

QUARTERLY OF APPLIED MATHEMATICS

Vol. XXXI

JULY 1973

No. 2

ANALYTICAL STRUCTURE OF A GENERALIZED DIRECT-INTERACTION APPROXIMATION*

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Abstract. As a mathematically tractable example, we have investigated the stochastic dynamic problem of an irreversible second-order chemical reaction. A generalized direct-interaction approximation has been devised to close off the hierarchy of moment equations at the arbitrary moment level, and then the results of such a closure technique have been compared term-by-term with the exact moment solutions. This shows qualitatively how the expansion terms summed up in the direct-interaction approximation are different from the classes of expansion terms present in the exact moment solutions. A quantitative comparison of the covariances indicates that the direct-interaction equations which are closed at the triple moment level represent a meaningful statistical approximation of the lowest order for the second-order reactive problem at hand.

The problem statement. As a mathematically tractable example, we shall consider in this paper the stochastic dynamical problem governed by

$$d\psi(t)/dt = -K\psi(t)^2, \quad (1.1)$$

where K is assumed constant. Physically, (1.1) describes the depletion of the reactant species $\psi(t)$ by an isothermal, irreversible second-order chemical reaction with the reaction-rate constant K [1, 2]. Considering an ensemble of realizations each governed by (1.1), a stochastic dynamical problem can be formulated by assigning certain statistical properties to the initial ensemble. For the deterministic solution of (1.1)

$$\psi(t) = \frac{\psi(t_0)}{1 + K\psi(t_0)(t - t_0)}, \quad (1.2)$$

where t_0 is the initial time, the stochastic dynamics may be described by the statistical distribution of $\psi(t_0)$. In particular, the moments of one time argument are completely determined by the initial distribution $P[\psi(t_0)]$. For $t_0 = 0$, the mean value is

$$\Psi(t) = \langle \psi(t) \rangle = \int \left(\frac{x}{1 + Ktx} \right) dP(x). \quad (1.3)$$

* Received October 4, 1971. Dr. Kraichnan's suggestions on the first draft of the manuscript have been extremely helpful.

And the covariance and triple moment about the $\Psi(t)$ are

$$U(t) = \langle (\psi(t) - \Psi(t))^2 \rangle = \langle \psi(t)^2 \rangle - \Psi(t)^2, \quad (1.4)$$

$$V(t) = \langle (\psi(t) - \Psi(t))^3 \rangle = \langle \psi(t)^3 \rangle - 3\langle \psi(t)^2 \rangle \Psi(t) + 2\Psi(t)^3, \quad (1.5)$$

where $\langle \psi(t)^m \rangle = \int (x/(1 + Ktx))^m dP(x)$. Furthermore, similar expressions can be written down for the quadruple moment $W(t)$, the quintuple moment $X(t)$, the hextuple moment $Y(t)$, etc.

For the turbulent flow problem, however, it is not practical to evolve an ensemble of flow systems each of which obeys the Navier–Stokes equations. Therefore the most practical alternative is to recast the Navier–Stokes equations into a set of moment equations, thereby evolving as many moments as are necessary for conveying the desired statistical information. This is the moment formulation. Due to the nonlinearity of the Navier–Stokes equations, the moment formulation will give rise to the hierarchy of unclosed moment equations. Although many suggestions have been made in the past for the closure problem, it appears that closing of the moment equations is still an art rather than a science [3]. In this paper, using the simple system (1.1), we shall demonstrate how the hierarchy of moment equations can be closed at any moment level by a generalized direct-interaction (DI) approximation, and then compare the results of such a closure scheme with the exact moment solutions qualitatively as well as quantitatively. First of all, the first three sets of closed statistical equations have been constructed respectively for the covariance, triple moment, and quadruple moment levels of closure (Secs. 2–6). The closed statistical equations of the covariance level are the same as the Kraichnan’s DI equations [1, 4], and the DI system closed at a certain moment level contains all the DI systems of lower-order moment level (Sec. 7). For this reason we are justified in calling the present closure scheme a generalized DI approximation, whose consistency is demonstrated by the shell structure of the DI systems. Secondly, the exact moments (1.3)–(1.5) have been transformed into the same form as the DI approximation results (Sec. 8). The term-by-term comparison shows qualitatively how the expansion terms included in the DI approximation are different from those terms present in the exact moment solutions. For the second-order reactive problem, the DI system of the triple moment level represents a meaningful statistical approximation of the lowest order in that it can describe the decay of the reactant species fluctuations whose distribution is not only asymmetric but also defined only for the positive random variable. On the other hand, the usual DI equations of the covariance level collect only the terms whose coefficients are of the even powers in $(-K)$, and hence the reactant fluctuations must be restricted to have a symmetric distribution at this stage of moment formulation. The covariance level formulation is inadequate because the distinction is completely lost between the depletive and generative type of chemical reactions. This has been substantiated by a quantitative comparison of the covariances evolved from an initial Helmholtz distribution [5] (Sec. 9).

2. The collective representation. To obtain the moment equations about the mean value, we split $\psi(t)$ into the mean $\Psi(t)$ and the fluctuation $\psi'(t)$. The mean equation is

$$(d/dt + K\Psi(t))\Psi(t) = -KU(t), \quad (2.1)$$

and the fluctuation equation becomes

$$(d/dt + 2K\Psi(t))\psi'(t) = K(U(t) - \psi'(t)^2). \tag{2.2}$$

Associated with (2.2) is the linearized response equation

$$(d/dt + 2K\Psi(t))G(t, t') = -2K\psi'(t)G(t, t') + \delta(t - t'), \tag{2.3}$$

with respect to an infinitesimal disturbance. Although we wish to work with the more familiar discrete Fourier modes, the absence of spatial coordinates in (2.1)–(2.3) prevents us from seeking a Fourier representation. Therefore, we shall resort to Kraichnan's artifice [4] of the collective representation by introducing an ensemble of M identically and independently distributed systems. The $\langle \rangle$ now represents an average over such M realizations; we shall be concerned with the limit as $M \rightarrow \infty$. Although the collective representation for the present problem can be obtained directly from (2.2), we shall follow the formal procedure of Kraichnan [4] in the interest of general readers.

Upon denoting the $\psi(t)$ of an individual realization by $\psi_{[n]}(t)$, we write (1.1) for the n th system as

$$d\psi_{[n]}(t)/dt = -K\psi_{[n]}(t)^2. \tag{2.4}$$

Further, denoting by $G_{[n,m]}(t, t')$ the response of the n th system with respect to an infinitesimal perturbation of the m th system, the linearized response equation for the n th system is

$$dG_{[n,m]}(t, t')/dt = -2K\psi_{[n]}(t)G_{[n,m]}(t, t') + \delta(t - t'). \tag{2.5}$$

For the collective representation, we introduce $(\alpha, \gamma = 0, \pm 1, \dots, \pm S, S = \text{integers}, M = 2S + 1)$

$$\psi_\alpha(t) = M^{-1/2} \sum_n \exp(i2\pi\alpha n/M) \psi_{[n]}(t), \tag{2.6}$$

$$G_{\alpha,\gamma}(t, t') = M^{-1} \sum_n \exp(i2\pi(\alpha - \gamma)n/M) G_{[n,m]}(t, t'). \tag{2.7}$$

The reality requirement states $\psi_\alpha(t) = \psi_{-\alpha}^*(t)$. Using the identities $\delta_{n,m} = M^{-1} \sum_\alpha \exp(i2\pi\alpha(n - m)/M)$ and $\delta_{\alpha,\beta} = M^{-1} \sum_n \exp(i2\pi n(\alpha - \beta)/M)$, the set of (2.4) and (2.5) gives

$$d\psi_\alpha(t)/dt = -KM^{-1/2} \sum_\beta \psi_\beta(t) \psi_{\alpha-\beta}(t), \tag{2.8}$$

$$dG_{\alpha,\gamma}(t, t')/dt = -2KM^{-1/2} \sum_\beta \psi_\beta(t) G_{\alpha-\beta,\gamma}(t, t') + \delta_{\alpha,\gamma} \delta(t - t'), \tag{2.9}$$

where $\alpha - \beta$ is interpreted using the cyclic convention $\alpha = \alpha \pm M$. Using the statistical sharpness $\langle |M^{-1/2}\psi_\alpha(t) - \Psi(t)|^2 \rangle = 0(M^{-1})$, (2.8) gives the mean equation for $\alpha = 0$

$$(d/dt + K\Psi(t))\Psi(t) = -KM^{-1} \sum_\beta' \langle \psi_\beta(t) \psi_\beta^*(t) \rangle, \tag{2.10}$$

where \sum_β' omits $\beta = 0$. For α and $\delta \neq 0$, (2.8) and (2.9) give

$$(d/dt + 2K\Psi(t))\psi_\alpha(t) = -KM^{-1/2} \sum_\beta'' \psi_\beta(t) \psi_{\alpha-\beta}(t), \tag{2.11}$$

$$(d/dt + 2K\Psi(t))G_{\alpha,\gamma}(t, t') = -2KM^{-1/2} \sum_\beta'' \psi_\beta(t) G_{\alpha-\beta,\gamma}(t, t') + \delta_{\alpha,\gamma} \delta(t - t'), \tag{2.12}$$

where \sum''_{β} omits both $\beta = 0$ and $\beta = \alpha$. Eqs. (2.11) and (2.12) are the collective representations of (2.2) and (2.3), respectively; they may be compared structurally with Kraichnan's Eqs. (11.12) and (11.13) of [4].

A minimal set of the statistical properties necessary for the present work will be summarized. First, the averages of the ψ 's obey

$$\langle \psi_{\alpha}(t) \psi_{\beta}(t') \cdots \psi_{\mu}(t'') \rangle = 0, \quad (\alpha + \beta + \cdots + \mu \neq 0) \quad (2.13)$$

and

$$\langle \psi_{\alpha}(t) \cdots \psi_{\beta}(t') \psi_{\mu}(s) \cdots \psi_{\gamma}(s') \rangle \rightarrow \langle \psi_{\alpha}(t) \cdots \psi_{\beta}(t') \rangle \langle \psi_{\mu}(s) \cdots \psi_{\gamma}(s') \rangle + 0(M^{-1}), \quad (2.14)$$

where $\alpha + \cdots + \beta = 0$ and $\mu + \cdots + \gamma = 0$. The Fourier modes in the homogeneous velocity field obey the statistical properties quite similar to the above, as shown by Orszag [6]. Secondly, the moments containing a factor of $G_{\alpha,\gamma}$ obey

$$\langle G_{\alpha-\delta,\alpha}(t, t') \psi_{\beta}(t'') \cdots \psi_{\mu}(t''') \rangle = 0, \quad (\delta \neq \beta + \cdots + \mu) \quad (2.15)$$

and

$$\begin{aligned} & \langle G_{\alpha-\delta,\alpha}(t, t') \psi_{\beta}(t'') \cdots \psi_{\rho}(t''') \psi_{\rho}(s) \cdots \psi_{\gamma}(s') \rangle \\ & \rightarrow \langle G_{\alpha-\delta,\alpha}(t, t') \psi_{\beta}(t'') \cdots \psi_{\mu}(t''') \rangle \langle \psi_{\rho}(s) \cdots \psi_{\gamma}(s') \rangle + 0(M^{-1}), \end{aligned} \quad (2.16)$$

where $\delta = \beta + \cdots + \mu$ and $\rho + \cdots + \gamma = 0$. Kraichnan [4] has shown (2.13) and (2.14) from the fact that M realizations have identical and independent distributions, and a similar argument can be used to justify (2.15) and (2.16). Lastly, using the statistical sharpness of $G_{\alpha,\alpha}$, the diagonal $G_{\alpha,\alpha}$ can be separated from the average of the ψ factors

$$\langle G_{\alpha,\alpha}(t, t') \psi_{\beta}(s) \cdots \psi_{\mu}(s') \rangle \rightarrow \langle G_{\alpha,\alpha}(t, t') \rangle \langle \psi_{\beta}(s) \cdots \psi_{\mu}(s') \rangle, \quad (2.17)$$

in the limit as $M \rightarrow \infty$. Orszag [6] has criticized (2.17) on the ground that it destroys the Galilean invariance of the DI equations for the turbulent flow problem.

3. The class A moments. Instead of (2.11) and (2.12), let us consider the following equations:

$$(d/dt + 2K\Psi(t))\psi_{\alpha}(t) = -KM^{-1/2} \sum''_{\beta} \phi_{\alpha,\beta,\alpha-\beta} \psi_{\beta}(t) \psi_{\alpha-\beta}(t), \quad (3.1)$$

$$\begin{aligned} & (d/dt + 2K\Psi(t))G_{\alpha,\gamma}(t, t') \\ & = -2KM^{-1/2} \sum''_{\beta} \phi_{\alpha,\beta,\alpha-\beta} \psi_{\beta}(t) G_{\alpha-\beta,\gamma}(t, t') + \delta_{\alpha,\gamma} \delta(t - t'). \end{aligned} \quad (3.2)$$

Unlike the random coupling model of Kraichnan [4], the $\phi_{\alpha,\beta,\alpha-\beta}$ will have no dynamic significance since all $\phi = 1$. We have introduced the ϕ into (3.1) and (3.2) simply to construct diagrams by the following rules. (i) Associate the $\phi_{\alpha,\beta,\alpha-\beta}$ with an open circle to which three broken lines are attached. (ii) Label each broken line with one of three indices of the ϕ . (iii) Indicate the sense of indices by attaching an incoming arrow head with α and an outgoing arrow head with both β and $\alpha - \beta$, so that the incoming indices add up to the outgoing indices. (iv) Reverse the arrow head of, say, α to represent $-\alpha$. (v) Replace the broken line with α by a wavy line to represent ψ_{α} . And (vi) replace both the incoming broken line with α and the outgoing broken line with $\alpha - \beta$ by solid lines to represent $G_{\alpha-\beta,\alpha}$.

First of all, the covariance $\langle \psi_{\alpha} \psi_{\alpha}^* \rangle$ will be treated as diagrammatically equivalent to $\langle \phi_{\alpha,\beta,\alpha-\beta} \phi_{\beta,\alpha,\alpha-\beta} \psi_{\alpha} \psi_{\alpha}^* \rangle$, as shown in Fig. 1. We shall then call the following higher-order

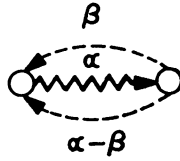


FIG. 1. Covariance.

moments the class *A* moments:

- I. $\langle \phi_{\alpha, \beta, \alpha-\beta} \psi_{\beta} \psi_{\alpha-\beta}^* \rangle,$
- II. $\langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\beta, \sigma, \beta-\sigma} \psi_{\sigma} \psi_{\beta-\sigma} \psi_{\alpha-\beta}^* \rangle,$
- III. $\langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\beta, \sigma, \beta-\sigma} \phi_{\sigma, \mu, \sigma-\mu} \psi_{\sigma-\mu} \psi_{\beta-\sigma} \psi_{\alpha-\beta}^* \rangle,$
- IV. $\langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\beta, \sigma, \beta-\sigma} \phi_{\sigma, \mu, \sigma-\mu} \phi_{\mu, \rho, \mu-\rho} \psi_{\rho} \psi_{\mu-\rho} \psi_{\sigma-\mu} \psi_{\beta-\sigma} \psi_{\alpha-\beta}^* \rangle,$
- V. $\langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\beta, \sigma, \beta-\sigma} \phi_{\sigma, \mu, \sigma-\mu} \phi_{\mu, \rho, \mu-\rho} \psi_{\rho} \psi_{\beta-\sigma-\rho} \psi_{\sigma-\mu} \psi_{\alpha-\beta}^* \rangle,$

The above moments are represented diagrammatically in Fig. 2. We have replaced the

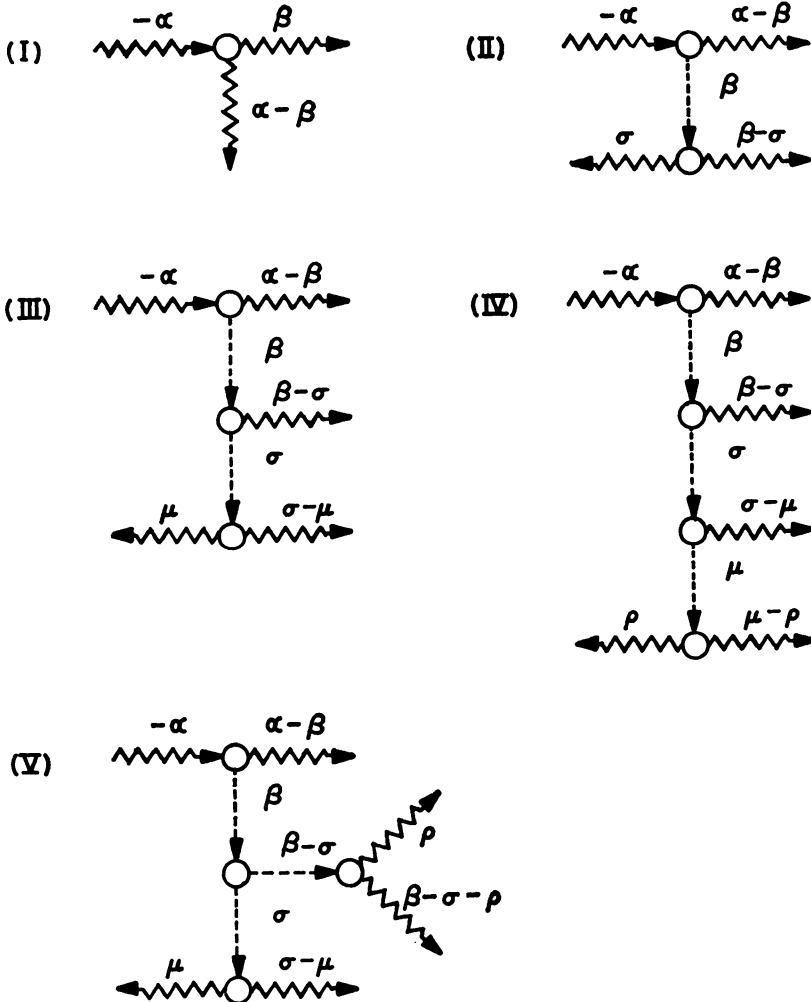


FIG. 2. Class *A* moments.

first α of the leading ϕ by $-\alpha$, so as to assure zero sum of the indices as required by (2.13). The class *A* moments all have a unique feature of containing the least number of open circles to connect the wavy lines in each diagram. Hence, we include in class *B* all other moments that would involve more than the minimum number of open circles as required by (I)–(V). For instance, a typical triple moment of class *B* that we shall encounter later is of the form

$$\langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\alpha - \beta, \sigma, \alpha - \beta - \sigma} \phi_{\alpha - \beta - \sigma, -\beta - \sigma, \alpha} \psi_{\beta} \psi_{\sigma} \psi_{\beta + \sigma}^* \rangle, \quad (3.3)$$

whose diagram is given in Fig. 3.

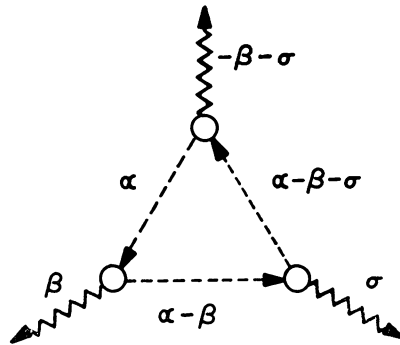


FIG. 3. A triple moment of class *B*.

Next, the averaged response function is $\langle G_{\alpha, \alpha} \rangle$. For the higher-order associated moments containing a factor of G , we include in class *A* the following ones with the diagrams shown in Fig. 4:

Again, the class *A* associated moments involve the smallest number of open circles to connect the two solid lines and several wavy lines in each diagram; all other moments requiring more than the minimum number of open circles will be included in class *B*. The particular configurations of the diagrams are, of course, immaterial because the diagrams are a topological representation.

4. The hierarchy of moment equations. The objective of this section is to build up the hierarchy of moment equations containing only the class *A* moments. To do this requires defining precisely certain moments which will appear in the statistical equations to be derived. Comparing the right-hand sides of (2.1) and (2.10), we arrive at the covariance definition

$$U(t, t') = M^{-1} \sum_{\alpha} \langle \psi_{\alpha}(t) \psi_{\alpha}^*(t') \rangle.$$

And analogously we define the averaged response function as

$$G(t, t') = M^{-1} \sum_{\alpha} \langle G_{\alpha, \alpha}(t, t') \rangle.$$

The above definitions, however, do not require $\langle \psi_{\alpha} \psi_{\alpha}^* \rangle$ and $\langle G_{\alpha, \alpha} \rangle$ to be independent of α , as in Kraichnan's formulation [4]. The higher-order moments are systematically

- VI. $\langle \phi_{\alpha, \beta, \alpha-\beta} \psi_{\beta} G_{\alpha-\beta, \alpha} \rangle,$
- VII. $\langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\alpha-\beta, \sigma, \alpha-\beta-\sigma} \psi_{\beta} \psi_{\sigma} G_{\alpha-\beta-\sigma, \alpha} \rangle,$
- VIII. $\langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\alpha-\beta, \sigma, \alpha-\beta-\sigma} \phi_{\alpha-\beta-\sigma, \mu, \alpha-\beta-\sigma-\mu} \psi_{\beta} \psi_{\sigma} \psi_{\mu} G_{\alpha-\beta-\sigma-\mu, \alpha} \rangle,$
- IX. $\langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\alpha-\beta, \sigma, \alpha-\beta-\sigma} \phi_{\alpha-\beta-\sigma, \mu, \alpha-\beta-\sigma-\mu} \phi_{\alpha-\beta-\sigma-\mu, \rho, \alpha-\beta-\sigma-\mu-\rho} \psi_{\beta} \psi_{\sigma} \psi_{\mu} \psi_{\rho} G_{\alpha-\beta-\sigma-\mu-\rho, \alpha} \rangle.$

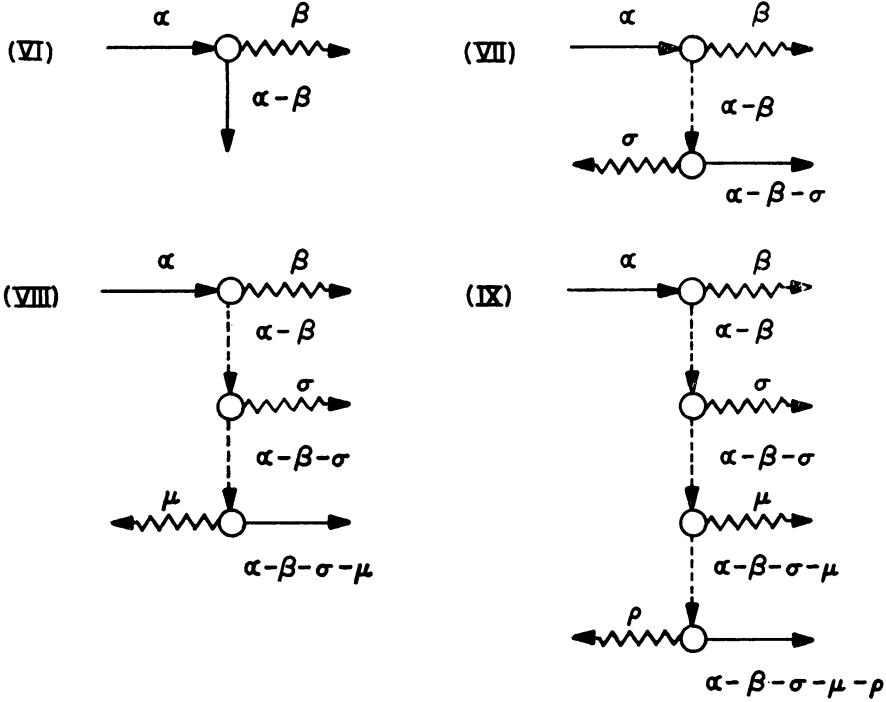


FIG. 4. Class A associated moments.

defined as

$$V(t, t', t'') = M^{-3/2} \sum_{\alpha} \sum_{\beta} \langle \phi_{\alpha, \beta, \alpha-\beta} \psi_{\beta}(t) \psi_{\alpha-\beta}(t') \psi_{\alpha}^*(t'') \rangle,$$

$$W(t, t', t'', t''') = M^{-2} \sum_{\alpha} \sum_{\beta} \sum_{\substack{\sigma \neq \alpha \\ \neq \beta-\alpha}} \langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\alpha-\beta, \sigma, \beta-\sigma} \psi_{\sigma}(t) \psi_{\beta-\sigma}(t') \psi_{\alpha-\beta}(t'') \psi_{\alpha}^*(t''') \rangle,$$

$$X(t, t', t'', t''', t^{iv}) = M^{-5/2} \sum_{\alpha} \sum_{\beta} \sum_{\substack{\sigma \neq \alpha \\ \neq \beta-\alpha}} \sum_{\substack{\mu \neq \beta \\ \neq \sigma-\beta}} \langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\beta, \sigma, \beta-\sigma} \phi_{\sigma, \mu, \sigma-\mu} \psi_{\mu}(t) \cdot \psi_{\sigma-\mu}(t') \psi_{\beta-\sigma}(t'') \psi_{\alpha-\beta}(t''') \psi_{\alpha}^*(t^{iv}) \rangle,$$

$$Y(t, t', t'', t''', t^{iv}, t^v) = M^{-3} \sum_{\alpha} \sum_{\beta} \sum_{\substack{\sigma \neq \alpha \\ \neq \beta-\alpha}} \sum_{\substack{\mu \neq \beta \\ \neq \sigma-\beta}} \sum_{\substack{\rho \neq \sigma \\ \neq \mu-\sigma}} \langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\beta, \sigma, \beta-\sigma} \phi_{\sigma, \mu, \sigma-\mu} \cdot \phi_{\mu, \rho, \mu-\rho} \psi_{\rho}(t) \psi_{\mu-\rho}(t') \psi_{\sigma-\mu}(t'') \psi_{\beta-\sigma}(t''') \psi_{\alpha-\beta}(t^{iv}) \psi_{\alpha}^*(t^v) \rangle,$$

$$\tilde{Y}(t, t', t'', t''', t^{iv}, t^v) = M^{-3} \sum_{\alpha} \sum_{\beta} \sum_{\substack{\sigma \neq \alpha \\ \neq \beta-\alpha}} \sum_{\substack{\mu \neq \beta \\ \neq \sigma-\beta}} \sum_{\substack{\rho \neq \sigma \\ \neq \mu-\sigma}} \langle \phi_{\alpha, \beta, \alpha-\beta} \phi_{\beta, \sigma, \beta-\sigma} \phi_{\sigma, \mu, \sigma-\mu} \cdot \phi_{\beta-\sigma, \rho, \beta-\sigma-\rho} \psi_{\rho}(t) \psi_{\sigma-\mu}(t') \psi_{\rho}(t'') \psi_{\beta-\sigma-\rho}(t''') \psi_{\alpha-\beta}(t^{iv}) \psi_{\alpha}^*(t^v) \rangle,$$

$$F(t, t', t'') = M^{-3/2} \sum_{\alpha} \sum_{\beta} \langle \phi_{\alpha, \beta, \alpha-\beta} G_{\alpha-\beta, \alpha}(t, t') \psi_{\beta}(t'') \rangle,$$

$$\begin{aligned}
E(t, t', t'', t''') &= M^{-2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma \neq \beta}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\alpha - \beta, \sigma, \alpha - \beta - \sigma} G_{\alpha - \beta - \sigma, \alpha}(t, t') \psi_{\sigma}(t'') \psi_{\beta}(t''') \rangle, \\
D(t, t', t'', t''', t^{iv}) &= M^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma \neq \beta}'' \sum_{\mu \neq \sigma}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\alpha - \beta, \sigma, \alpha - \beta - \sigma} \phi_{\alpha - \beta - \sigma, \mu, \alpha - \beta - \sigma - \mu} \\
&\quad \cdot G_{\alpha - \beta - \sigma - \mu, \alpha}(t, t') \psi_{\mu}(t'') \psi_{\sigma}(t''') \psi_{\beta}(t^{iv}) \rangle, \\
C(t, t', t'', t''', t^{iv}, t^v) \\
&= M^{-3} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma \neq \beta}'' \sum_{\mu \neq \sigma}'' \sum_{\rho \neq \mu}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\alpha - \beta, \sigma, \alpha - \beta - \sigma} \phi_{\alpha - \beta - \sigma, \mu, \alpha - \beta - \sigma - \mu} \\
&\quad \cdot \phi_{\alpha - \beta - \sigma - \mu, \rho, \alpha - \beta - \sigma - \mu - \rho} G_{\alpha - \beta - \sigma - \mu - \rho, \alpha}(t, t') \psi_{\rho}(t'') \psi_{\mu}(t''') \psi_{\sigma}(t^{iv}) \psi_{\beta}(t^v) \rangle.
\end{aligned}$$

Note that only the class A moments (I)–(IX) are involved in the above.

4.1 Covariance equations. For the covariance and averaged response equations, we apply $M^{-1} \sum_{\alpha}' \psi_{\alpha}^*(t')$ and $M^{-1} \sum_{\alpha}'$ respectively to (3.1) and (3.2) and obtain the following equations after averaging:

$$(d/dt + 2K\Psi(t))U(t, t') = -KV(t, t, t'), \quad (4.1)$$

$$(d/dt + 2K\Psi(t))G(t, t') = -2KF(t, t', t) + \delta(t - t'), \quad (4.2)$$

with $G(t, t') = 0$ if $t < t'$. The covariance is symmetric in t and t' ; hence the redundant equation for U in t' can be written down immediately from (4.1).

4.2 Triple moment equations. Apply $M^{-3/2} \sum_{\alpha}' \sum_{\beta'}' \phi_{\alpha, \beta, \alpha - \beta} \psi_{\alpha - \beta}(t') \psi_{\alpha}^*(t'')$ to the equation for $\psi_{\beta}(t)$. Upon averaging, we have

$$\begin{aligned}
&(d/dt + 2K\Psi(t))V(t, t', t'') \\
&= -KM^{-2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\beta, \sigma, \beta - \sigma} \psi_{\sigma}(t) \psi_{\beta - \sigma}(t) \psi_{\alpha - \beta}(t') \psi_{\alpha}^*(t'') \rangle. \quad (4.3)
\end{aligned}$$

The right-hand-side moment has the diagram (II) of Fig. 2 which reduces to a closed loop similar to Fig. 1 when $\sigma = \alpha$ and $\beta = \alpha$. This means that analytically the right-hand side becomes

$$\begin{aligned}
&-2KM^{-2} \sum_{\alpha}' \sum_{\beta}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\beta, \alpha, \beta - \alpha} \psi_{\alpha}(t) \psi_{\beta - \alpha}(t) \psi_{\alpha - \beta}(t') \psi_{\alpha}^*(t'') \rangle \\
&-KM^{-2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \mu \neq \beta - \alpha}}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\beta, \sigma, \beta - \sigma} \psi_{\sigma}(t) \psi_{\beta - \sigma}(t) \psi_{\alpha - \beta}(t') \psi_{\alpha}^*(t'') \rangle.
\end{aligned}$$

Therefore, in view of (2.14), the V equation becomes

$$(d/dt + 2K\Psi(t))V(t, t', t'') = -2KU(t, t')U(t, t'') - KW(t, t, t', t''). \quad (4.4)$$

Since V is symmetric in all time arguments, the redundant equations for V in t' and t'' can be obtained by interchanging the time arguments in (4.4).

For the derivation of the F equation, we apply $M^{-3/2} \sum_{\alpha}' \sum_{\beta'}' \phi_{\alpha, \beta, \alpha - \beta} \psi_{\beta}(t'')$ to the equation for $G_{\alpha - \beta, \alpha}(t, t')$ and obtain after averaging

$$\begin{aligned}
&(d/dt + 2K\Psi(t))F(t, t', t'') \\
&= -2KM^{-2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\alpha - \beta, \sigma, \alpha - \beta - \sigma} G_{\alpha - \beta - \sigma, \alpha}(t, t') \psi_{\sigma}(t) \psi_{\beta}(t'') \rangle. \quad (4.5)
\end{aligned}$$

Since $\sigma = -\beta$ gives rise to a term which involves the diagonal $G_{\alpha,\alpha}$, we write the right-hand side as

$$\begin{aligned}
 & -2KM^{-2} \sum_{\alpha}' \sum_{\beta}'' \langle \phi_{\alpha,\beta,\alpha-\beta} \phi_{\alpha-\beta,-\beta,\alpha} G_{\alpha,\alpha}(t, t') \psi_{\beta}^*(t) \psi_{\beta}(t'') \rangle \\
 & -2KM^{-2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma \neq -\beta}'' \langle \phi_{\alpha,\beta,\alpha-\beta} \phi_{\alpha-\beta,\sigma,\alpha-\beta-\sigma} G_{\alpha-\beta-\sigma,\alpha}(t, t') \psi_{\sigma}(t) \psi_{\beta}(t'') \rangle.
 \end{aligned}$$

Then, invoking (2.17), the F equation becomes

$$(d/dt + 2K\Psi(t))F(t, t', t'') = -2KU(t, t'')G(t, t') - 2KE(t, t', t, t''), \quad (4.6)$$

with $F(t', t', t'') = 0$ and $F = 0$ if $t < t'$.

4.3 Quadruple moment equations. It is by now evident how the quadruple moment equations can be constructed. First, to obtain the W equation, we apply

$$M^{-2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \neq \beta - \alpha}}'' \phi_{\alpha,\beta,\alpha-\beta} \phi_{\beta,\sigma,\beta-\sigma} \psi_{\beta-\sigma}(t') \psi_{\alpha-\beta}(t'') \psi_{\alpha}^*(t''')$$

to the equation for $\psi_{\sigma}(t)$:

$$\begin{aligned}
 (d/dt + 2K\Psi(t))W(t, t', t'', t''') = & -KM^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \neq \beta - \alpha}}'' \sum_{\mu}'' \langle \phi_{\alpha,\beta,\alpha-\beta} \phi_{\beta,\sigma,\beta-\sigma} \\
 & \cdot \phi_{\sigma,\mu,\sigma-\mu} \psi_{\mu}(t) \psi_{\sigma-\mu}(t) \psi_{\beta-\sigma}(t') \psi_{\alpha-\beta}(t'') \psi_{\alpha}^*(t''') \rangle. \quad (4.7)
 \end{aligned}$$

We see from the diagram (III) in Fig. 2 that there are three possibilities for splitting the right-hand side into lower-order moments. For the first case of $\mu = \beta$ and $\sigma - \beta$, the right-hand side gives rise to

$$\begin{aligned}
 -2KM^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \neq \beta - \alpha}}'' \langle \phi_{\alpha,\beta,\alpha-\beta} \phi_{\beta,\sigma,\beta-\sigma} \phi_{\sigma,\beta,\sigma-\beta} \\
 \cdot \psi_{\beta}(t) \psi_{\sigma-\beta}(t) \psi_{\beta-\sigma}(t') \psi_{\alpha-\beta}(t'') \psi_{\alpha}^*(t''') \rangle, \quad (4.8)
 \end{aligned}$$

which, by virtue of (2.14), represents the decomposition into the lower-order moments of class A . On the other hand, the second ($\mu = \alpha$ and $\beta - \alpha$) and third ($\mu = \sigma - \alpha$ and $\sigma - \alpha + \beta$) cases result in decompositions into the lower-order class B moments. For instance, in the second case, the right-hand side of (4.7) gives rise to

$$\begin{aligned}
 -2KM^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \neq \beta - \alpha}}'' \langle \phi_{\alpha,\beta,\alpha-\beta} \phi_{\beta,\sigma,\beta-\sigma} \phi_{\sigma,\alpha,\sigma-\alpha} \psi_{\sigma-\alpha}(t) \psi_{\beta-\sigma}(t') \psi_{\alpha-\beta}(t'') \rangle \\
 \cdot \langle \psi_{\alpha}(t) \psi_{\alpha}^*(t''') \rangle. \quad (4.9)
 \end{aligned}$$

Note, however, that the first $\langle \rangle$ factor is a triple moment of class B which has the diagrammatic representation of Fig. 3. Thus, considering only the decomposition (4.8) into the class A moments, the W equation becomes

$$(d/dt + 2K\Psi(t))W(t, t', t'', t''') = -2KU(t, t'')V(t, t'', t''') - KX(t, t, t', t'', t'''). \quad (4.10)$$

Again, the three redundant equations for W in t' , t'' and t''' can be written down immediately from (4.10) because W is symmetric in all time arguments.

Next, for the derivation of the E equation, we apply $M^{-2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma \neq -\beta}'' \phi_{\alpha,\beta,\alpha-\beta} \phi_{\alpha-\beta,\sigma,\alpha-\beta-\sigma} \psi_{\sigma}(t'') \psi_{\beta}(t''')$ to the equation for $G_{\alpha-\beta-\sigma,\alpha}(t, t')$:

$$\begin{aligned}
 (d/dt + 2K\Psi(t))E(t, t', t'', t''') = & -2KM^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma \neq -\beta}'' \sum_{\mu}'' \langle \phi_{\alpha,\beta,\alpha-\beta} \phi_{\alpha-\beta,\sigma,\alpha-\beta-\sigma} \\
 & \cdot \phi_{\alpha-\beta-\sigma,\mu,\alpha-\beta-\sigma-\mu} G_{\alpha-\beta-\sigma-\mu,\alpha}(t, t') \psi_{\mu}(t) \psi_{\sigma}(t'') \psi_{\beta}(t''') \rangle. \quad (4.11)
 \end{aligned}$$

Under $\mu = -\sigma$, the right-hand side gives rise to

$$-2KM^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \sigma \neq -\beta}}'' \langle \phi_{\alpha, \beta, \alpha - \beta} \phi_{\alpha - \beta, \sigma, \alpha - \beta - \sigma} \phi_{\alpha - \beta - \sigma, -\sigma, \alpha - \beta} \\ \cdot G_{\alpha - \beta, \alpha}(t, t') \psi_{\sigma}^*(t) \psi_{\sigma}(t'') \psi_{\beta}(t''') \rangle,$$

which can be decomposed into the lower-order class A moments. Hence, considering this decomposition, the E equation becomes

$$(d/dt + 2K\Psi(t))E(t, t', t'', t''') = -2KU(t, t'')F(t, t', t''') - 2KD(t, t', t, t'', t'''), \quad (4.12)$$

with $E(t', t', t'', t''') = 0$ and $E = 0$ if $t < t'$. For $\mu = -\beta - \sigma$, the right-hand side of (4.11) can further be split into lower-order moments; however, it gives rise to a triple moment of class B .

4.4 Quintuple moment equations. For the moment equations for U , V , and W , it was possible to derive the redundant equations in t' , t'' , and t''' by simply interchanging the time arguments. This came about from the complete symmetry of U , V , and W diagrams with respect to the time arguments in that arbitrary relabeling of the wavy lines did not alter the diagrammatic structure. On the other hand, the diagram (III) in Fig. 2 is symmetric only with respect to the four wavy lines with $-\alpha$, $\alpha - \beta$, μ , and $\sigma - \mu$, but not with respect to the wavy line with $\beta - \sigma$. Therefore, we must have two distinct moment equations for X ; one to represent the four moment equations in t , t' , t'' , t^{iv} , and the other in t'' . To obtain the first X equation, we apply

$$M^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \sigma \neq -\beta}}'' \sum_{\substack{\mu \neq \beta \\ \mu \neq -\sigma}}'' \phi_{\alpha, \beta, \alpha - \beta} \phi_{\beta, \sigma, \beta - \sigma} \phi_{\sigma, \mu, \sigma - \mu} \psi_{\sigma - \mu}(t') \psi_{\beta - \sigma}(t'') \psi_{\alpha - \beta}(t''') \psi_{\alpha}^*(t^{iv})$$

to the equation for $\psi_{\mu}(t)$. Of the several decompositions, only the case of $\rho = \sigma$ and $\mu - \sigma$ will result in decomposition into the lower-order class A moments; hence, we have

$$(d/dt + 2K\Psi(t))X(t, t', t'', t''', t^{iv}) = -2KU(t, t')W(t, t'', t''', t^{iv}) \\ - KY(t, t, t, t', t'', t''', t^{iv}). \quad (4.13)$$

This represents the redundant equations in t' , t'' , t^{iv} because X is symmetric in those time arguments. For the second X equation in t'' , we apply

$$M^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\substack{\sigma \neq \alpha \\ \sigma \neq -\beta}}'' \sum_{\substack{\mu \neq \beta \\ \mu \neq -\sigma}}'' \phi_{\alpha, \beta, \alpha - \beta} \phi_{\beta, \sigma, \beta - \sigma} \phi_{\sigma, \mu, \sigma - \mu} \psi_{\mu}(t) \psi_{\sigma - \mu}(t') \psi_{\alpha - \beta}(t''') \psi_{\alpha}^*(t^{iv})$$

to the equation for $\psi_{\beta - \sigma}(t')$. Considering only the case of $\rho = -\sigma$ and β , which gives rise to the lower-order class A moments, we have

$$(d/dt'' + 2K\Psi(t''))X(t, t', t'', t''', t^{iv}) = -2KV(t, t', t'')V(t'', t''', t^{iv}) \\ - K\tilde{Y}(t, t', t'', t'', t''', t^{iv}). \quad (4.14)$$

Finally, for the derivation of the D equation, we apply

$$M^{-5/2} \sum_{\alpha}' \sum_{\beta}'' \sum_{\sigma \neq -\beta}'' \sum_{\mu \neq -\sigma}'' \phi_{\alpha, \beta, \alpha - \beta} \phi_{\alpha - \beta, \sigma, \alpha - \beta - \sigma} \phi_{\alpha - \beta - \sigma, \mu, \alpha - \beta - \sigma - \mu} \psi_{\mu}(t') \psi_{\sigma}(t'') \psi_{\beta}(t''')$$

to the equation for $G_{\alpha - \beta - \sigma - \mu, \alpha}(t, t')$. Considering only the decomposition under $\rho = -\mu$,

we have

$$(d/dt + 2K\Psi(t))D(t, t', t'', t''', t^{iv}) = -2KU(t, t'')E(t, t', t''', t^{iv}) - 2KC(t, t', t, t'', t''', t^{iv}), \quad (4.15)$$

with $D(t', t', \dots) = 0$ and $D = 0$ if $t < t'$.

5. The closure procedure. Starting from the mean equation (2.1), us let summarize the unclosed moment equations derived in the previous section:

$$(d/dt + K\Psi(t))\Psi(t) = -KU(t), \quad (5.1)$$

$$(d/dt + 2K\Psi(t))U(t, t') = -KV(t, t, t'),$$

Redundant equation for U in t' ; (5.2)

$$(d/dt + 2K\Psi(t))G(t, t') = -2KF(t, t', t) + \delta(t - t'),$$

$$(d/dt + 2K\Psi(t))V(t, t', t'') = -2KU(t, t')U(t, t'') - KW(t, t, t', t''),$$

Redundant equations for V in t' and t'' , (5.3)

$$(d/dt + 2K\Psi(t))F(t, t', t'') = -2KU(t, t'')G(t, t') - 2KE(t, t', t, t''),$$

$$(d/dt + 2K\Psi(t))W(t, t', t'', t''') = -2KU(t, t')V(t, t'', t''') - KX(t, t, t', t'', t'''),$$

Redundant equations for W in t' , t'' , and t''' , (5.4)

$$(d/dt + 2K\Psi(t))E(t, t', t'', t''') = -2KU(t, t''')F(t, t', t''') - 2KD(t, t', t, t'', t'''),$$

$$(d/dt + 2K\Psi(t))X(t, t', t'', t''', t^{iv}) = -2KU(t, t'')W(t, t''', t''', t^{iv}) - KY(t, t, t', t'', t''', t^{iv}),$$

Redundant equations for X in t' , t'' , and t^{iv} , (5.5)

$$(d/dt'' + 2K\Psi(t''))X(t, t', t'', t''', t^{iv}) = -2KV(t, t', t'')V(t'', t''', t^{iv})$$

$$- K\tilde{Y}(t, t', t'', t'', t''', t^{iv}),$$

$$(d/dt + 2K\Psi(t))D(t, t', t'', t''', t^{iv}) = -2KU(t, t'')E(t, t', t''', t^{iv})$$

$$- 2KC(t, t', t, t'', t''', t^{iv}).$$

Similarly, the moment equations of any order can be derived by the procedure of Sec. 4 so as to build up the hierarchy of unclosed statistical equations. We must point out that the first right-hand-side terms of (5.3)–(5.5) are the decompositions of the respective second right-hand-side terms into the lower-order moments of class A. In the hierarchy of moment equations formulated by Orszag [6], a similar decomposition has been achieved by invoking the ordering property of the Fourier modes in a homogeneous velocity field. His decomposition, however, does not differentiate the lower-order moments of class A from those of class B which are excluded from our hierarchy.

Considering the first right-hand-side terms of (5.3)–(5.5) as a small perturbation, we can close the statistical equations at any moment level by using the modal-interaction perturbation technique of [7].

Covariance level closure. For closure at the covariance level, we restrict ourselves to (5.1)–(5.3). Then, by treating the first right-hand-side terms of (5.3) as being small

in comparison to the second terms, we can write down the perturbation solutions by the following prescription, to be justified in the Appendix. For the V equation in t , we have the perturbation solution

$$V(t, t', t'') = \int_{t_0}^t ds G(t, s) [-2KU(t', s)U(t'', s)], \quad (5.6)$$

where the square bracket contains the first right-hand-side term of the V equation. And for the perturbation solution for F we have

$$F(t, t', t'') = \int_{t_0}^t ds G(t, s) [-2KU(t'', s)G(s, t')], \quad (5.7)$$

where the square bracket contains the first right-hand-side term of the F equation. In fact, (5.6) reflects the perturbation of V induced by $-2KU(t, t')U(t, t'')$, and similarly the perturbation of F due to $-2KU(t, t'')G(t, t')$ is expressed by (5.7). Since (5.6) represents the perturbation in t , similar perturbations in t' and t'' can be obtained from the redundant equations for V in those time arguments. Then, adding up the three perturbations, the perturbation solution reflecting evolution in three time arguments becomes

$$\begin{aligned} V(\bar{t}, \bar{t}', \bar{t}'') &= V(t_0) - 2K \int_{t_0}^t ds G(t, s) U(t', s) U(t'', s) \\ &\quad - 2K \int_{t_0}^{t'} ds G(t', s) U(t, s) U(t'', s) - 2K \int_{t_0}^{t''} ds G(t'', s) U(t, s) U(t', s), \end{aligned} \quad (5.8)$$

where $V(t_0) = V(t_0, t_0, t_0)$. Note that (5.7) is consistent with the initial condition $F(t', t', t'') = 0$.

Triple moment level closure. For the triple moment level of closure, we consider (5.1)–(5.4). Then, using the prescription similar to (5.6) and (5.7), the perturbation solutions of (5.4) can be written down at once:

$$\begin{aligned} W(\bar{t}, \bar{t}', \bar{t}'', \bar{t}''') &= W(t_0) - 2K \int_{t_0}^t ds G(t, s) U(t', s) V(t'', t''', s) \\ &\quad - 2K \int_{t_0}^{t'} ds G(t', s) U(t, s) V(t'', t''', s) - 2K \int_{t_0}^{t''} ds G(t'', s) U(t''', s) V(t, t', s) \\ &\quad - 2K \int_{t_0}^{t'''} ds G(t''', s) U(t'', s) V(t, t', s), \end{aligned} \quad (5.9)$$

where $W(t_0) = W(t_0, \dots)$, and

$$E(t, t', t'', t''') = -2K \int_{t_0}^t ds G(t, s) U(t'', s) F(s, t', t'''), \quad (5.10)$$

under $E(t', t', \dots) = 0$.

Quadruple moment level closure. We consider (5.1)–(5.5) for the quadruple moment level of closure. The perturbation solutions of (5.5) are

$$\begin{aligned}
 X(t, t', t'', t''', t^{iv}) = & X(t_0) - 2K \int_{t_0}^t ds G(t, s) U(t', s) W(t'', t''', t^{iv}, s) \\
 & - 2K \int_{t_0}^{t'} ds G(t', s) U(t, s) W(t'', t''', t^{iv}, s) \\
 & - 2K \int_{t_0}^{t''} ds G(t'', s) U(t^{iv}, s) W(t, t', t'', s) \\
 & - 2K \int_{t_0}^{t'''} ds G(t''', s) U(t''', s) W(t, t', t'', s) \\
 & - 2K \int_{t_0}^{t^{iv}} ds G(t^{iv}, s) U(t''', s) W(t, t', t'', s) \\
 & - 2K \int_{t_0}^{t'''} ds G(t''', s) V(t, t', s) V(t''', t^{iv}, s), \tag{5.11}
 \end{aligned}$$

where $X(t_0) = X(t_0, \dots)$, and

$$D(t, t', t'', t''', t^{iv}) = -2K \int_{t'}^{t''} ds G(t, s) U(t'', s) E(s, t', t''', t^{iv}), \tag{5.12}$$

under $D(t', t', \dots) = 0$.

6. The systems of the direct-interaction equations. We can now close (5.2) by introducing into the right-hand sides the perturbation solutions (5.7) and (5.8). To be consistent with the covariance level of formulation, the triple moment $V(t_0)$ in the final result must be suppressed. We then find that the statistical equations so closed are identical to Kraichnan's DI equations [1] for U and G . For this reason, we shall call the set of (5.1)–(5.3) closed by (5.9) and (5.10) the DI equations of the triple moment level and, similarly, the set of (5.1)–(5.4) closed by (5.11) and (5.12) the DI equations of the quadruple moment level. Our use of the DI approximation here has been motivated by the structural similarity. The DI systems of both the triple moment and quadruple moment levels do not, however, seem to satisfy the realizability requirement that Kraichnan [4] has exhibited for the DI equations of the covariance level from the existence of a model dynamic representation. It is not certain whether the existence of a model dynamic representation is so essential for a good turbulence theory. At any rate, let us denote by DI-(n) the DI equations closed at the n th-order moment level. For convenience, we set $t_0 = 0$ and define $\tau = K\Psi(0)t$. Further, $\hat{\Psi}(\tau) = \Psi(\tau)/\Psi(0)$, $\hat{U}(\) = U(\)/U(0)$, $\hat{V}(\) = V(\)/U(0)^{3/2}$, and $\hat{W}(\) = W(\)/U(0)^2$. Then the first three systems of the DI equations containing the parameter $\alpha = \Psi(0)/U(0)^{1/2}$ become DI-(2)

$$(d/d\tau + \hat{\Psi}(\tau))\hat{\Psi}(\tau) = -\alpha^{-2}\hat{U}(\tau), \tag{6.1}$$

$$\begin{aligned}
 (d/d\tau + 2\hat{\Psi}(\tau))\hat{U}(\tau, \tau') \\
 = 4\alpha^{-2} \int_0^\tau ds G(\tau, s)\hat{U}(\tau, s)\hat{U}(\tau', s) + 2\alpha^{-2} \int_0^{\tau'} ds G(\tau', s)\hat{U}(\tau, s)^2, \tag{6.2}
 \end{aligned}$$

$$(d/d\tau + 2\hat{\Psi}(\tau))G(\tau, \tau') = 4\alpha^{-2} \int_{\tau'}^\tau ds G(\tau, s)\hat{U}(\tau, s)G(s, \tau') + \delta(\tau - \tau'). \tag{6.3}$$

DI-(3)

$\hat{\Psi}$ —Eq. (6.1),

$$(d/d\tau + 2\hat{\Psi}(\tau))\hat{U}(\tau, \tau') = -\alpha^{-1}\hat{V}(\tau, \tau, \tau'), \tag{6.4}$$

$$(d/d\tau + 2\hat{\Psi}(\tau))G(\tau, \tau') = -2\alpha^{-1}F(\tau, \tau', \tau) + \delta(\tau - \tau'), \quad (6.5)$$

$$(d/d\tau + 2\hat{\Psi}(\tau))\hat{V}(\tau, \tau', \tau'')$$

$$\begin{aligned} &= -2\alpha^{-1}\hat{U}(\tau, \tau'')\hat{U}(\tau, \tau') + 4\alpha^{-2} \int_0^\tau ds G(\tau, s)\hat{U}(\tau, s)\hat{V}(\tau', \tau'', s) \\ &\quad + 2\alpha^{-2} \int_0^{\tau'} ds G(\tau', s)\hat{U}(\tau'', s)\hat{V}(\tau, \tau, s) \\ &\quad + 2\alpha^{-2} \int_0^{\tau''} ds G(\tau'', s)\hat{U}(\tau', s)\hat{V}(\tau, \tau, s), \end{aligned} \quad (6.6)$$

$$(d/d\tau + 2\hat{\Psi}(\tau))F(\tau, \tau', \tau'')$$

$$= -2\alpha^{-1}\hat{U}(\tau, \tau'')G(\tau, \tau') + 4\alpha^{-2} \int_{\tau'}^\tau ds G(\tau, s)\hat{U}(\tau, s)F(s, \tau', \tau''). \quad (6.7)$$

DI-(4)

$\hat{\Psi}$ —Eq. (6.1), \hat{U} —Eq. (6.4), G —Eq. (6.5),

$$(d/d\tau + 2\hat{\Psi}(\tau))\hat{V}(\tau, \tau', \tau'') = -2\alpha^{-1}\hat{U}(\tau, \tau'')\hat{U}(\tau, \tau') - \alpha^{-1}\hat{W}(\tau, \tau, \tau', \tau''), \quad (6.8)$$

$$(d/d\tau + 2\hat{\Psi}(\tau))F(\tau, \tau', \tau'') = -2\alpha^{-1}\hat{U}(\tau, \tau'')G(\tau, \tau') - 2\alpha^{-1}E(\tau, \tau', \tau, \tau''), \quad (6.9)$$

$$(d/d\tau + 2\hat{\Psi}(\tau))\hat{W}(\tau, \tau', \tau'', \tau''') = -2\alpha^{-1}\hat{U}(\tau, \tau')\hat{V}(\tau, \tau'', \tau''')$$

$$\begin{aligned} &\quad + 4\alpha^{-2} \int_0^\tau ds G(\tau, s)\hat{U}(\tau, s)\hat{W}(\tau', \tau'', \tau''', s) \\ &\quad + 2\alpha^{-2} \int_0^{\tau'} ds G(\tau', s)\hat{V}(\tau, \tau, s)\hat{V}(\tau'', \tau''', s) \\ &\quad + 2\alpha^{-2} \int_0^{\tau''} ds G(\tau'', s)\hat{U}(\tau''', s)\hat{W}(\tau', \tau, \tau, s) \\ &\quad + 2\alpha^{-2} \int_0^{\tau'''} ds G(\tau''', s)\hat{U}(\tau'', s)\hat{W}(\tau', \tau, \tau, s), \end{aligned} \quad (6.10)$$

$$(d/d\tau + 2\hat{\Psi}(\tau))E(\tau, \tau', \tau'', \tau''') = -2\alpha^{-1}\hat{U}(\tau, \tau'')F(\tau, \tau', \tau''')$$

$$+ 4\alpha^{-2} \int_{\tau'}^\tau ds G(\tau, s)\hat{U}(\tau, s)E(s, \tau', \tau'', \tau'''). \quad (6.11)$$

Initially, $\hat{\Psi}(0) = \hat{U}(0) = 1$ by the normalization, whereas the $\hat{V}(0)$ and $\hat{W}(0)$ will have to be specified by the initial distribution. Furthermore, we have $G(\tau', \tau') = 1$, $F(\tau, \tau', \cdot) = 0$, and $E(\tau', \tau', \cdot) = 0$.

7. Series solutions of three direct-interaction systems. The generalized DI approximation has closed the hierarchy of moment equations by the convolution type of integrals involving only the known statistical functions at each level of closure. Since these integrals represent summation of certain classes of the expansion terms, it is essential that the nonlinear integral equations of DI-(2), DI-(3), and DI-(4) be solved by means other than the series expansion technique. For instance, the DI-(2) has already been integrated numerically in an isotropic mixing field [1], and a similar numerical

scheme can be extended to both DI-(3) and DI-(4), although they are substantially more complicated. However, for the purpose of exhibiting the structure of the DI systems, we shall propose here to obtain the series solutions of the form

$$\begin{aligned} \hat{\Psi}(\tau) &= \hat{\Psi}_0(\tau) + \alpha^{-1}\hat{\Psi}_1(\tau) + \alpha^{-2}\hat{\Psi}_2(\tau) + \dots, \\ \hat{U}(\tau, \tau') &= \hat{U}_0(\tau, \tau') + \alpha^{-1}\hat{U}_1(\tau, \tau') + \alpha^{-2}\hat{U}_2(\tau, \tau') + \dots, \\ G(\tau, \tau') &= G_0(\tau, \tau') + \alpha^{-1}G_1(\tau, \tau') + \alpha^{-2}G_2(\tau, \tau') + \dots, \end{aligned} \tag{7.1}$$

No similar expressions exist for \hat{V} , F , etc. For the consistent initial conditions, the zeroth orders retain the exact initial values and all remaining higher orders have the zero initial values:

$$\begin{aligned} \hat{\Psi}_0(0) &= \hat{\Psi}(0), & \hat{U}_0(0) &= \hat{U}(0), & G_0(\tau', \tau') &= 1, \dots, \\ \hat{\Psi}_i(0) &= 0, & \hat{U}_i(0) &= 0, & G_i(\tau', \tau') &= 0, \dots \text{ for } i \geq 1, \end{aligned} \tag{7.2}$$

where the dots denote the initial conditions for \hat{V} , F , etc.

7.1 *The DI-(2) system.* Introduce (7.1) into the DI-(2) and sort out successively three equations of the same order. For the zeroth-order set, we have

$$(d/d\tau + \hat{\Psi}_0(\tau))\hat{\Psi}_0(\tau) = 0, \tag{7.3}$$

$$(d/d\tau + 2\hat{\Psi}_0(\tau))\hat{U}_0(\tau, \tau') = 0, \tag{7.4}$$

$$(d/d\tau + 2\hat{\Psi}_0(\tau))G_0(\tau, \tau') = \delta(\tau - \tau'). \tag{7.5}$$

Since the solution of (7.3) is $\hat{\Psi}_0(\tau) = (1 + \tau)^{-1}$, it is convenient to introduce a new variable $\eta = 1 + \tau$ into (7.4) and (7.5):

$$(d/d\eta + 2\eta^{-1})\hat{U}_0(\eta, \eta') = 0, \tag{7.6}$$

$$(d/d\eta + 2\eta^{-1})G_0(\eta, \eta') = \delta(\eta - \eta'). \tag{7.7}$$

Integrating (7.6) from $1 \rightarrow \eta$ and then the redundant equation for U_0 in η' from $1 \rightarrow \eta'$, we have

$$\hat{U}_0(\eta, \eta') = \hat{U}(1)\eta^{-2}\eta'^{-2}. \tag{7.8}$$

And the solution of (7.7) is

$$G_0(\eta, \eta') = (\eta'/\eta)^2. \tag{7.9}$$

In view of (7.2), the first-order set is vacuous: $\hat{\Psi}_1 = \hat{U}_1 = G_1 = 0$. Indeed, this is expected from the fact that DI-(2) contains the parameter α^{-2} ; hence all the odd-order sets are likewise vacuous. For the second-order set, we have

$$(d/d\eta + 2\eta^{-1})\hat{\Psi}_2(\eta) = -\hat{U}_0(\eta), \tag{7.10}$$

$$\begin{aligned} (d/d\eta + 2\eta^{-1})\hat{U}_2(\eta, \eta') &= -2\hat{\Psi}_2(\eta)\hat{U}_0(\eta, \eta') + 4 \int_1^\eta ds G_0(\eta, s)\hat{U}_0(\eta, s)\hat{U}_0(\eta', s) \\ &\quad + 2 \int_1^{\eta'} ds G_0(\eta', s)\hat{U}_0(\eta, s)\hat{U}_0(\eta, s), \end{aligned} \tag{7.11}$$

$$(d/d\eta + 2\eta^{-1})G_2(\eta, \eta') = -2\hat{\Psi}_2(\eta)G_0(\eta, \eta') + 4 \int_1^\eta ds G_0(\eta, s)\hat{U}_0(\eta, s)G_0(s, \eta). \tag{7.12}$$

First of all, (7.10) gives

$$\hat{\Psi}_2(\eta) = -\hat{U}(1)\eta^{-2}(1 - \eta^{-1}). \quad (7.13)$$

With this $\hat{\Psi}_2$, the right-hand side of (7.11), denoted by $Q(\eta, \eta')$, is known explicitly. Let us consider (7.11) and the redundant equation in η' :

$$(d/d\eta + 2\eta^{-1})\hat{U}_2(\eta, \eta') = Q(\eta, \eta'), \quad (7.14a)$$

$$(d/d\eta' + 2\eta'^{-1})\hat{U}_2(\eta, \eta') = Q(\eta', \eta). \quad (7.14b)$$

We first integrate (7.14a) from $1 \rightarrow \eta$ and then 7.14(b) from $1 \rightarrow \eta'$:

$$\hat{U}_2(\eta, \eta') = \eta^{-2}\eta'^{-2} \int_1^\eta \xi^2 Q(\xi, 1) d\xi + \eta'^{-2} \int_1^{\eta'} \xi^2 Q(\xi, \eta) d\xi.$$

Or, after some manipulations, we have

$$\hat{U}_2(\eta, \eta') = \hat{U}(1)^2\eta^{-2}\eta'^{-2}[3(1 - \eta^{-1})^2 + 3(1 - \eta'^{-1})^2 + 2(1 - \eta^{-1})(1 - \eta'^{-1})]. \quad (7.15)$$

For $\eta = \eta'$, (7.15) gives the simultaneous-time covariance

$$\hat{U}_2(\eta) = 8\hat{U}(1)^2\eta^{-4}(1 - \eta^{-1})^2. \quad (7.16)$$

Note that (7.16) could have been obtained more directly from the following equation

$$(d/d\eta + 4\eta^{-1})\hat{U}_2(\eta) = 2Q(\eta), \quad (7.17)$$

which was obtained by adding up the two equations of (7.14) and then setting $\eta = \eta'$. The nonsimultaneous-time information is lost in (7.17) because it evolves along the diagonal of the $\eta - \eta'$ plane. However, this is a useful short cut that we shall use for the computation of the simultaneous-time moments. Lastly, to complete the solution of the second-order set, we obtain from (7.12)

$$G_2(\eta, \eta') = 2\hat{U}(1)(\eta'/\eta)^2[(\eta'^{-1} - \eta^{-1}) - \frac{1}{2}(\eta'^{-2} - \eta^{-2}) + (\eta'^{-1} - \eta^{-1})^2]. \quad (7.18)$$

As pointed out previously, the third-order set is vacuous. Then, the mean value and simultaneous-time covariance up to the fourth-order become

$$\begin{aligned} \hat{\Psi}(\eta) &= \eta^{-1}\{1 - \hat{U}(1)\eta^{-1}(1 - \eta^{-1})/\alpha^2 - 3\hat{U}(1)^2\eta^{-1}(1 - \eta^{-1})^3/\alpha^4 + \dots\}, \\ \hat{U}(\eta) &= \eta^{-4}\{\hat{U}(1) + 8\hat{U}(1)^2((1 - \eta^{-1})/\alpha)^2 + 45\hat{U}(1)^3((1 - \eta^{-1})/\alpha)^4 + \dots\}, \end{aligned} \quad (7.19)$$

where the three dots denote the higher-order terms in even powers of α^{-1} .

7.2 The DI-(3) system. By repeating the series solution procedure of Sec. 7.1, we obtain the following results for DI-(3):

$$\begin{aligned} \hat{\Psi}(\eta) &= \eta^{-1}\left\{1 - \hat{U}(1)\eta^{-1}\frac{(1 - \eta^{-1})}{\alpha^2} + \hat{V}(1)\eta^{-1}\frac{(1 - \eta^{-1})^2}{\alpha^3} \right. \\ &\quad \left. - 3\hat{U}(1)^2\eta^{-1}\frac{(1 - \eta^{-1})^3}{\alpha^4} + \dots\right\}, \\ \hat{U}(\eta) &= \eta^{-4}\left\{\hat{U}(1) - 2\hat{V}(1)\left(\frac{1 - \eta^{-1}}{\alpha}\right) + 8\hat{U}(1)^2\left(\frac{1 - \eta^{-1}}{\alpha}\right)^2 - 22\hat{U}(1)\hat{V}(1)\left(\frac{1 - \eta^{-1}}{\alpha}\right)^3 \right. \\ &\quad \left. + (45\hat{U}(1)^3 + 13\hat{V}(1)^2)\left(\frac{1 - \eta^{-1}}{\alpha}\right)^4 + \dots\right\}, \end{aligned}$$

$$\hat{V}(\eta) = \eta^{-6} \left\{ \hat{V}(1) - 6\hat{U}(1)^2 \left(\frac{1-\eta^{-1}}{\alpha} \right) + 27\hat{U}(1)\hat{V}(1) \left(\frac{1-\eta^{-1}}{\alpha} \right)^2 - (68\hat{U}(1)^3 + 22\hat{V}(1)^2) \left(\frac{1-\eta^{-1}}{\alpha} \right)^3 + \dots \right\}, \quad (7.20)$$

where the three dots denote the higher-order terms whose coefficients involve $\hat{U}(1)$ and/or $\hat{V}(1)$. By comparing (7.19) with (7.20), we find that the DI-(2) is embedded in the DI-(3). In other words, the $\hat{\Psi}(\eta)$ and $\hat{U}(\eta)$ of (7.19) reappear in (7.20) as the terms whose coefficients involve only the $\hat{U}(1)$.

7.3 *The DI-(4) system.* Finally, the series solution of DI-(4) yields

$$\begin{aligned} \hat{\Psi}(\eta) &= \eta^{-1} \left\{ 1 - \hat{U}(1)\eta^{-1} \frac{(1-\eta^{-1})}{\alpha^2} + \hat{V}(1)\eta^{-1} \frac{(1-\eta^{-1})^2}{\alpha^3} - (\hat{W}(1) + 3\hat{U}(1)^2)\eta^{-1} \frac{(1-\eta^{-1})^3}{\alpha^4} + \dots \right\}, \\ \hat{U}(\eta) &= \eta^{-4} \left\{ \hat{U}(1) - 2\hat{V}(1) \left(\frac{1-\eta^{-1}}{\alpha} \right) + (3\hat{W}(1) + 8\hat{U}(1)^2) \left(\frac{1-\eta^{-1}}{\alpha} \right)^2 - 22\hat{U}(1)\hat{V}(1) \left(\frac{1-\eta^{-1}}{\alpha} \right)^3 + (45\hat{U}(1)^3 + 13\hat{V}(1)^2 + 29\hat{U}(1)\hat{W}(1)) \left(\frac{1-\eta^{-1}}{\alpha} \right)^4 + \dots \right\}, \\ \hat{V}(\eta) &= \eta^{-6} \left\{ \hat{V}(1) - (3\hat{W}(1) + 6\hat{U}(1)^2) \left(\frac{1-\eta^{-1}}{\alpha} \right) + 27\hat{U}(1)\hat{V}(1) \left(\frac{1-\eta^{-1}}{\alpha} \right)^2 - (68\hat{U}(1)^3 + 22\hat{V}(1)^2 + 50\hat{U}(1)\hat{W}(1)) \left(\frac{1-\eta^{-1}}{\alpha} \right)^3 + \dots \right\}. \end{aligned} \quad (7.21)$$

The solution for $\hat{W}(\eta)$ will not be presented here. Again, we observe that (7.20) is embedded in (7.21). Therefore, from the structure of the lower-order DI systems, it may be inferred that the DI-(n) is properly embedded in the DI-($n + 1$). This shell structure of the DI systems will be taken as the consistency of the generalized DI approximation. In closing, we must point out that the terms which are not underscored in (7.21) can be traced to the zeroth orders, \hat{U}_0 , \hat{V}_0 , and \hat{W}_0 ; hence, they reflect the initial moment values. On the other hand, the underscored terms can be attributed to the nonlinearity of the problem.

8. **Expansion and partial summation of the exact moments.** In order to provide a term-by-term comparison of the DI approximation results with the exact moments, we transform (1.3)–(1.5) into the same form as (7.21). Using the notations of Sec. 6, we have

$$\hat{\Psi}(\tau) = \int \left(\frac{x}{\Psi(0) + \tau x} \right) dP(x), \quad (8.1)$$

$$\hat{U}(\tau) = \alpha^2 \left\{ \int \left(\frac{x}{\Psi(0) + \tau x} \right)^2 dP(x) - \hat{\Psi}(\tau)^2 \right\}, \quad (8.2)$$

$$\hat{V}(\tau) = \alpha^3 \left\{ \int \left(\frac{x}{\Psi(0) + \tau x} \right)^3 dP(x) - 3\hat{\Psi}(\tau) \int \left(\frac{x}{\Psi(0) + \tau x} \right)^2 dP(x) + 2\hat{\Psi}(\tau)^3 \right\}, \quad (8.3)$$

and similar expressions can be written for $\hat{W}(\tau)$, $\hat{X}(\tau)$, and $\hat{Y}(\tau)$.

8.1 *Expansion in τ .* Expanding the factors $(\Psi(0) + \tau x)^{-n}$ in powers of τ , we may put (8.1)–(8.3) in the form

$$\hat{\Psi}(\tau) = \Psi(0)^{-1} \left\{ M_1 - M_2 \left(\frac{\tau}{\Psi(0)} \right) + M_3 \left(\frac{\tau}{\Psi(0)} \right)^2 - M_4 \left(\frac{\tau}{\Psi(0)} \right)^3 + \dots \right\}, \quad (8.4)$$

$$\begin{aligned} \hat{U}(\tau) = & \left(\frac{\alpha}{\Psi(0)} \right)^2 \left\{ M_2 - 2M_3 \left(\frac{\tau}{\Psi(0)} \right) + 3M_4 \left(\frac{\tau}{\Psi(0)} \right)^2 - 4M_5 \left(\frac{\tau}{\Psi(0)} \right)^3 + 5M_6 \left(\frac{\tau}{\Psi(0)} \right)^4 + \dots \right. \\ & - \left[M_1^2 - 2M_1 M_2 \left(\frac{\tau}{\Psi(0)} \right) + (2M_1 M_3 + M_2^2) \left(\frac{\tau}{\Psi(0)} \right)^2 - 2(M_1 M_4 + M_2 M_3) \left(\frac{\tau}{\Psi(0)} \right)^3 \right. \\ & \left. \left. + (M_3^2 + 2M_1 M_5 + 2M_2 M_4) \left(\frac{\tau}{\Psi(0)} \right)^4 + \dots \right] \right\}, \quad (8.5) \end{aligned}$$

$$\begin{aligned} \hat{V}(\tau) = & \left(\frac{\alpha}{\Psi(0)} \right)^3 \left\{ M_3 - 3M_4 \left(\frac{\tau}{\Psi(0)} \right) + 6M_5 \left(\frac{\tau}{\Psi(0)} \right)^2 - 10M_6 \left(\frac{\tau}{\Psi(0)} \right)^3 + \dots \right. \\ & - 3 \left[M_1 M_2 - (2M_1 M_3 + M_2^2) \left(\frac{\tau}{\Psi(0)} \right) + 3(M_1 M_4 + M_2 M_3) \left(\frac{\tau}{\Psi(0)} \right)^2 \right. \\ & \left. - 2(2M_1 M_5 + 2M_2 M_4 + M_3^2) \left(\frac{\tau}{\Psi(0)} \right)^3 + \dots \right] \\ & + 2 \left[M_1^3 - 3M_1^2 M_2 \left(\frac{\tau}{\Psi(0)} \right) + 3(M_1^2 M_3 + M_1 M_2^2) \left(\frac{\tau}{\Psi(0)} \right)^2 \right. \\ & \left. \left. - (3M_1^2 M_4 + 6M_1 M_2 M_3 + M_2^3) \left(\frac{\tau}{\Psi(0)} \right)^3 + \dots \right] \right\}, \quad (8.6) \end{aligned}$$

where $M_m = \int x^m dP(x)$. Since the M 's are the moments about the origin, they can be expressed in terms of the moments about the mean. To do this requires setting $\tau = 0$ in (8.1)–(8.3) and solving successively for the M 's:

$$M_1 = \Psi(0), \quad M_2 = \Psi(0)^2 \left(\frac{\hat{U}(0)}{\alpha^2} + 1 \right), \quad M_3 = \Psi(0)^3 \left(\frac{\hat{V}(0)}{\alpha^3} + \frac{3\hat{U}(0)}{\alpha^2} + 1 \right), \quad (8.7)$$

and $M_4 - M_6$ can be expressed similarly from the \hat{W} , \hat{X} , and \hat{Y} .

Now introduce the M 's into (8.4)–(8.6). After regrouping the terms of the same powers in α^{-1} , the resulting expansions may be rearranged in the form

$$\begin{aligned} \hat{\Psi}(\tau) = & (1 - \tau + \tau^2 - \tau^3 + \tau^4 + \dots) - \hat{U}(0)(\tau/\alpha^2)(1 - 3\tau + 6\tau^2 - 10\tau^3 + \dots) \\ & + \hat{V}(0)(\tau^2/\alpha^3)(1 - 4\tau + 10\tau^2 + \dots) \\ & - \hat{W}(0)(\tau^3/\alpha^4)(1 - 5\tau + \dots) + O(\alpha^{-5}), \quad (8.8) \end{aligned}$$

$$\begin{aligned} \hat{U}(\tau) = & \hat{U}(0)(1 - 4\tau + 10\tau^2 - 20\tau^3 + 35\tau^4 + \dots) \\ & - 2\hat{V}(0)(\tau/\alpha)(1 - 5\tau + 15\tau^2 - 35\tau^3 + \dots) \\ & + (3\hat{W}(0) - \hat{U}(0)^2)(\tau/\alpha)^2(1 - 6\tau + 21\tau^2 + \dots) \\ & - (4\hat{X}(0) - 2\hat{U}(0)\hat{V}(0))(\tau/\alpha)^3(1 - 7\tau + \dots) \\ & + (5\hat{Y}(0) - \hat{V}(0)^2 - 2\hat{U}(0)\hat{W}(0))(\tau/\alpha)^4(1 + \dots) + O(\alpha^{-5}), \quad (8.9) \end{aligned}$$

$$\begin{aligned}
 \hat{V}(\tau) &= \hat{V}(0)(1 - 6\tau + 21\tau^2 - 56\tau^3 + \dots) \\
 &\quad - 3(\hat{W}(0) - \hat{U}(0)^2)(\tau/\alpha)(1 - 7\tau + 28\tau^2 + \dots) \\
 &\quad + 3(2\hat{X}(0) - 3\hat{U}(0)\hat{V}(0))(\tau/\alpha)^2(1 - 8\tau + \dots) \\
 &\quad - (10\hat{Y}(0) - 12\hat{U}(0)\hat{W}(0) - 6\hat{V}(0)^2 + 2\hat{U}(0)^3)(\tau/\alpha)^3(1 + \dots) + O(\alpha^{-4}).
 \end{aligned} \tag{8.10}$$

8.2 *Summation of the τ expansions.* At this point, we may consider (8.8)–(8.10) as the two-dimensional expansions in τ and α^{-1} . However; the τ expansions can be summed up to all orders by noting $(1 + \tau)^{-n} = 1 - n\tau + n(n + 1)\tau^2/2! - \dots$. Under the assumption that the τ expansions in (8.8)–(8.10) represent the leading terms of infinite series, we can replace them by the $(1 + \tau)^{-n}$ factors and obtain the consolidated expressions

$$\begin{aligned}
 \hat{\Psi}(\tau) &= (1 + \tau)^{-1} - \hat{U}(0)(\tau/\alpha^2)(1 + \tau)^{-3} + \hat{V}(0)(\tau^2/\alpha^3)(1 + \tau)^{-4} \\
 &\quad - \hat{W}(0)(\tau^3/\alpha^4)(1 + \tau)^{-5} + O(\alpha^{-5}).
 \end{aligned} \tag{8.11}$$

$$\begin{aligned}
 \hat{U}(\tau) &= \hat{U}(0)(1 + \tau)^{-4} - 2\hat{V}(0)(\tau/\alpha)(1 + \tau)^{-5} + (3\hat{W}(0) - \hat{U}(0)^2)(\tau/\alpha)^2(1 + \tau)^{-6} \\
 &\quad - (4\hat{X}(0) - 2\hat{U}(0)\hat{V}(0))(\tau/\alpha)^3(1 + \tau)^{-7} \\
 &\quad + (5\hat{Y}(0) - \hat{V}(0)^2 - 2\hat{U}(0)\hat{W}(0))(\tau/\alpha)^4(1 + \tau)^{-8} + O(\alpha^{-5}),
 \end{aligned} \tag{8.12}$$

$$\begin{aligned}
 \hat{V}(\tau) &= \hat{V}(0)(1 + \tau)^{-6} - 3(\hat{W}(0) - \hat{U}(0)^2)(\tau/\alpha)(1 + \tau)^{-7} \\
 &\quad + 3(2\hat{X}(0) - 3\hat{U}(0)\hat{V}(0))(\tau/\alpha)^2(1 + \tau)^{-8} \\
 &\quad - (10\hat{Y}(0) - 12\hat{U}(0)\hat{W}(0) - 6\hat{V}(0)^2 + 2\hat{U}(0)^3)(\tau/\alpha)^3(1 + \tau)^{-9} + O(\alpha^{-4}).
 \end{aligned} \tag{8.13}$$

Finally, we introduce $\eta = 1 + \tau$ into (8.11)–(8.13) and suppress both the $\hat{X}(0)$ and $\hat{Y}(0)$, which are superfluous for the quadruple-moment level of closure:

$$\begin{aligned}
 \hat{\Psi}(\eta) &= \eta^{-1} \left\{ 1 - \hat{U}(1)\eta^{-1} \frac{(1 - \eta^{-1})}{\alpha^2} \right. \\
 &\quad \left. + \hat{V}(1)\eta^{-1} \frac{(1 - \eta^{-1})^2}{\alpha^3} - \hat{W}(1)\eta^{-1} \frac{(1 - \eta^{-1})^3}{\alpha^4} + \dots \right\},
 \end{aligned} \tag{8.14}$$

$$\begin{aligned}
 \hat{U}(\eta) &= \eta^{-4} \left\{ \hat{U}(1) - 2\hat{V}(1) \left(\frac{1 - \eta^{-1}}{\alpha} \right) \right. \\
 &\quad + (3\hat{W}(1) - \hat{U}(1)^2) \left(\frac{1 - \eta^{-1}}{\alpha} \right)^2 + 2\hat{U}(1)\hat{V}(1) \left(\frac{1 - \eta^{-1}}{\alpha} \right)^3 \\
 &\quad \left. - (2\hat{U}(1)\hat{W}(1) + \hat{V}(1)^2) \left(\frac{1 - \eta^{-1}}{\alpha} \right)^4 + \dots \right\},
 \end{aligned} \tag{8.15}$$

$$\begin{aligned}
 \hat{V}(\eta) &= \eta^{-6} \left\{ \hat{V}(1) - (3\hat{W}(1) - 3\hat{U}(1)^2) \left(\frac{1 - \eta^{-1}}{\alpha} \right) - 9\hat{U}(1)\hat{V}(1) \left(\frac{1 - \eta^{-1}}{\alpha} \right)^2 \right. \\
 &\quad \left. + (12\hat{U}(1)\hat{W}(1) + 6\hat{V}(1)^2 - 2\hat{U}(1)^3) \left(\frac{1 - \eta^{-1}}{\alpha} \right)^3 + \dots \right\}.
 \end{aligned} \tag{8.16}$$

In analogy to (7.21), we have underscored the terms whose coefficients are the products of initial moments. By comparing the underscored terms in the above with those in (7.21), we see at once how the classes of expansion terms summed up in the DI approximation are different from those present in the exact moment solutions. In particular, for $\hat{V}(1) = \hat{W}(1) = 0$, (8.15) gives

$$\hat{U}(\eta) = \eta^{-4} \{ \hat{U}(1) - \hat{U}(1)^2 ((1 - \eta^{-1})/\alpha)^2 \}, \quad (8.17)$$

which should be compared with the covariance of DI-(2) given in (7.19). Note that (8.17) is a polynomial in $(1 - \eta^{-1})/\alpha$ with the negative second-order term, whereas the covariance of (7.19) is an infinite series with the positive coefficients.

9. Concluding remarks. Unlike the Burgers-model turbulence, the stochastic nonlinear system (1.1) cannot serve as a meaningful prototype of the real turbulence because it does not share certain dynamical features with the Navier–Stokes equations. The dynamic system described by (1.1) is purely dissipative. Furthermore, the absence of the convection term in (1.1) deprives the present problem of many of the dynamic features, such as the energy conservation, equipartition energy state, and the Galilean invariance, which distinguish the turbulent flow problem from other stochastic processes. Nevertheless, the quadratic nonlinearity of (1.1) causes the same sort of closure difficulty as the Navier–Stokes equations. Hence we can investigate the analytical structure of the generalized DI approximation in depth, thanks to the mathematical amenability of (1.1).

From the standpoint of the moment formulation, it is not surprising that the DI-(3) represents a meaningful statistical approximation of the lowest order for the second-order reactive problem. This is because the DI-(2) collects only the terms whose coefficients are of the even powers in $(-K)$; hence the distinction is completely lost between the depletive and generative type of chemical reactions. On the other hand, the DI-(3) can adequately describe the decay of the reactant fluctuation intensity, for it can cope with an asymmetric initial distribution which extends only to the positive range, to be consistent with the requirement that $\psi(t)$ is a positive random variable. For quantitative comparisons, we consider for $P(x)$ the asymmetric Helmert distribution [5]

$$P(x) = \frac{1}{n!} \int_0^x y^n e^{-y} dy. \quad (9.1)$$

Here, for convenience, we take $n = \frac{1}{2}, 1, \frac{3}{2}, \dots$. O'Brien [8] has used (9.1) with $n = 3.5$ to test his closure formula. For the initial distribution (9.1), we have

$$\Psi(0) = \alpha^2, \quad \hat{V}(0) = 2/\alpha, \quad \hat{W}(0) = 3(1 + 2/\alpha^2), \quad (9.2)$$

where $\alpha^2 = n + 1$. In the limit as $n \rightarrow \infty$, (9.1) reduces to a Gaussian distribution. Although the series solutions of the DI systems obtained in Sec. 7 were not meant for computational purposes, we shall use them here to provide comparison of the covariances. To avoid any convergence difficulty, we take a large value of $\alpha = 3.0$. Fig. 5 compares the three covariances of the form $\hat{U}(\tau)(1 + \tau)^4$ computed by the second expressions of (7.19), (7.20), and (7.21), respectively. Also included in the figure as a reference is the exact covariance computed by (8.2). In support of the previous discussion, the DI-(3) gives the best approximation to the exact covariance for the entire time range of the figure. On the other hand, both the DI-(2) and DI-(4) appear to be the moment formulations of the wrong order for the present reactive problem. As mentioned before,

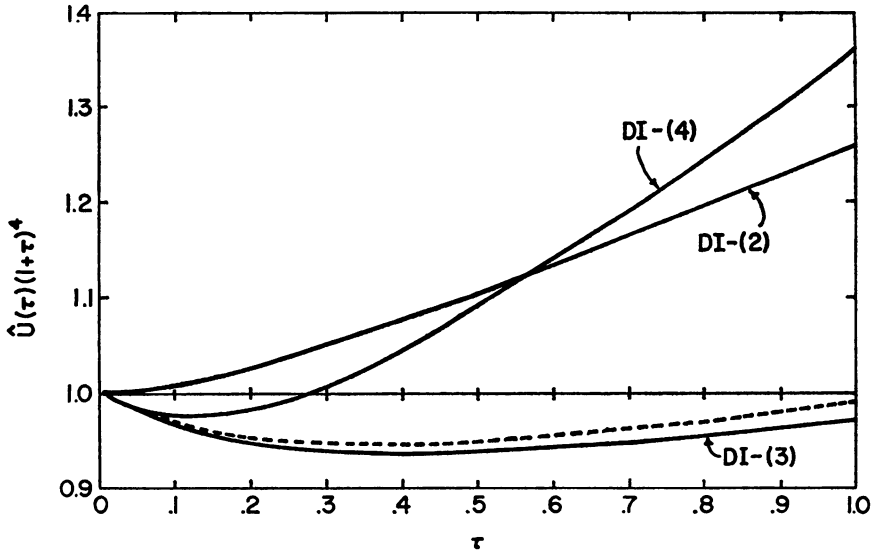


Fig. 5. Comparison of covariances.— direct-interaction approximation; — exact covariance.

the inadequacy of DI-(2) can be ascribed to the imposition of a symmetric initial distribution that reactant fluctuations cannot physically obey, whereas the failure of DI-(4) is, perhaps, a manifestation that summing up more expansion terms does not necessarily give a better approximation [3]. The mean value $\Psi(t)$ is rather insensitive to the closure scheme, for the effect of fluctuations is only secondary in the mean equation (6.1).

Appendix: The modal-interaction perturbation technique. Using the modal-interaction perturbation technique of [7], we can obtain the perturbation solutions of (5.3)–(5.5) to express the effect of the respective first right-hand-side terms treated as a small perturbation. Without repetition, we shall discuss here only the solution of (5.3) in detail because the solution of (5.4) and (5.5) can be carried out in a completely analogous manner. Returning to (3.1) and (3.2), the equations for $\psi_\beta(t)$ and $G_{\alpha-\beta,\alpha}(t, t')$ are respectively

$$(d/dt + 2K\Psi(t))\psi_\beta(t) = -KM^{-1/2} \sum_{\sigma}'' \phi_{\beta,\sigma,\beta-\sigma}\psi_\sigma(t)\psi_{\beta-\sigma}(t), \tag{A1}$$

$$(d/dt + 2K\Psi(t))G_{\alpha-\beta,\alpha}(t, t') = -2KM^{-1/2} \sum_{\sigma}'' \phi_{\alpha-\beta,\sigma,\alpha-\beta-\sigma}\psi_\sigma(t)G_{\alpha-\beta-\sigma,\alpha}(t, t'). \tag{A2}$$

Rewrite the above as

$$(d/dt + 2K\Psi(t))\psi_\beta(t) = -2KM^{-1/2}\phi_{\beta,\alpha,\beta-\alpha}\psi_\alpha(t)\psi_{\alpha-\beta}^*(t) - KM^{-1/2} \sum_{\substack{\sigma \neq \alpha \\ \sigma \neq \beta - \alpha}}'' \phi_{\beta,\sigma,\beta-\sigma}\psi_\sigma(t)\psi_{\beta-\sigma}(t), \tag{A3}$$

$$(d/dt + 2K\Psi(t))G_{\alpha-\beta,\alpha}(t, t') = -2KM^{-1/2}\phi_{\alpha-\beta,-\beta,\alpha}\psi_\beta^*(t)G_{\alpha,\alpha}(t, t') - 2KM^{-1/2} \sum_{\sigma \neq \beta}'' \phi_{\alpha-\beta,\sigma,\alpha-\beta-\sigma}\psi_\sigma(t)G_{\alpha-\beta-\sigma,\alpha}(t, t'). \tag{A4}$$

Observe the following: if we apply $M^{-3/2} \sum'_\alpha \sum''_{\beta'} \phi_{\alpha,\beta,\alpha-\beta} \psi_{\alpha-\beta}(t') \psi_\alpha^*(t'')$ to (A3), then the resulting equation after averaging is simply (4.4). Further, we can obtain (4.6) by applying $M^{-3/2} \sum'_\alpha \sum''_{\beta'} \phi_{\alpha,\beta,\alpha-\beta} \psi_\beta(t'')$ to (A4) and averaging the resulting equation. Here (2.14) and (2.17) have been used. Since the first right-hand-side terms of (A3) and (A4) are of $O(M^{-1/2})$, this justifies our claim that the first right-hand-side terms of (5.3) are small in comparison to the second terms of the equations. It has been shown in [7] that the perturbation $\Delta\psi_\beta(t)$ of $O(M^{-1/2})$ due to the first right-hand-side term of (A3) is

$$\Delta\psi_\beta(t) = -2KM^{-1/2} \int_{t_0}^t ds \phi_{\beta,\alpha,\beta-\alpha} G_{\beta,\beta}(t,s) \psi_\alpha(s) \psi_{\alpha-\beta}^*(s). \quad (\text{A5})$$

Upon applying $M^{-3/2} \sum'_\alpha \sum''_{\beta'} \phi_{\alpha,\beta,\alpha-\beta} \psi_{\alpha-\beta}(t') \psi_\alpha^*(t'')$ to (A5) and averaging, we obtain the perturbation $\Delta V(t, t', t'')$ induced by $-2KU(t, t')U(t, t'')$, i.e.

$$\Delta V(t, t', t'') = -2K \int_{t_0}^t ds G(t, s) U(t', s) U(t'', s).$$

Hence this justifies the prescription (5.6). Similarly, the perturbation $\Delta G_{\alpha-\beta,\alpha}(t, t')$ of $O(M^{-1/2})$ due to the first right-hand-side term of (A4) is

$$\Delta G_{\alpha-\beta,\alpha}(t, t') = -2KM^{-1/2} \int_{t_0}^t ds \phi_{\alpha-\beta,-\beta,\alpha} G_{\alpha-\beta,\alpha-\beta}(t,s) \psi_\beta^*(s) G_{\alpha,\alpha}(s, t'). \quad (\text{A6})$$

By applying $M^{-3/2} \sum'_\alpha \sum''_{\beta'} \phi_{\alpha,\beta,\alpha-\beta} \psi_\beta(t'')$ to (A6) and averaging, we obtain the perturbation $\Delta F(t, t', t'')$ induced by $-2KU(t, t'')G(t, t')$, i.e.

$$\Delta F(t, t', t'') = -2K \int_{t_0}^t ds G(t, s) U(t'', s) G(s, t').$$

Therefore this justifies the prescription (5.7). A similar argument can be used to justify (5.9)–(5.12).

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