subsequently. If the equations faithfully preserve these general properties, it may then be pertinent to try to establish more reasonable Lagrangian velocity fields and to explore their role in mixing and in enhancing reaction rates.

Equations (4.6)-(4.18) can now be rewritten in the light of the above definitions. In displaying the results below we have omitted the circumflex (') throughout.

\[
\frac{d\overline{\Gamma}}{dt} = -\overline{\Gamma}(t) - \frac{4\pi}{\alpha^2} \int_0^\infty k^2 \psi(k,t|t) \, dk \tag{5.1}
\]

\[
\left( \frac{\partial}{\partial t} + \frac{2k^2}{N Da} \right) \overline{T}(t) \psi(k,t|t) = 2S(k,t|t) + 2R(k,t|t) \tag{5.2}
\]

\[
\left( \frac{\partial}{\partial t} + \frac{k^2}{N Da} \right) \psi(k,t|t) = S(k,t|t) + S(k,t|t) + R(k,t|t) \tag{5.3}
\]

\[
\left( \frac{\partial}{\partial t} + \frac{k^2}{N Da} \right) G(k,t|t) = H(k,t|t) + H^+(k,t|t) + Q(k,t|t) \tag{5.4}
\]

\[
G(k,t|t) \equiv 1 \tag{5.5}
\]

where
\[
S(k,t|r) = \int ds \, G(k,r|s) \int_0^\infty P(q,k,t) \psi(q,t|s) dq \tag{5.6}
\]

\[
+ \int_0^t ds \, P(q,k,t) \psi(q,t|r) dq - \int ds \, \int_0^\infty P(q,k,t) \delta(q,t|s) dq \psi(k,t|s)
\]
\[ S(k,r|t) = -\int_0^r ds \int_0^\infty dq G(q,r|s) dq \psi(k,r|s) \]
\[ - \int_r^\infty ds \int_0^\infty dq G(q,r|s) dq \psi(k,r|s) \]
\[ + \int_0^\infty ds \int_0^\infty dq G(q,r|s) dq \psi(k,r|s) \]
\[ (5.7) \]

\[ R(k,t|r) = \frac{4\pi}{a^2 k} \int_0^r ds \int_0^\infty dq \psi(p,t|s) \psi(q,t|s) G(k,r|s) \]
\[ + \frac{8\pi}{a^2 k} \int_0^r ds \int_0^\infty dq \psi(p,t|s) \psi(q,t|s) \psi(k,r|s) \]
\[ (5.8) \]

\[ H(k,t|r) = \int_0^t \int_0^r ds G(q,t|r) dq \]
\[ - \int_0^\infty \int_0^r ds G(q,s|r) dq \]
\[ (5.9) \]

\[ H^+(k,t|r) = \int_0^t \int_0^r ds G(q,s|r) dq \]
\[ - \int_0^\infty \int_0^t dq G(k,t|r) \]
\[ (5.10) \]

\[ Q(k,t|r) = \frac{8\pi}{a^2 k} \int_0^t ds \int_0^{k+p} dq \psi(p,t|s) G(q,t|s) G(k,s|r) \]
\[ (5.11) \]

\[ P(q,k,t) = 2N^2 \pi^2 q^2 \left( e^{-2k^2 q^2} + \frac{e^{-2k^2 q^2}}{2\tau^2} + \frac{e^{-2k^2 q^2}}{4k^2 q^2} \right) \exp\left(-k^2 q^2 \tau\right), \]

and \( \tau = 1 + \frac{1}{2} N^2 (t - s)^2 \).

The integration of such a set of equations would proceed in exactly the same way as that developed for the D.I. equations with one important exception. In the D.I. equations the diagonal terms in the \((t,t')\) matrix are computed by using the symmetry of \(S(k,t,t')\) in \(t\) and \(t'\). For example, if
\( S(k, t + \Delta t, t) \) is known so is \( S(k, t, t + \Delta t) \) to which it is identically equal. Then the D.I. equations specifically state how \( S(k, t + \Delta t, t + \Delta t) \) is to be computed from \( S(k, t, t + \Delta t) \). However, the A.L.H.D.I. equation for \( S(k, t | t') \) is only valid for \( t > t' \) so that although symmetry in \( t \) and \( t' \) still holds, it is of no value to the computational program. Instead explicit use must be made of (5.2) whereby \( S(k, t + \Delta t, t + \Delta t) \) can be directly computed once \( S(k, t, t) \) and \( S(k, t + \Delta t, t) \) are established. With this exception a computing program for the A.L.H.D.I. equations can be based on that developed by Lee\(^3,4\) for the D.I. equations.

A subsequent report will give more detail on certain specific computational programs designed to examine critically some important consequences of the above A.L.H.D.I. equations.
6. AN INVARIANCE PROPERTY OF THE L.H.D.I. EQUATIONS

An important property of the exact equations which can be demonstrated easily from (2.1) and which is immediately evident on physical grounds is that when \( D \equiv 0 \) all single point functions of the concentration field decay at a rate independent of the turbulence. Since the turbulence plays no direct role in decreasing scalar intensity, but only alters the scale of the concentration field, and since the reaction term has no dependence on scale, it is evident that in the absence of molecular diffusion, turbulence does not play even an indirect role in the dynamics of decay of single point functions. This fact is another significant invariance which any proposed closure approximation should necessarily satisfy. It has not been consciously built into the L.H.D.I. approximation or into any other so that it is pertinent to ask of any closure whether it does in fact satisfy this further invariance.

We have shown elsewhere\(^5\) by numerical computation that Direct Interaction is seriously inadequate in this regard, which is perhaps not surprising since it has already been shown\(^1\) to fail to preserve another related velocity field property - Galilean Invariance.

In this section we prove that the L.H.D.I. approximation does in fact guarantee the exact preservation of the invariance described above. Specifically we show that when \( D \equiv 0 \) \( \bar{\Gamma}(t) \), \( \bar{\gamma}^{\infty}(t) \) and \( \bar{\gamma}^T(t) \) are described by closed sets of equations to which the velocity field makes no contribution. We first demonstrate an important property of the function \( G(k,t|\tau,\tau') \) which is preserved by the L.H.D.I. closure. Namely, when \( D \equiv 0 \)

\[
G(k, t|\tau, \tau') = G(\tau|\tau')
\]

(6.1)

Since \( G(\bar{x}, t|\tau, \bar{x}', \tau'|\tau') \) is the response measured at time \( \tau \) in the fluid element which is at \( \bar{x} \) at time \( t \) to a perturbation in the scalar field...
applied at time \( r' \) to the fluid element which is at \( x' \) at time \( t' \) and since the only mechanism which transfers scalar quantity from one element to another is molecular diffusion then, when \( D = 0 \), the following should be true:

\[
G(x, t | x', t') = \delta(x - x') G(r | r')
\]

and (4.1) follows.

Now the L.H.D.I. equations for \( G(k, t | r, t | r') \) is, from (2.27) and (2.29),

\[
\frac{\partial}{\partial t} G(k, t | r, t | r') = H(k, t | r, t | r') + H^+(k, t | r, t | r'),
\]

which is identically satisfied by (6.1) independently of the velocity field spectral description.

The equation for \( \gamma(t) \) can be obtained by integration of (4.2) overall wave numbers. In the absence of molecular diffusion the result is

\[
\left( \frac{3}{3t} + 4c\kappa(t) \right) \gamma(t) = 2 \int k^2 R(k, t | t, t | t) dk
\]

(6.2)

where the invariance \( K(5.13) \) has been used to eliminate the direct effects of the velocity field. Insofar as (6.2) contains the \( G \) function in \( R(k, t | t, t | t) \) the indirect role of velocity influence on \( \gamma(t) \) is possible. However, we now demonstrate that it does not in fact arise.

Using (3.2), (6.2) becomes

\[
\left( \frac{3}{3t} + 4c\kappa(t) \right) \gamma(t) = 12\pi c^2 \int \psi(p, t | t, t | s) G(t | s),
\]

or

\[
\left( \frac{3}{3t} + 4c\kappa(t) \right) \gamma(t) = 12\pi c^2 \int k^2 \psi(k, t | t, t | s) dk \int k^2 \psi(p, t | t, t | s) dp dq G(t | s).
\]

Hence

\[
\left( \frac{3}{3t} + 4c\kappa(t) \right) \gamma(t) = 12\pi c^2 \int \gamma^2(t | s) G(t | s),
\]

(6.3)
where the generalization \( \overline{\gamma}(t|s) = \int_0^\infty k^2 \psi(k,t|t,t|s) dk \) has been made and where \( \overline{\gamma}(t) \) has been written \( \overline{\gamma}(t|t) \) for notational convenience.

The equation for \( \overline{\gamma}(t|r) \) is, from (4.3),

\[
\left( \frac{\partial}{\partial t} + 2c\Gamma(t) \right) \overline{\gamma}(t|r) = 8\pi c^2 \int_{t_0}^t ds \overline{\gamma}(t|s) \overline{\gamma}(r|s) G(t|s) + 4\pi c^2 \int_{t_0}^r ds \overline{\gamma}^2(t|s) G(r|s)
\]

(6.4).

Finally, the equation for \( G(t|r) \) is obtained from (4.6) and (3.5).

We have

\[
\left( \frac{\partial}{\partial t} + 2c\Gamma(t) \right) G(t|r) = 8\pi c^2 \int_{t_0}^t ds G(s|r) G(t|s) \overline{\gamma}(r|s) \]

(6.5)

where (4.4) has been employed.

The set of equations (4.1)(6.3)(6.4) and (6.5) are closed and entirely independent of the velocity field specification. They determine the behaviour of \( \Gamma(t) \), and \( \overline{\gamma}(t) \) in terms of their initial descriptions. On multiplying (2.4) by \( \gamma \), taking an ensemble average and comparing the result with (6.3) in the limit \( D = 0 \), it is clear that in the L.H.D.I. approximation

\[
\overline{\gamma}^2(t) = 6\pi \int_{t_0}^t ds \overline{\gamma}^2(t|s) G(t|s)
\]

(6.6)

and all terms in the integrand are determined independently of the velocity field. Finally we remark that the above arguments apply equally well to the abridged form of the equations derived in section 4 so that they too preserve this particular invariance.
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