A consequence of the zero fourth cumulant approximation

By EDWARD E. O'BRIEN
State University of New York, Long Island Center, Oyster Bay, New York

AND GEORGE C. FRANCIS
Computing Laboratory, Ballistics Research Laboratory,
Aberdeen Proving Ground, Maryland

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Recent investigations by Kraichnan (1961) and Ogura (1961) have raised doubts concerning the usefulness of the zero fourth cumulant approximation in turbulence dynamics. It appears extremely tedious to examine, by numerical computation, the consequences of this approximation on the turbulent energy spectrum although the appropriate equations have been established by Proudman & Reid (1954) and Tatsumi (1957). It has proved possible, however, to compute numerically the sequences of an analogous assumption when applied to an isotropic passive scalar in isotropic turbulence.

The result of such computation, for specific initial conditions described herein, and for stationary turbulence, is that the scalar spectrum does develop negative values after a time approximately $2\Lambda/(\overline{u^2})^{\frac{1}{2}}$, where Λ is a length scale typical of the energy-containing components of both the turbulent and scalar spectra and $(\overline{u^2})^{\frac{1}{2}}$ is the root mean square turbulent velocity.

1. Introduction

The application of an hypothesis of Millionshtchikov (1941) to the problem of turbulent energy decay was made independently by Proudman & Reid (1954) and Tatsumi (1957). This hypothesis assumes that the relationship among mean values of quadruple velocity component products and those of double velocity component products is the one appropriate to a jointly normal probability distribution. The third-order convariances are not taken as zero, hence complete joint normality is not postulated.

A recent critical appraisal of the general cumulant discard approximation, of which the above hypothesis is a particular case, has been presented by Kraichnan (1961) and throws suspicion on its validity except in the situation of low Reynolds number or short time intervals from an initially multivariate normal distribution of velocity. Ogura (1961) has shown that Millionshtchikov's hypothesis leads, for two-dimensional turbulence, to a negative energy spectrum.

The present investigation first derives the dynamical consequences of the zero fourth cumulant approximation when applied to the behaviour of a scalar field. The field has been chosen to be one which undergoes a first-order reaction as well as turbulent mixing since this slight generalization adds no mathematical complexity to the problem and does provide an extra time-scale parameter. In physical terms this requires a dilute reactant with negligible temperature fluctuations.

The appropriate spectral equations so obtained are solved numerically for a particular set of initial conditions, and it is found that the scalar spectrum becomes negative. This result is obtained from two distinct computing programmes, both of which are described in some detail and it is taken to be secure evidence that the hypothesis investigated here can produce unphysical results when applied to scalar mixing.

2. The general equations of spectral transfer

For the problem of an isotropic concentration fluctuation field randomly convected by and reacting with an isotropic turbulent velocity field, two equations will be derived. The first of these relates the two-point, second-order correlation to a two-point, third-order correlation involving one velocity fluctuation component and two concentration fluctuations. The second equation relates a three-point third-order correlation to a three-point fourth-order one. In order to achieve this, certain simplifying restrictions are employed. Both the field of turbulence and the field of concentration fluctuations are assumed isotropic. The coefficients of viscosity and of diffusion, and the reaction rate constant, are taken independent of position and time. Also it is necessary that the reactant be dynamically passive and that the reaction, if any, be first order. As remarked in the previous section these conditions can best be obtained by a dilute reactant for which temperature fluctuations are negligible.

With these restrictions the material conservation equation for the reactant becomes

$$\frac{\partial \Gamma}{\partial t}+\frac{\partial (u_j\Gamma)}{\partial x_j} = D\frac{\partial^2 \Gamma}{\partial x_k \partial x_k}-C\Gamma, \tag{2.1}$$

where $\Gamma(\mathbf{x},t)$ is the concentration of the reactant, and is a random function of position and time, $u_j(\mathbf{x},t)$ is a turbulent incompressible velocity field assumed to be statistically specified *a priori*, D is the diffusion constant and C is the reaction rate constant.

Taking the mean of (2.1) and subtracting the result from (2.1) we obtain

$$\frac{\partial \gamma}{\partial t}+\frac{\partial (u_j\gamma)}{\partial x_j} = D\frac{\partial^2 \gamma}{\partial x_k \partial x_k}-C\gamma \tag{2.2}$$

for homogeneous turbulence, where $\gamma(\mathbf{x},t)$ is defined as the fluctuation of concentration about the mean at a point $\mathbf{x}$ and a time t. A correlation equation for the concentration at two space points $(\mathbf{x},\mathbf{x}')$ and identical times can be formed in the usual fashion (for example, see Batchelor 1953, p. 79). If we define a separation vector $\mathbf{r} = \mathbf{x}'-\mathbf{x}$ and for notational simplicity write $\gamma(\mathbf{x}',t)$ as γ', the probability average of the time rate of change of $\gamma\gamma'$ becomes

$$\frac{\partial}{\partial t}\overline{\gamma\gamma'}+\frac{\partial}{\partial r_j}(\overline{\gamma\gamma' u_j'-\gamma'\gamma u_j}) = 2D\frac{\partial^2 \overline{\gamma\gamma'}}{\partial r_k \partial r_k}-2C\overline{\gamma\gamma'}. \tag{2.3}$$

$\gamma(\mathbf{x}, t)$ is a stationary random function of $\mathbf{x}$. Therefore it follows from Cramer's theorem (1940) that since $\overline{\gamma\gamma'}(\mathbf{r})$ is the correlation scalar describing a homogeneous field of concentration fluctuations there exists a function $A(\mathbf{k})$ which is its Fourier transform. Furthermore, the assumption of isotropy implies that $A(\mathbf{k})$ can be written as a function of the magnitude of $\mathbf{k}$. We use

$$A(\mathbf{k}) = G(k^2), \tag{2.4}$$

and a Fourier transform of equation (2.3) yields

$$\frac{\partial}{\partial t} G(k^2, t) + 2(Dk^2 + C)\, G(k^2, t) = T(k^2, t), \tag{2.5}$$

where
$$T(k^2, t) = (2\pi)^{-3} \int \frac{\partial}{\partial r_j} \overline{(\gamma\gamma' u_j - \gamma\gamma' u_j')}\, e^{-i\mathbf{k}\cdot\mathbf{r}}\, d\mathbf{r}. \tag{2.6}$$

Corrsin (1951) has established, for isotropic fields, two properties of the triple moments occurring in equation (2.6) which are useful for our purposes:

$$\overline{\gamma\gamma' u_j}(\mathbf{r}) = -\overline{\gamma\gamma' u_j'}(\mathbf{r}), \tag{2.7}$$

and $\overline{\gamma\gamma' u_j}(\mathbf{r})$ behaves as a cubic near $|\mathbf{r}| = 0$.

Application of these two results to (2.6) shows that

$$\int_0^\infty k^2\, T(k^2, t)\, dk = 0. \tag{2.8}$$

Thus the transfer function $T(k^2, t)$ which can be interpreted as concentration spectral transfer from all other wave vectors to $\mathbf{k}$ has the obviously necessary property that its integral over wave-number space is zero. Hence it represents transfer of concentration 'energy' from one part of wave-number space to another without change in the total amount of such energy. It will be shown subsequently that the results obtained by the joint normal distribution hypotheses are quite consistent with this property.

Equation (2.5) is the first of the principal equations to be derived in this section. It is strikingly similar to the analogous correlation equation in isotropic turbulence, but it is easy to show that the similarity in form and in the behaviour of the transfer function is a direct consequence of the linearity of first-order reactions. Higher-order reactions introduce a new non-linear transfer phenomenon which does not have the property represented by (2.8).

The second-moment equation of interest may be obtained by first multiplying (2.2) by $\gamma' u_k''$ and multiplying (2.2) written for $\gamma'(\mathbf{x}', t)$ by $\gamma u_k''$. Add, then use of the incompressibility condition shows that

$$\begin{aligned} u_k'' \frac{\partial \gamma\gamma'}{\partial t} + u_k'' \left(\gamma' \frac{\partial u_j \gamma}{\partial x_j} + \gamma \frac{\partial u_j \gamma'}{\partial x_j}\right) \\ = u_k'' D \left(\gamma' \frac{\partial^2 \gamma}{\partial x_j \partial x_j} + \gamma \frac{\partial^2 \gamma'}{\partial x_j' \partial x_j'}\right) - 2C u_k'' \gamma\gamma'. \end{aligned} \tag{2.9}$$

The double prime refers to quantities at $\mathbf{x}''$, a third space point.

If the Navier–Stokes equations at $\mathbf{x}''$ are multiplied by $\gamma\gamma'$, we have

$$\gamma\gamma' \frac{\partial u_k''}{\partial t} + \gamma\gamma' u_p'' \frac{\partial u_k''}{\partial x_p''} = \nu\gamma\gamma' \frac{\partial^2 u_k''}{\partial x_j'' \partial x_j''} - \frac{1}{\rho}\gamma\gamma' \frac{\partial p''}{\partial x_k''}. \tag{2.10}$$

When equations (2.9) and (2.10) are added and averaged then

$$\frac{\partial}{\partial t}\overline{\gamma\gamma' u_k''}+\overline{u_k''\gamma'\frac{\partial(u_j\gamma)}{\partial x_j}}+\overline{u_k''\gamma\frac{\partial(u_j'\gamma')}{\partial x_j'}}+\overline{\gamma\gamma'\frac{\partial u_p'' u_k''}{\partial x_p''}}$$
$$=\overline{\nu\gamma\gamma'\frac{\partial^2 u_k''}{\partial x_j''\partial x_j''}}+\overline{Du_k''\left(\gamma'\frac{\partial^2\gamma}{\partial x_j\partial x_j}+\gamma\frac{\partial^2\gamma'}{\partial x_j'\partial x_j'}\right)}$$
$$-2C\overline{u_k''\gamma\gamma'}-\frac{1}{\rho}\frac{\partial\overline{\gamma\gamma' p''}}{\partial x_k''}. \tag{2.11}$$

Defining $\mathbf{r}'$ by the equation $\mathbf{x}'' = \mathbf{x}+\mathbf{r}'$ and as before $\mathbf{x}' = \mathbf{x}+\mathbf{r}$ then

$$\left.\begin{aligned}\partial/\partial x_i &= -(\partial/\partial r_i+\partial/\partial r_i'),\\ \partial/\partial x_i' &= \partial/\partial r_i,\\ \partial/\partial x_i'' &= \partial/\partial r_i'.\end{aligned}\right\} \tag{2.12}$$

Use of the incompressibility condition and (2.12) and the partial derivative with respect to r_k of (2.11) leads to the scalar equation

$$\left\{\frac{\partial}{\partial t}-2D\left(\frac{\partial^2}{\partial r_j\partial r_j'}+\frac{\partial^2}{\partial r_j\partial r_j}+\frac{1}{2}\frac{\partial^2}{\partial r_j'\partial r_j'}\right)-\nu\frac{\partial^2}{\partial r_j'\partial r_j'}+2C\right\}\frac{\partial}{\partial r_k}(\overline{\gamma\gamma' u_k''})$$
$$=\frac{\partial^2}{\partial r_j\partial r_k}(\overline{\gamma\gamma' u_k'' u_j})+\frac{\partial^2}{\partial r_j'\partial r_k}(\overline{\gamma\gamma' u_k'' u_j})$$
$$-\frac{\partial^2}{\partial r_j\partial r_k}(\overline{u_k'' u_j'\gamma\gamma'})-\frac{\partial^2}{\partial r_j\partial r_k}(\overline{\gamma\gamma' u_k'' u_\gamma''})$$
$$-\frac{1}{\rho}\frac{\partial^2}{\partial r_k'\partial r_k}(\overline{\gamma\gamma' p''}). \tag{2.13}$$

Equation (2.13) expresses the relation between third- and fourth-order means in its most appropriate form for our purposes. It is a straightforward calculation (O'Brien 1960) to demonstrate that the Fourier transform of $-2\partial(\overline{\gamma\gamma' u_k''})/\partial r_k$ defined by

$$\mathscr{L}(\mathbf{k},\mathbf{k}',t) = -2(2\pi)^{-6}\iint\frac{\partial}{\partial r_k}(\overline{\gamma\gamma' u_k''})\,e^{-i\mathbf{k}.\mathbf{r}-i\mathbf{k}'.\mathbf{r}'}\,d\mathbf{r}\,d\mathbf{r}'$$

is related to $T(k^2,t)$ in (2.5) by

$$T(k^2,t) = \int\mathscr{L}(\mathbf{k},\mathbf{k}',t)\,d\mathbf{k}'. \tag{2.14}$$

The left-hand side of (2.13) has an important symmetry property, demonstrated below, which is independent of the statistical assumption employed in reducing the right-hand side of (2.13) but which is useful in determining valid forms for the initial conditions on $\mathscr{L}(\mathbf{k},\mathbf{k}',t)$ and for showing that use of the zero fourth cumulant approximation is formally consistent with the requirements of equation (2.5). A Fourier transform of the left-hand side of (2.13) (and a multiplication by -2) produces the terms

$$\left\{\frac{\partial}{\partial t}+[2Dkk'\mu+2Dk^2+(\nu+D)\,k'^2+2C]\right\}\mathscr{L}(\mathbf{k},\mathbf{k}',t),$$

where
$$\mu = \frac{\mathbf{k}.\mathbf{k}'}{|\mathbf{k}|\,|\mathbf{k}'|}.$$

The symmetry property mentioned above can best be seen if we adopt Tatsumi's (1957) notation and define a new vector $\mathbf{k}''$ by the relation

$$\mathbf{k}+\mathbf{k}'+\mathbf{k}'' = 0, \tag{2.15}$$

and let
$$\mu' = \frac{\mathbf{k}.\mathbf{k}''}{|\mathbf{k}|\,|\mathbf{k}''|}.$$

Then
$$k'^2 = k^2+k''^2+2\mu'kk'' \quad \text{and} \quad k''^2 = k^2+k'^2+2\mu kk'.$$

Let $\mathscr{L}(\mathbf{k},\mathbf{k}',t)$, which by isotropy can be written $\mathscr{L}_1(k,k',\mu,t)$, be considered as a function of $\mathbf{k}$, $\mathbf{k}''$ and t. We write it as

$$L(\mathbf{k},\mathbf{k}'',t) = L(k,k'',\mu',t) = \mathscr{L}_1(k,k',\mu,t).$$

If we adopt $\mathbf{k}''$ as defined by (2.15) as an independent variable instead of $\mathbf{k}'$, we find that the transformed left-hand side of (2.13) takes the form

$$\frac{\partial L}{\partial t}(k,k'',\mu',t)+f(k,k'',\mu')\,L(k,k'',\mu',t),$$

where
$$f(k,k'',\mu') = D(k^2+k''^2)+\nu k'^2+2C$$

and is therefore symmetric in $\mathbf{k}$ and $\mathbf{k}''$.

The significance of this symmetry can be demonstrated by combining (2.5) and (2.14). That the latter equation can be written

$$T(k^2,t) = \int L(\mathbf{k},\mathbf{k}'',t)\,d\mathbf{k}''$$

is physically evident and easily proved by noting that the Jacobian

$$\frac{\partial(k',\mu)}{\partial(k'',\mu')} = -\frac{k''^2}{k'^2}.$$

Hence from (2.5) and (2.14)

$$\iint L(\mathbf{k},\mathbf{k}'',t)\,d\mathbf{k}\,d\mathbf{k}'' = 0. \tag{2.16}$$

A sufficient condition for this identify to be satisfied is that L be antisymmetric in $\mathbf{k}$ and $\mathbf{k}''$, and a consequence of the symmetry of $f(k,k'',\mu')$ in $\mathbf{k}$ and $\mathbf{k}''$ is that the symmetric and antisymmetric parts of $L(\mathbf{k},\mathbf{k}'',t)$ obey independent equations.

From (2.16) it seems wise to choose $L(\mathbf{k},\mathbf{k}'',0)$ as antisymmetric in $\mathbf{k}$ and $\mathbf{k}''$. The formal consistency of the zero fourth cumulant approximation becomes evident when, as we show in §3, the Fourier transform of the right-hand side of (2.15) is then also antisymmetric in $\mathbf{k}$ and $\mathbf{k}''$. Under these conditions no symmetric terms in $L(\mathbf{k},\mathbf{k}'',t)$ can be generated and (2.16) is identically satisfied for all t.

It is suspected, although not proved, that the complete right-hand side of (2.13) without approximation may also have this antisymmetry property and that therefore $L(\mathbf{k},\mathbf{k}'',t)$ may be quite generally interpreted as representing concentration 'energy' transfer from wave vector $\mathbf{k}$ to wave vector $\mathbf{k}''$.

3. Spectral decay equations

It is possible to obtain a determinate set of decay equations by assuming that all fourth-order covariances are related to second-order ones in the same manner as they are for Gaussian variables. Under this zero fourth cumulant approximation the mean of four fluctuating components a, b, c and d, each with zero means, will satisfy the equation

$$\overline{abcd} = \overline{ab}\,\overline{cd}+\overline{ac}\,\overline{bd}+\overline{ad}\,\overline{bc}. \tag{3.1}$$

If a, b, c and d were true Gaussian variables any odd-order mean would vanish. Evidently to require this would eliminate completely the inertial transfer effects from equation (2.5) and thus destroy the very property which is central to the problem.

Use of (3.1) for fourth-order moments consisting of two velocities and two concentrations leads to reductions of all the terms on the right-hand side of (2.13). For example

$$\overline{u_k'' u_j \gamma\gamma'} = \overline{u_k'' u_j}\;\overline{\gamma\gamma'}+\overline{u_k''\gamma}\;\overline{u_j\gamma'}+\overline{u_k''\gamma'}\;\overline{u_j\gamma},$$

but all first-order isotropic solenoidal tensors (such as $\overline{u_j\gamma'}$, etc.) are zero, so that we have simply

$$\overline{u_k'' u_i \gamma\gamma'} = \overline{u_k'' u_j}(\mathbf{r}')\,\overline{\gamma\gamma'}(\mathbf{r}),$$

and Q_{kj}, the Fourier transform of $\overline{\gamma\gamma' u_k'' u_j}$, becomes

$$Q_{kj} = \phi_{jk}(\mathbf{k}')\,G(k^2), \tag{3.2}$$

where $\phi_{jk}(\mathbf{k}')$ is the Fourier transform of $\overline{u_j u_k''}(\mathbf{r}')$. Similarly, by redefining variables

$$\mathbf{r} = \boldsymbol{\xi};\quad \mathbf{r}'-\mathbf{r} = \eta$$

and taking the Fourier transform of $\overline{u_k'' u_j' \gamma\gamma'}$ it can be shown (O'Brien 1960) that, if Q_{kj}' is this Fourier transform,

$$Q_{kj}' = \phi_{jk}(\mathbf{k}')\,G(k''^2). \tag{3.3}$$

The remaining term on the right-hand side of (2.13) can be shown to be identically zero under this hypothesis by the following reasoning:

$$\overline{u_k'' u_j'' \gamma\gamma'}(\mathbf{r},\mathbf{r}') = \overline{u_k'' u_j''}(0)\,\overline{\gamma\gamma'}(\mathbf{r}),$$

which is independent of $\mathbf{r}'$ and so

$$\frac{\partial}{\partial r_j'}(\overline{u_k'' u_j'' \gamma\gamma'(\mathbf{r})}) = 0.$$

Furthermore,
$$\frac{\partial}{\partial r_j'}(\overline{u_k'' u_j \gamma\gamma'}) = 0,$$

by incompressibility when $\overline{u_k'' u_j \gamma\gamma'} = \overline{u_k'' u_j}\,\overline{\gamma\gamma'}$.

Finally we note a result of Reid (1955) who demonstrated that the triple correlation $\overline{\gamma\gamma' p''}$ is identically zero under the hypothesis we are investigating.

The quantity $\phi_{jk}(\mathbf{k}')$ which occurs in (3.2) and (3.3) and is defined following (3.2) can, for isotropic fields using invariance theory (Robertson 1940) and the incompressibility condition $k_j'\phi_{jk}(\mathbf{k}') = 0$, be written in the form

$$\phi_{jk}(\mathbf{k}') = \phi(k'^2)\left[\delta_{jk}-\frac{k_j' k_k'}{k'^2}\right], \tag{3.4}$$

where $\phi(k'^2)$ is an arbitrary scalar and will be given its usual title of turbulent energy spectrum density.

Equations (3.2), (3.3) and (3.4) can be used to write the transform of equation (2.13) in its most concise form as

$$\left\{\frac{\partial}{\partial t}+f(k,k'',\mu')\right\}L(k,k'',\mu',t) = 2k^2(1-\mu^2)\,\phi(k'^2)\,[G(k''^2,t)-G(k^2,t)], \quad (3.5)$$

where $$f(k,k'',\mu') = Dk^2+Dk''^2+\nu k'^2+2C$$

was defined in the preceding section.

For convenience we rewrite equations (2.5) and (2.14) of §2 since they, in conjunction with (3.5), form the closed set of moment equations we desired to establish:

$$\frac{\partial G}{\partial t}(k^2,t)+[2Dk^2+2C]\,G(k^2,t) = T(k^2,t), \quad (2.5)$$

$$2\pi\int_0^\infty\int_{-1}^{+1} L(k,k'',\mu',t)\,k''^2\,dk''\,d\mu' = T(k^2,t). \quad (2.14)$$

Structurally equations (2.5) and (3.5) are quite similar and their solutions can be written immediately in the following manner:

$$L(k,k'',\mu',t) = L(k,k'',\mu',0)\exp\{-f(k,k'',\mu')\,t\}$$
$$+\,2k^2(1-\mu^2)\int_0^t \exp\{-f[t-t']\}\,\phi(k'^2,t)\,[G(k''^2,t')-G(k^2,t')]\,dt', \quad (3.6)$$

$$G(k^2,t) = G(k^2,0)\exp\{-2[Dk^2+C]\,t\}$$
$$+\int_0^t T(k^2,t')\exp\{-2[Dk^2+C]\,[t-t']\}\,dt'. \quad (3.7$$

The simultaneous equations (3.6) and (3.7) together with (2.14) constitute the fundamental equations for the present study. In the next section certain definite conditions on $G(k^2,t)$ and $\phi(k'^2,t)$ are specified and numerical calculations are presented for the decay of a particular concentration spectrum.

4. Numerical computation of the decay of a typical spectrum

It is quite possible to rewrite equations (2.5) and (3.5) in dimensionless form. However, unless the length and velocity scales are chosen to be time independent we can no longer present the solutions in the form (3.6) and (3.7). Since we will be concerned only with stationary turbulence the root-mean-square velocity $(\overline{u^2})^{\frac{1}{2}}$ used to non-dimensionalize the velocity fluctuation will be truly time independent. The same, unfortunately, is not true of the length scale which should alter as the concentration spectrum decays in time. We therefore have been forced to choose as the length scale one which is appropriate to the initial concentration spectrum. We have in fact chosen to use the inverse of the wave-number at which the initial concentration spectrum peaks. It can easily be shown that for the particular spectrum which we have examined numerically this is of the same order as the concentration microscale.

If we denote this length scale by Λ the following dimensionless numbers arise:

$$N_R = \frac{(\overline{u^2})^{\frac{1}{2}}\,\Lambda}{\nu}, \quad \text{a Reynolds number,}$$

$$N_P = \frac{(\overline{u^2})^{\frac{1}{2}}\,\Lambda}{D}, \quad \text{a Peclet number,}$$

$$N_D = \frac{\Lambda C}{(\overline{u^2})^{\frac{1}{2}}}, \quad \text{a Damkohler number of the first kind.}$$

The use of Λ in the Reynolds number will not be misleading since for the spectra we examine Λ is of the same order as the turbulence microscale.

Equations (3.6), (3.7) and (2.14) will yield information about the time and wave-number behaviour of a concentration spectrum density $G(k^2, t)$ only when two initial conditions $G(k^2, 0)$ and $L(k, k'', \mu, 0)$ and the turbulence spectrum density $\phi(k'^2, t)$ are specified. One restriction that $L(k, k'', \mu', 0)$ be antisymmetric in k and k'' has already been noted. It would evidently be useful to examine a wide range of initial conditions to explore the degree of dependence of the solution on the initial statistical distribution. We will, however, consider only the most simple situation, that in which initially the statistical distribution third-order moments are zero and hence $T(k^2, 0)$ and $L(k, k'', \mu', 0)$ are zero. Such a situation occurs for example if one assumes an initially multivariate normal distribution, and concentration fields are initially uncorrelated.

One spectrum for which $T(k^2, 0)$ might be expected to be zero is the one typical of the final period of decay (Proudman & Reid 1954) when there is, in fact, negligible energy transfer. Its precise definition will be described subsequently. It should be emphasized that use of such a spectrum does not limit us to the final period but merely provides us with a consistent and well-behaved initial condition.

With these ideas in mind the spectrum whose decay we have computed has the following description:

$$\left.\begin{aligned} G(k^2, 0) &= B_1 e^{-k^2}, \\ \phi(k'^2, 0) &= \phi(k'^2, t) = (3\pi^{\frac{3}{2}})^{-1} k'^2 e^{-k'^2}, \\ \mathscr{L}_1(k, k', \mu, 0) &= 0, \end{aligned}\right\} \tag{4.1}$$

where the independent variables are now all dimensionless.

When the above information is inserted into the non-dimensional forms of (3.7), (3.6) and (2.14) we obtain

$$\begin{aligned} G(k^2, t) = G(k^2, 0) \exp\left[-2\left\{\frac{k^2}{N_P} + N_D\right\} t\right] \\ + \int_0^t \exp\left[-2\left\{\frac{k^2}{N_P} + N_D\right\}(t - t')\right] T(k^2, t')\, dt', \end{aligned} \tag{4.2}$$

$$\begin{aligned} \mathscr{L}_1(k, k', \mu, t) &= \frac{2k^2}{3\pi^{\frac{3}{2}}} k'^2 e^{-k'^2}(1 - \mu^2) \\ &\times \int_0^t \exp\left[-\left\{\frac{2kk'\mu}{N_P} + \frac{2k^2}{N_P} + \left(\frac{1}{N_P} + \frac{1}{N_R}\right) k'^2 + 2N_D\right\}(t - t')\right] [G(k''^2, t') - G(k^2, t')]\, dt', \end{aligned} \tag{4.3}$$

$$T(k^2, t) = 2\pi \int_0^\infty \int_{-1}^{+1} \mathscr{L}_1(k, k', \mu, t)\, k'^2\, dk'\, d\mu, \tag{4.4}$$

where $G(k^2, 0) = B_1 e^{-k^2}$ and, from previous remarks on the scaling, the non-dimensional time t is actual time multiplied by $(\overline{u^2})^{\frac{1}{2}}/\Lambda$.

The above equations are linear in B_1, hence in the graphical presentation of results it is pertinent to leave the ordinate arbitrary. The use of k, k', μ and t as independent variables rather than k, k'', μ' and t is for convenience only.

From (4.2) it follows that

$$G(k^2, t+\tau) = G(k^2, t)\exp\left[-2\left(\frac{k^2}{N_P}+N_D\right)\tau\right] + \int_t^{t+\tau}\exp\left[-2\left(\frac{k^2}{N_P}+N_D\right)(t+\tau-t')\right]T(k^2, t')\,dt'.$$

Hence

$$G(k^2, t+\tau) \approx G(k^2, t)\exp\left[-2\left(\frac{k^2}{N_P}+N_D\right)\tau\right] + \tfrac{1}{2}\tau\left[\exp\left\{-2\left(\frac{k^2}{N_P}+N_D\right)\tau\right\}T(k^2, t)+T(k^2, t+\tau)\right]$$

by the trapezoidal rule (error of order τ^2). Similarly, letting

$$y(k, k', \mu) = \frac{2kk'\mu}{N_P}+\frac{2k^2}{N_P}+\left(\frac{1}{N_P}+\frac{1}{N_R}\right)k'^2+2N_D,$$

we obtain, from (4.3),

$$\mathscr{L}_1(t+\tau) = e^{-y\tau}L(t)+\tfrac{2}{3}\pi^{-\frac{3}{2}}k^2k'^2e^{-k'^2}(1-\mu^2)\,\tau[e^{-y\tau}\{G(k''^2, t)-G(k^2, t)\}],$$

where the less precise rectangular rule rather than the trapezoidal is used because $G(k^2, t+\tau)$ is not known at this stage. From $\mathscr{L}_1(t+\tau)$ a double summation leads to $T(k^2, t+\tau)$, which is then used to calculate $G(k^2, t+\tau)$:

$$T(k^2, t+\tau) \approx \sum_{0<k'<M}[2\pi k'^2 W_{k'} \sum_{-1<\mu<1}\{\mathscr{L}_1(k, k', \mu, t+\tau)\,W_\mu\}],$$

where $W_{k'}$ and W_μ are weights related to the points chosen. The particular summations used were those of Gauss quadrature (Gowan, Davids & Sevenson 1942) which gives high accuracy for relatively few summands. (In the case considered, 16 values of $k' < 12$ and 9 values of μ were sufficient.)

The final approximation made was related to $G(k''^2, t)$. Such values were obtained by quadratic interpolation in the table of $G(k^2, t)$, with any values outside the table, or any negative values obtained through inaccuracies, being taken as zero. As a check on the accuracy of the calculations we have used the condition (2.8) of conservation of 'energy' in convective transfer,

$$\int_0^\infty k^2T(k^2, t)\,dk = 0.$$

Since $k^2T < 0$ for small k, say $k < k_1$, and $k^2T > 0$ for larger k, $\left|\int_0^{k_1} k^2T\,dk\right|$ should equal $\left|\int_{k_1}^\infty k^2T\,dk\right|$. At every time-step these integrals were evaluated and compared. At no time did they differ by more than 4 % using a combination of trapezoidal and Simpson's rules.

It had been hoped that the above programme could have been carried out for enough different values of N_P, N_R and N_D to include most physically interesting systems. Initially N_R was chosen as 50, N_P as 3500 (a Schmidt number of 700), and N_D as 0·04 to give a fairly strong reactive effect. The number of values of k, k' and μ, nearly a maximum compatible with the memory space available on the computer, were 30 for k, 16 for k', and 9 for μ.

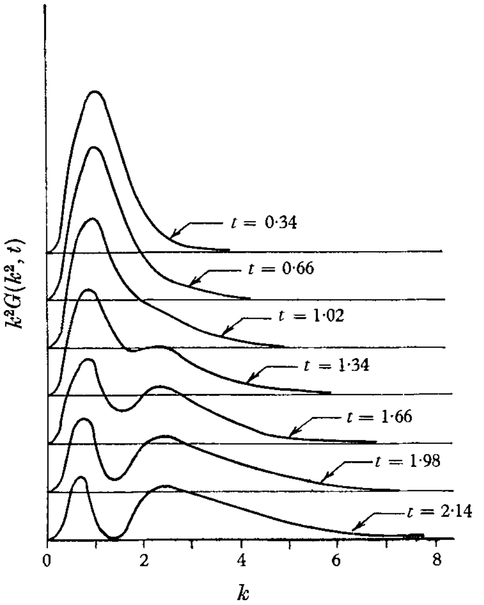

FIGURE 1. Three-dimensional concentration 'energy' spectrum $k^2G(k^2, t)$. Ordinate scale arbitrary. $N_R = 50$, $N_P = 3500$, $N_D = 0{\cdot}04$.

Figure 1 shows the behaviour of the concentration spectrum $k^2G(k^2,t)$ and figure 2 presents the transfer function $k^2T(k^2,t)$ as decay proceeds. It is evident that an unexpected dip occurs in the scalar spectrum near $k = 1{\cdot}4$ which is also approximately the dimensionless wave-number corresponding to the peak of the turbulence energy spectrum. It is very convenient for calculation purposes to maintain, as we have done here, the turbulent energy spectrum peak and scalar spectrum peak at the same order of wave-number, but without examining other arrangements where the two spectra are widely separated it is impossible to determine in general how intimately the turbulent spectrum and the phenomenon of a dip formation are related. This has not been done. The occurrence of negative values of the scalar spectrum (at $t = 2{\cdot}14$) led us to decide to verify that this was a real effect by examining the same spectrum's decay using a second programme quite distinct from the first.

An alternative calculation of the same spectral decay problem as above, (4.1), can be made by carrying out the integration over μ analytically before devising a computing programme. When this is done it becomes evident that for the spectrum under consideration there is a considerable time duration during which the diffusive terms (and reaction term) have negligible influence. Since the non-diffusive case can be examined with considerably improved accuracy a computation of this has been carried out.

If (3.5) is inserted into (2.5), the initial condition $L(k, k'', \mu', 0) = 0$ is used and $\phi(k'^2, t)$ is taken independent of time, we have

$$\frac{\partial G}{\partial t}(k^2,t)+2[Dk^2+C]\,G(k^2,t) = \int_{\mathbf{k}''}\int_0^t 2k^2\exp\{-f(k,k'',\mu')\,[t-t']\}\,(1-\mu^2)\,\phi(k'^2) \times [G(k''^2,t')-G(k^2,t')]\,dt'\,d\mathbf{k}''. \quad (4.5)$$

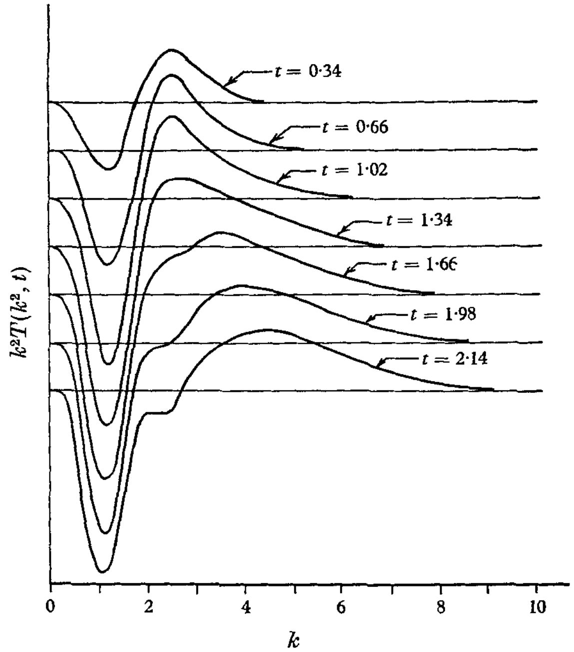

FIGURE 2. Three-dimensional concentration spectral transfer function $k^2T(k^2, t)$. Ordinate scale arbitrary. $N_R = 50$, $N_P = 3500$, $N_D = 0{\cdot}04$.

The further assumption that diffusive terms be neglected reduced (4.5) to

$$\frac{\partial G(k^2,t)}{\partial t} = \int_{\mathbf{k}''} 2k^2(1-\mu^2)\;\phi(k'^2)\left\{\int_0^t [G(k''^2,t')-G(k^2,t')]\,dt'\right\}d\mathbf{k}''. \quad (4.6)$$

If a function $F(k^2,t)$ is defined by $F(k^2,t) = \int_0^t G(k^2,t')\,dt'$ and if the sine rule is applied to the vector triad $\mathbf{k}+\mathbf{k}'+\mathbf{k}'' = 0$ to yield $k'^2(1-\mu^2) = k''^2(1-\mu'^2)$ then (4.6) becomes

$$\frac{\partial^2 F(k^2,t)}{\partial t^2} + g(\mathbf{k})\,F(k^2,t) = \int_{\mathbf{k}''} K(\mathbf{k},\mathbf{k}'')\,F(k''^2,t)\,d\mathbf{k}'', \quad (4.7)$$

where $K(\mathbf{k},\mathbf{k}'') = [2k^2k''^2(1-\mu'^2)\,\phi(k'^2)]/k'^2$ is symmetric in $\mathbf{k}$ and $\mathbf{k}''$ and not less than 0 for all $\mathbf{k}$ and $\mathbf{k}''$ and

$$g(\mathbf{k}) = \int_{\mathbf{k}''} K(\mathbf{k},\mathbf{k}'')\,d\mathbf{k}''.$$

From the form of $\phi(k'^2)$ as given by (4.1), $K(\mathbf{k}, \mathbf{k}')$ and $g(\mathbf{k})$ can be calculated. When this is done and (4.7) is re-expressed in scalar form it becomes

$$\frac{\partial^2 F(k^2,t)}{\partial t^2} = -2\pi^{\frac{3}{2}}k^2 F(k^2,t) + \pi k^{-2}\int_0^\infty kk''\exp\{-(k^2+k''^2)\}F(k''^2,t) \times [(2kk''+1)\exp(-2kk'') + (2kk''-1)\exp(2kk'')]\,dk'', \quad (4.8)$$

with the initial conditions

$$F(k^2,0) = 0,$$

$$\frac{\partial F}{\partial t}(k^2,0) = G(k^2,0) = B_1 e^{-k^2}.$$

With k held fixed and the integral replaced by an approximating sum, the differential equation becomes an ordinary one of second order, or a pair of first order:

$$\frac{dF}{dt}(k^2,t) = G(k^2,t),$$

$$\frac{dG}{dt}(k^2,t) = -2\pi^{\frac{3}{2}}k^2F(k^2,t) + I(k^2,t),$$

where $I(k^2,t) = 0$ if $k = 0$ and, otherwise,

$$I(k^2,t) = \frac{\pi}{k^2}\sum_{0\leqslant k''\leqslant M} W_{k''}kk''F(k''^2,t) \times [(2kk''+1)\exp\{-(k+k'')^2\} + (2kk''-1)\exp\{+(k-k'')^2\}],$$

where $W_{k''}$ is the weight associated with point k'', and M is an upper bound to the region of significance (an 'approximation' to ∞).

Selecting the k''-values from among the k-values eliminates the inaccuracy of interpolating for $F(k''^2,t)$. (Such specification was not possible in the more general problem discussed earlier.)

The differential equations were integrated by the Runge–Kutta–Gill method (Gill 1951), a standard procedure for high-speed electronic computers, not convenient for hand calculations.

The condition of conservation of 'energy' in convective transfer becomes in the new formulation

$$\int_0^\infty 2\pi^{\frac{3}{2}}k^4F(k^2,t)\,dk = \int_0^\infty k^2I(k^2,t)\,dk.$$

Again this condition is used as a continuous check on the accuracy of the calculations. Both integrals were evaluated (by Simpson's rule) at every t, and the procedure was stopped when they differed by as much as 5 %.

For the specific case considered in this paper, 19 values of k and 25 values of t'', $0 \leqslant k'' \leqslant 11{\cdot}8$, were used. The summation over k'' to obtain $I(k^2,t)$ used Simpson's rule also.

Figures 3 and 4 present the behaviour of the concentration spectrum $k^2G(k^2,t)$ and the transfer function $k^2T(k^2,t)$ respectively. The very close similarity between figures 1, 2 and 3, 4 show that diffusive terms are indeed negligible for a considerable time as we surmised and, more significantly, that the joint normal distribution hypothesis can lead to a negative spectrum.

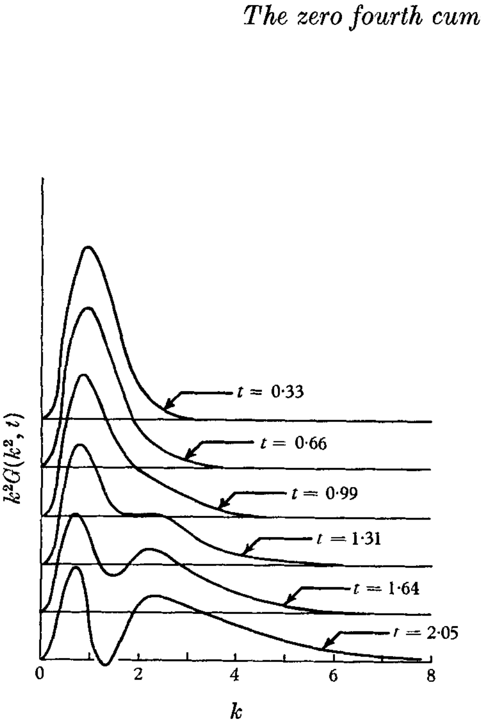

FIGURE 3. Three-dimensional concentration 'energy' spectrum $k^2G(k^2, t)$. Ordinate scale arbitrary. $N_R = \infty$, $N_P = \infty$, $N_D = 0$.

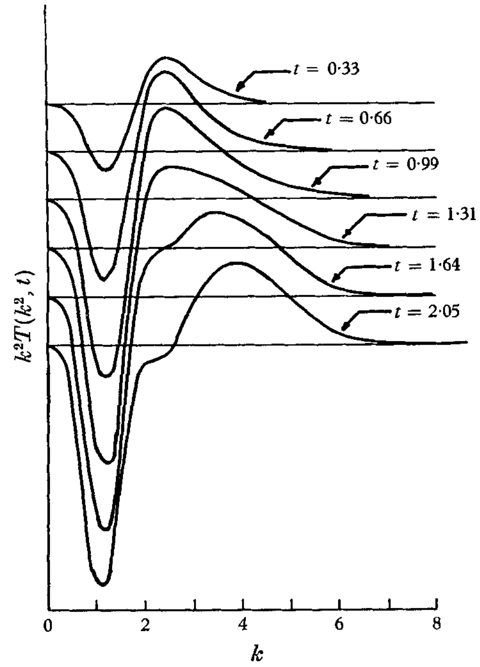

FIGURE 4. Three-dimensional concentration spectral transfer function $k^2T(k^2, t)$. Ordinate scale arbitrary. $N_R = \infty$, $N_P = \infty$, $N_D = 0$.

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REFERENCES

BATCHELOR, G. K. 1953 *The Theory of Homogeneous Turbulence*. Cambridge University Press.

CORRSIN, S. 1951 *J. Aero. Sci.* **18** (6), 417.

CRAMER, H. 1940 *Ann. Math.* **41**, 215.

GILL, S. 1951 *Proc. Camb. Phil. Soc.* **47** (1), 96.

GOWAN, A. N., DAVIDS, N. & SEVENSON, A. 1942 *Bull. Amer. Math. Soc.* **48**, 10, 739.

KRAICHNAN, R. 1961 *N.Y.U. Res. Rep. HSN*-3.

Millionshtchikov, M. 1941 *C.R. Acad. Sci., U.S.S.R.*, **32**, 615, 619.

O'Brien, E. E. 1960 Doctoral Disseration, Department of Mechanics, The Johns Hopkins University.

Ogura, Y. 1961 *Tech. Rep. Dynamical Weather Prediction Project M.I.T.*

Proudman, I. & Reid, W. H. 1954 *Phil. Trans.* A, **247**, 163.

Reid, W. H. 1955 *Proc. Camb. Phil. Soc.* **51**, 350.

Robertson, H. P. 1940 *Proc. Camb. Phil. Soc.* **36**, 209.

Tatsumi, T. 1957 *Proc. Roy. Soc.* A, **239**, 16.