J. Fluid Mech. (2000), *vol.* 412, *pp.* 303–329. Printed in the United Kingdom

© 2000 Cambridge University Press

On constructing realizable, conservative mixed scalar equations using the eddy-damped quasi-normal Markovian theory

By MARK ULITSKY[1] AND LANCE R. COLLINS[2]†

[1]Group T-3, MS B216, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

[2]Department of Chemical Engineering, The Pennsylvania State University, University Park, PA 16802, USA

(Received 20 September 1999 and in revised form 14 January 2000)

The eddy-damped quasi-normal Markovian (EDQNM) turbulence theory has been applied to the covariance spectrum of two passive isotropic scalars with different diffusivities in stationary isotropic turbulence. A rigorous application of EDQNM, which introduces no new modelling assumptions or constants, is shown to yield a covariance spectrum that violates the Cauchy–Schwartz inequality over some of the wavenumbers. One approach to this problem is to derive a model based on a stochastic differential equation, as its presence guarantees realizability. For an isotropic scalar, it is possible to construct a Langevin equation for the Fourier transform of the scalar concentrations that is consistent with each EDQNM scalar autocorrelation spectrum. The Langevin equations can then be used to construct a model for the covariance spectrum that is realizable. However, the resulting covariance transfer term does not properly conserve the scalar covariance, and so the model is still not satisfactory. The problem can be traced to the Markovianization step, which leads to the presence of the scalar diffusivities in the transfer functions in an unphysical fashion. A simple fix is described which reconciles the two approaches and gives conservative, realizable results for all time.

Next, we apply the EDQNM theory to a more general system involving the mixing of anisotropic scalars. Anisotropy in this case results from a uniform mean gradient of the two scalar concentrations in one direction. As with the isotropic scalars, direct application of the EDQNM closure results in a covariance spectrum that violates the Cauchy–Schwartz inequality; however, in this case it is not as simple to construct a Langevin model that reproduces all of the spectral interactions that result from the EDQNM procedure. Nevertheless, we show that the same modification of the inverse time scale as is done for the isotropic scalar results in an anisotropic scalar covariance spectrum that is realizable for all times.

1. Introduction

There are several important applications which involve the mixing of either inert or reacting species in a turbulent flow. The most celebrated example is combustion, in which fuel and oxidizer mix and react to produce combustion products and a great deal of energy. The rate of reaction in turbulent flames is often sufficiently large that the overall system is mixing limited, underscoring the need for reliable descriptions

† Author to whom correspondence should be addressed.

of turbulent mixing. In non-premixed combustion, the interaction of several chemical species with different molecular diffusivities leads to the phenomenon known as differential diffusion. This complex phenomenon has been observed experimentally in both non-reacting (Kerstein *et al.* 1989) and reacting (Drake, Pitz & Lapp 1986; Chen, Bilger & Dibble 1990; Vranos *et al.* 1992) flows. Most theoretical models of turbulent diffusion flames make the simplifying assumption that all diffusivities (i.e. that of each species and temperature) are identical (often referred to in the literature as the unity Lewis number assumption). This simplification is extremely attractive, since it ultimately leads to a Shvab–Zeldovich conserved scalar formulation (Williams 1985), regardless of the number of reacting species involved. While the unity Lewis number assumption may be computationally efficient, it is suspect at low to moderate Reynolds numbers where significant effects due to differential diffusion have been observed (Bilger & Dibble 1982).

Recent numerical studies by Yeung & Pope (1993) and later by Yeung (1996) have demonstrated that differential diffusion, being of molecular origin, occurs initially at high wavenumbers and then progresses to lower wavenumbers with time. These results suggest that a model based on a spectral or Fourier representation of the velocity and scalar fields can provide valuable information about this process. Moreover, a spectral description of mixing is attractive because the linear diffusion terms are exact, and thus effects such as differential diffusion should be well captured.

In this study, we consider the interaction of two passive scalars (with different diffusivities) using the eddy-damped quasi-normal Markovian (EDQNM) theory. The EDQNM theory has previously been shown to be an effective tool for investigating both turbulent energy (Andre & Lesieur 1977; Lesieur 1987) and passive scalar spectra (Herring *et al.* 1982; Nakauchi, Oshima & Saito 1989). Originally, we intended to extend the axisymmetric spectral model of Herr, Wang & Collins (1996) for a single scalar to the more general case of multiple scalars with different diffusivities. However, despite the fact that the present model introduces no additional assumptions or new constants, the EDQNM scalar covariance spectrum is shown to violate a Cauchy–Schwartz condition over a range of wavenumbers.

To assist in understanding the origin of the unrealizable spectra, a parallel study of two forced *isotropic* scalars is presented. One advantage of the simpler isotropic system is that it is possible to develop an alternative model based on a Langevin equation. As noted and exploited in much of Kraichnan's earlier work (an excellent review of this work can be found in Kraichnan 1991), models developed from stochastic differential equations are guaranteed to be realizable. Indeed, the Langevin-based model appears to solve the problem; however, closer scrutiny shows that the model for the covariance transfer spectrum derived via the Langevin equation does not properly conserve the scalar covariance, and thus is unphysical as well. Comparing the EDQNM and Langevin-based closures shows that they can be made consistent only by slightly altering the inverse time scale that results from Marovianization. It should be noted that the inverse time scales from all three scalar spectra must be modified. That is, it is not sufficient to simply change the inverse time scale for the scalar covariance spectrum. Given this modification, the model is both conservative and realizable for all time. Also, for a Schmidt number of unity, the autocorrelation spectra will be equivalent to the standard EDQNM model for these spectra.

In principle, a similar approach can be taken to correct the more general axisymmetric scalar spectrum that arises in the presence of uniform mean scalar gradients; unfortunately, there appears to be no equivalent Langevin model for the anisotropic scalar spectrum that reproduces all of the spectral interactions from the EDQNM theory in a consistent fashion. However, if we substitute the modified inverse time scale derived earlier for the isotropic scalar, the resulting EDQNM model yields a realizable covariance spectrum for all times and for all combinations of the scalar diffusivities we considered. A relatively simple *a posteriori* argument is presented that explains why the modification to the inverse time scale yields realizable spectra for this case.

The paper is organized as follows. We begin with a brief summary of the equations for the isotropic scalar spectrum in § 2. Section 3 then gives the results of the EDQNM model, including evidence of unrealizable spectra. A modified model is then developed from the Langevin equation analysis, and the model is shown to be realizable; however, the transfer spectrum from the Langevin analysis does not properly conserve the covariance spectrum until the inverse time scales from the autocorrelation spectra are modified. Results from the original EDQNM, Langevin, and modified Langevin model are given. We then derive the EDQNM model for the covariance spectrum of scalars with uniform mean gradients in § 4 and show example calculations of this model in § 5. Conclusions are given in § 6.

2. Isotropic scalar equations

The equation governing the advection and diffusion of a passive scalar with constant physical properties in an incompressible flow field is given by

$$\frac{\partial \phi_\alpha}{\partial t} + \frac{\partial}{\partial x_i}(u_i \phi_\alpha) = \mathscr{D}_\alpha \frac{\partial^2 \phi_\alpha}{\partial x_i \partial x_i}, \tag{2.1}$$

where ϕ_α is the local concentration of species α, u_i is the Navier–Stokes velocity, and $\mathscr{D}_\alpha$ denotes the molecular diffusivity. As the turbulence and scalar fields are isotropic, we can assume without loss of generality that there is zero mean flow and zero mean scalar (Lesieur 1987; Hinze 1987). Thus, after a Reynolds decomposition, we can simply replace the velocity and scalar by the fluctuating quantities, i.e. $u_i = u_i'$ and $\phi_\alpha = \phi_\alpha'$.

It is convenient to express the equations for the scalar fluctuations (ϕ_α' and ϕ_β') in non-dimensional form using the integral length scale of the turbulence L (for x_i), the r.m.s. fluctuating velocity u' (for u_i), the large-eddy turnover time L/u' (for t), and the characteristic scalar fluctuation $(\epsilon_\phi L/u')^{1/2}$ (for ϕ_α' and ϕ_β' respectively), where ϵ_ϕ is the steady-state scalar dissipation rate. The dimensionless equations are

$$\frac{\partial \phi_\alpha'}{\partial t} + \frac{\partial}{\partial x_i}(u_i' \phi_\alpha') = \frac{1}{Pe_\alpha} \frac{\partial^2 \phi_\alpha'}{\partial x_i \partial x_i}, \tag{2.2}$$

$$\frac{\partial \phi_\beta'}{\partial t} + \frac{\partial}{\partial x_i}(u_i' \phi_\beta') = \frac{1}{Pe_\beta} \frac{\partial^2 \phi_\beta'}{\partial x_i \partial x_i}, \tag{2.3}$$

where Pe_α, the mass transfer Péclet number, is defined in terms of the Reynolds and Schmidt numbers ($Re_L = u'L/\nu$ and $Sc_\alpha = \nu/\mathscr{D}_\alpha$) as $Pe_\alpha = Re_L Sc_\alpha$. The definitions of Pe_β and Sc_β follow by analogy. Also note that in order to maintain a reasonable nomenclature, the same variables (e.g. x_i, t, u_i', and ϕ') have been used to represent dimensionless and dimensional quantities. Since all subsequent equations will be expressed in dimensionless form, except where noted, this practice should not cause any confusion. Equations (2.2) and (2.3) represent the starting point for deriving the EDQNM transport equations for the scalar autocorrelation and covariance spectra.

2.1. *Two-point correlations*

The ultimate aim of the EDQNM analysis is to derive closed expressions for the scalar autocorrelation and covariance spectra. These three scalar spectra will involve the Fourier transform of the following two-point physical space correlations:

$$R_{ij}(\boldsymbol{x}_1,\boldsymbol{x}_2) \equiv \overline{u_i'(\boldsymbol{x}_1)u_j'(\boldsymbol{x}_2)}, \tag{2.4}$$

$$B^{\alpha}(\boldsymbol{x}_1,\boldsymbol{x}_2) \equiv \overline{\phi_\alpha'(\boldsymbol{x}_1)\phi_\alpha'(\boldsymbol{x}_2)}, \tag{2.5}$$

$$B^{\beta}(\boldsymbol{x}_1,\boldsymbol{x}_2) \equiv \overline{\phi_\beta'(\boldsymbol{x}_1)\phi_\beta'(\boldsymbol{x}_2)}, \tag{2.6}$$

$$B^{\alpha\beta}(\boldsymbol{x}_1,\boldsymbol{x}_2) \equiv \overline{\phi_\alpha'(\boldsymbol{x}_1)\phi_\beta'(\boldsymbol{x}_2)}. \tag{2.7}$$

For an arbitrary two-point correlation $A(\boldsymbol{x}_1,\boldsymbol{x}_2)$, the definition of the Fourier transform is given by

$$A(\boldsymbol{k},\boldsymbol{p}) \equiv \iint A(\boldsymbol{x}_1,\boldsymbol{x}_2)\mathrm{e}^{-\mathrm{i}(\boldsymbol{k}\cdot\boldsymbol{x}_1+\boldsymbol{p}\cdot\boldsymbol{x}_2)}\,\mathrm{d}\boldsymbol{x}_1\,\mathrm{d}\boldsymbol{x}_2, \tag{2.8}$$

and its inverse,

$$A(\boldsymbol{x}_1,\boldsymbol{x}_2) \equiv \iint A(\boldsymbol{k},\boldsymbol{p})\mathrm{e}^{+\mathrm{i}(\boldsymbol{k}\cdot\boldsymbol{x}_1+\boldsymbol{p}\cdot\boldsymbol{x}_2)}\,\hat{\mathrm{d}}\boldsymbol{k}\,\hat{\mathrm{d}}\boldsymbol{p}, \tag{2.9}$$

in which $\hat{\mathrm{d}}\boldsymbol{k} \equiv \mathrm{d}\boldsymbol{k}/(2\pi)^3$ and $\hat{\mathrm{d}}\boldsymbol{p} \equiv \mathrm{d}\boldsymbol{p}/(2\pi)^3$.

For isotropic mirror-symmetric turbulence, the Reynolds stress tensor, $R_{ij}(\boldsymbol{k},\boldsymbol{p})$, takes on a particularly simple form in Fourier space which has been discussed extensively in the literature (Andre & Lesieur 1977; Orszag 1970; Leslie 1973; Tatsumi 1980; Lesieur 1987). Likewise, the scalar autocorrelation spectra, $B^{\alpha}(\boldsymbol{k},\boldsymbol{p})$ and $B^{\beta}(\boldsymbol{k},\boldsymbol{p})$, have been discussed extensively in earlier work (Herring *et al.* 1982; Nakauchi *et al.* 1989; Lesieur 1987). Therefore, we state without proof the following results which have been derived by appealing to tensor invariant arguments, isotropy, continuity, and the reality condition (Batchelor 1953):

$$R_{ij}(\boldsymbol{k},\boldsymbol{p}) = \hat{\delta}(\boldsymbol{k}+\boldsymbol{p})P_{ij}(\boldsymbol{k})R(k), \tag{2.10}$$

$$B^{\alpha}(\boldsymbol{k},\boldsymbol{p}) = 2\hat{\delta}(\boldsymbol{k}+\boldsymbol{p})B^{\alpha}(k), \tag{2.11}$$

$$B^{\beta}(\boldsymbol{k},\boldsymbol{p}) = 2\hat{\delta}(\boldsymbol{k}+\boldsymbol{p})B^{\beta}(k), \tag{2.12}$$

$$B^{\alpha\beta}(\boldsymbol{k},\boldsymbol{p}) = 2\hat{\delta}(\boldsymbol{k}+\boldsymbol{p})B^{\alpha\beta}(k), \tag{2.13}$$

where

$$P_{ij}(\boldsymbol{k}) \equiv \delta_{ij} - \frac{k_ik_j}{k^2}, \tag{2.14}$$

$R(k)$, $B^{\alpha}(k)$, $B^{\beta}(k)$, $B^{\alpha\beta}(k)$ are scalar functions of the wavenumber $k \equiv |\boldsymbol{k}|$ only, $\hat{\delta}(\ )$ is the three-dimensional Dirac delta function multiplied by $(2\pi)^3$, and δ_{ij} is the Kronecker delta function. Note that we distinguish scalar functions (e.g. $B^{\alpha\beta}(k)$) from the original vector functions (e.g. $B^{\alpha\beta}(\boldsymbol{k},\boldsymbol{p})$) by the arguments. In general, $B^{\alpha\beta}(k)$ can be a complex function of the wavenumber k (autocorrelations are by definition real functions). However, homogeneity and the reality condition relate the transforms of $B^{\alpha\beta}(\boldsymbol{x}_1,\boldsymbol{x}_2)$ and $B^{\alpha\beta}(\boldsymbol{x}_2,\boldsymbol{x}_1)$ as being complex conjugates of each other. As the turbulence is defined to be mirror symmetric (i.e. no helicity), the function $B^{\alpha\beta}(k)$ will remain a real function for all time.

Although we now have relations for the scalar wavenumber spectra in terms of their vector counterparts, it is actually more common to work with spherical integrals of the Fourier transformed two-point correlations given above. Thus an energy spectrum, $E(k)$, can be defined as

$$E(k) \equiv \frac{1}{(2\pi)^3} \iint R_{ij}(\boldsymbol{k},\boldsymbol{p})\,\hat{\mathrm{d}}\boldsymbol{p}\,k^2\,\mathrm{d}\Omega_k = \frac{k^2 R(k)}{2\pi^2}, \tag{2.15}$$

where the solid angle $d\Omega_k \equiv \sin\theta_k\,\mathrm{d}\theta_k\,\mathrm{d}\phi_k$ and the limits on the spherical angles are $0 \leqslant \theta_k \leqslant \pi$ and $0 \leqslant \phi_k \leqslant 2\pi$. The scalar spectra can be similarly defined by

$$E^i_\phi(k) \equiv \frac{1}{(2\pi)^3} \iint B^i(\boldsymbol{k},\boldsymbol{p})\,\hat{\mathrm{d}}\boldsymbol{p}\,k^2\,\mathrm{d}\Omega_k = \frac{k^2 B^i(k)}{\pi^2}, \tag{2.16}$$

where i refers to α, β, or $\alpha\beta$. The transport equation for $E^\alpha_\phi(k)$ can be written in general as

$$\left[\frac{\partial}{\partial t} + \frac{2}{Pe_\alpha}k^2\right] E^\alpha_\phi(k,t) = Tr^\alpha(k,t). \tag{2.17}$$

In this equation, only the transfer spectrum, $Tr^\alpha(k,t)$, needs to be modelled, since the diffusion terms are exact. The non-local wavenumber representation of the transfer spectrum as given by EDQNM (Lesieur 1987) is

$$Tr^\alpha(k,t) = \iint_\Delta \left[g_1(k,p,q)E(p)E^\alpha_\phi(q,t) - g_2(k,p,q)E(p)E^\alpha_\phi(k,t)\right] \theta\left({}^\alpha\mu^{pkq}_M\right)\,\mathrm{d}p\,\mathrm{d}q, \tag{2.18}$$

where g_1 and g_2 are geometric factors, Δ indicates that the integration only occurs over that portion of the (p,q)-plane for which k, p, and q form a triad (triangle), and θ is a time-dependent function which comes from the Markovianization of the triple correlation. The inverse time scale, ${}^\alpha\mu^{pkq}_M$, is defined by

$${}^\alpha\mu^{pkq}_M = c_{1M}\,\mu^p + c_{2M}(\mu^k + \mu^q) + \frac{1}{Re_L}p^2 + \frac{1}{Pe_\alpha}k^2 + \frac{1}{Pe_\alpha}q^2, \tag{2.19}$$

where μ^k is calculated in the manner suggested by Pouquet *et al.* (1975) and takes the form

$$\mu^k = \frac{1}{\sqrt{2}\pi}\sqrt{\int_0^k \tilde{k}^4 R(\tilde{k})\,\mathrm{d}\tilde{k}}. \tag{2.20}$$

The time-dependent function, $\theta(\gamma)$, is given by

$$\theta(\gamma) = \frac{1 - \mathrm{e}^{-\gamma t}}{\gamma} \tag{2.21}$$

and the geometric factors, g_1 and g_2, by

$$g_1(k,p,q) = \frac{N^2 k}{p^3 q}, \tag{2.22}$$

$$g_2(k,p,q) = \frac{N^2 q}{p^3 k}, \tag{2.23}$$

where

$$N^2 \equiv \frac{(k+p+q)(k+p-q)(p+q-k)(q+k-p)}{4}. \tag{2.24}$$

(See the Appendix for a clarification of the relationship between the present notation and the classical notation found in, for example, Lesieur 1987.) By replacing α in (2.17)–(2.19) with β, one obtains the evolution equation for E^{β}_{ϕ}.

The transport equation for $E^{\alpha\beta}(k,t)$ can be written in an analogous fashion to that for $E^{\alpha}(k,t)$

$$\left[\frac{\partial}{\partial t} + \left(\frac{1}{Pe_\alpha} + \frac{1}{Pe_\beta}\right) k^2\right] E^{\alpha\beta}_{\phi}(k,t) = Tr^{\alpha\beta}(k,t), \tag{2.25}$$

where again, only the transfer spectrum needs to be modelled. As the derivation of the EDQNM expression for $Tr^{\alpha\beta}(k,t)$ follows as a straightforward extension of the one for $Tr^{\alpha}(k,t)$ (which is discussed in detail in Lesieur 1987), we will simply list the final result

$$Tr^{\alpha\beta}(k,t) = \iint_{\Delta} [g_1(k,p,q)E(p)E^{\alpha\beta}_{\phi}(q,t) - g_2(k,p,q)E(p)E^{\alpha\beta}_{\phi}(k,t)] \times \frac{\left[\theta\left({}^{\alpha\beta}\mu^{pkq}_M\right) + \theta\left({}^{\beta\alpha}\mu^{pkq}_M\right)\right]}{2}\,\mathrm{d}p\,\mathrm{d}q, \tag{2.26}$$

where the inverse time scale ${}^{\alpha\beta}\mu^{pkq}_M$ is defined as

$${}^{\alpha\beta}\mu^{pkq}_M = c_{1M}\,\mu^p + c_{2M}(\mu^k + \mu^q) + \frac{1}{Re_L}p^2 + \frac{1}{Pe_\alpha}k^2 + \frac{1}{Pe_\beta}q^2 \tag{2.27}$$

(simply switch α and β to obtain ${}^{\beta\alpha}\mu^{pkq}_M$).

Conservation of each species implies that the transfer process is merely a redistributive one, and thus the integral of the transfer spectrum over all wavenumbers must be zero. In particular, conservation will be guaranteed by satisfying two criteria. The first is that the same θ weighting factor must multiply the positive and negative terms in the transfer spectrum. This is clearly evident in (2.18) and (2.26). The second criterion is that the θ weighting factor must be symmetric under an interchange of k and q. This guarantees that the integral over all wavenumbers, k, of the positive and negative terms of the transfer function precisely cancel. Thus, the EDQNM model conserves both the scalar autocorrelations and covariance.

There are two unknown constants that must be specified to complete the model. Following the analysis of Andre & Lesieur (1977), the coefficients c_{1M} and c_{2M} are assigned values of 0.36.

2.2. *Forcing*

As the equations for the scalar spectra contain no source terms, the standard practice is to initialize the spectra to non-zero values and let them decay in either decaying or stationary turbulence. However, in order to highlight the problems related to realizability and conservation of transfer, it will be convenient to add an external forcing term $F(k)$ to the right-hand sides of (2.17) and (2.25). This forcing term will allow us to initialize all three scalar spectra to zero at $t = 0$, and the spectra will build up as result of the forcing and eventually reach a stationary state due to a balance between the forcing and scalar dissipation. The mechanics of the forcing are quite simple and the method used here is similar to the one described by Lesieur (1987) to obtain a stationary energy spectrum. Note that the forcing term is assumed not to affect the closure models for the transfer spectra (i.e. (2.18) and (2.26)).

To mimic the source terms in the scalar equations that arise in the presence of a uniform mean scalar gradient (discussed in § 4 and § 5 in greater detail), the forcing is

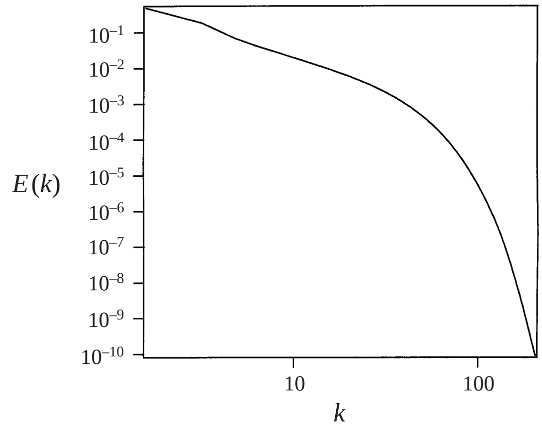

FIGURE 1. EDQNM stationary energy spectrum for $Re_L = 594$. This spectrum was used for all scalar runs and relevant length/time scale information concerning this spectrum can be found in table 1. Note that the ordinate and abscissa of this and all future plots are dimensionless based on the turbulence intensity, u', and integral length scale, L.

designed to add scalar energy to the first two wavenumbers only. Non-local transfer, moderated by scalar dissipation, will then fill in the rest of the scalar spectrum. We choose $F(1) = 0.12$, $F(2) = 0.50$ and $F(k) = 0$ for all other wavenumbers (note that here 1 and 2 refer to the first two wavenumbers, corresponding to $k = 1.59$ and $k = 3.18$ respectively). At steady state, this corresponds to a scalar dissipation rate $\epsilon_\phi = 1.0$ for each of the scalars. It should be noted that each scalar is forced identically so that ϕ'_α and ϕ'_β are initially perfectly correlated. However with time, the scalars will decorrelate as a result of differences in their molecular diffusivities (i.e. differential diffusion).

3. Isotropic scalar results and discussion

The transport equations for the scalar autocorrelation and covariance spectra ($E^\alpha_\phi(k,t)$, $E^\beta_\phi(k,t)$, and $E^{\alpha\beta}_\phi(k,t)$) combined with a stationary EDQNM energy spectrum $E(k)$ (see Lesieur 1987), have been solved numerically on a uniform grid with 128 wavenumbers and non-dimensional $\Delta k = 1.59$ (i.e. the model is approximating a 256^3 direct numerical simulation). For more detailed information on the numerical scheme used to solve the coupled integro-differential equations, the interested reader should refer to Herr *et al.* (1996).

There are three dimensionless parameters in the present study (Re_L, Sc_α, and Sc_β), although no attempt here is made to vary the intensity or composition of the stationary isotropic turbulence. That is, a single energy spectrum with $Re_L = 594$ has been utilized for all runs. This spectrum is shown in figure 1 and a summary of the relevant single-point length/time scale statistics associated with the spectrum can be found in table 1. Note that the energy spectrum and Fourier wavenumber have been made dimensionless by using the variables u' and L, where u' is the r.m.s. fluctuating velocity and L is the integral length scale of the turbulence. Adequate resolution of the dissipation region (small length scales) is assured by adhering to the criterion suggested by Eswaran & Pope (1988) that $k_{max}\eta > 1$, where η is the Kolmogorov length scale.

Parameter	Definition	Value
u'	turbulence intensity	1.016
ϵ	dissipation rate	0.25
ν	kinematic viscosity	0.00272
L	integral length scale	1.591
λ	Taylor microscale	0.410
η	Kolmogorov length scale	0.0168
T_e	eddy turnover time	1.566
Re_L	Reynolds number (integral scale)	594
Re_λ	Reynolds number (microscale)	153

TABLE 1. Turbulence parameters associated with the energy spectrum.

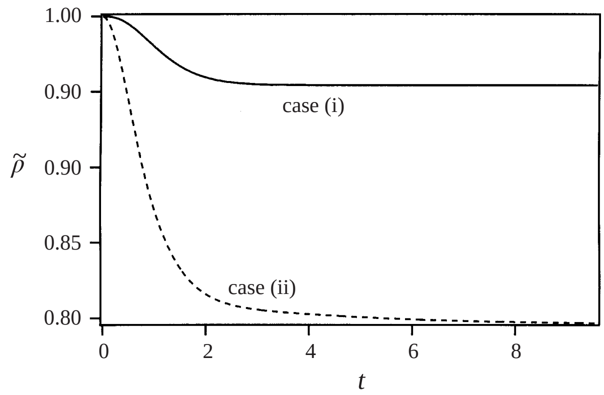

FIGURE 2. Correlation coefficient vs. dimensionless time for $Sc_\alpha = 1$ and $Sc_\beta = 0.1$ (hereafter case (i)) and $Sc_\alpha = 1$ and $Sc_\beta = 0.01$ (hereafter case (ii)). Notice that for both cases $\tilde{\rho} \leqslant 1$ for all time, indicating the integral Cauchy–Schwartz condition is satisfied.

3.1. *Realizability*

Following Yeung & Pope (1993) and Yeung (1996), it is convenient to define a coherency spectrum, which is a normalized covariance spectrum, as

$$\rho(k) \equiv \frac{E_\phi^{\alpha\beta}(k)}{\sqrt{E_\phi^\alpha(k) E_\phi^\beta(k)}}. \tag{3.1}$$

The Cauchy–Schwartz condition implies that $\rho(k) \leqslant 1$ for all k. Before considering $\rho(k)$ however, it will be insightful to consider the time history of the correlation coefficient, a single-point statistic denoted by $\tilde{\rho}$ and defined as

$$\tilde{\rho} \equiv \frac{\overline{\phi'_\alpha \phi'_\beta}}{\sqrt{\overline{\phi'_\alpha \phi'_\alpha}\ \overline{\phi'_\beta \phi'_\beta}}}, \tag{3.2}$$

where the overbars denote total mean quantities (e.g. $\overline{\phi'_\alpha \phi'_\alpha} = \int_0^{k_{max}} E_\phi^\alpha(k)\,\mathrm{d}k$, and similar equations apply for the other two mean quantities in (3.2)). The Cauchy–Schwartz condition also implies that $\tilde{\rho}(t) \leqslant 1$ for all time.

Figure 2 shows the correlation coefficient as a function of dimensionless time for the case where $Sc_\alpha = 1$ and $Sc_\beta = 0.1$ and also for $Sc_\alpha = 1$ and $Sc_\beta = 0.01$. These

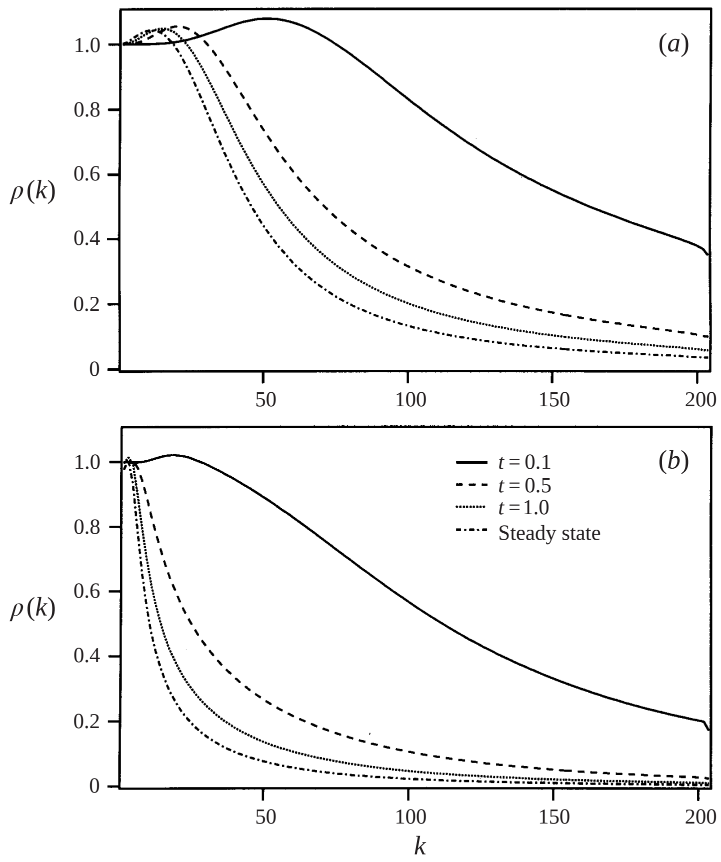

FIGURE 3. Evolution of the coherency spectrum for (*a*) case (i) and (*b*) case (ii). Notice that $\rho(k) > 1$ over a range of wavenumbers in violation of the Cauchy–Schwartz inequality. The violation occurs initially at high wavenumbers and moves to lower wavenumbers with increasing time, although it is still evident even after the spectrum reaches steady state.

two sets of Schmidt numbers will be referred to so frequently that it will be useful to denote ($Sc_\alpha = 1$, $Sc_\beta = 0.1$) by (i) and ($Sc_\alpha = 1$, $Sc_\beta = 0.01$) by (ii). The coefficient starts near unity because the two scalars have identical initial conditions and are forced identically, and are therefore nearly perfectly correlated at short times. With increasing time, the effects of differential diffusion are felt, causing the correlation coefficient to decrease. Notice that the correlation coefficient is less than unity for all time and is therefore realizable. Thus, although we will show that the coherency spectrum, $\rho(k)$, violates the Cauchy–Schwartz inequality over a range of wavenumbers, integrated quantities such as the correlation coefficient remain realizable for all time. From the figure, it is clear that ϕ'_α and ϕ'_β for case (ii) not only decorrelate much more rapidly at short times, as is evident from the steeper slope at $t = 0$, but also to a greater extent than is observed in case (i).

Figure 3 shows the evolution of the coherency spectrum for cases (i) and (ii). Here it is apparent that $\rho(k)$ exceeds unity, violating the Cauchy–Schwartz inequality over a range of wavenumbers. Notice that the violation at short times occurs at high wavenumbers, but as time increases the range of wavenumbers in violation of the Cauchy–Schwartz inequality moves toward lower wavenumbers (i.e. larger scales). It is worth noting that a careful examination of the numerical algorithm used to solve the integro-differential equations shows that the unrealizable spectra are not a numerical artifact, but result from the EDQNM closure. The result is surprising since the present model is a straightforward extension (with no additional assumptions) of the earlier models (Lesieur 1987). Apparently, requiring that $\rho(k) \leqslant 1$ provides a more stringent realizability test of the EDQNM procedure than simple positive definiteness of the autocorrelation spectra. Because of the complexity of the integral responsible for scalar transfer, it is difficult to identify the cause of the partially unrealizable solution. To assist in this regard, we shall develop a spectral equation based on a stochastic model of scalar transport. The advantage of this approach is that the existence of an underlying stochastic model guarantees that the model is realizable.

3.2. *Langevin model*

Under certain circumstances, it is possible to prove that the EDQNM spectral model is related to a Langevin equation. For example, the EDQNM closure for the energy equation was shown by Orszag (1973) to be the exact solution for an ensemble of velocities that each obey the following Langevin equation:

$$\frac{\partial \hat{u}_i(\boldsymbol{k},t)}{\partial t} = -\gamma_u(k,t)\hat{u}_i(\boldsymbol{k},t) + q_i(\boldsymbol{k},t), \tag{3.3}$$

where $\hat{u}_i(\boldsymbol{k},t)$ is a mode of the velocity field for a specific member of the ensemble, $\gamma_u(k,t)$ is a non-stochastic damping function and $q_i(\boldsymbol{k},t)$ is a stochastic forcing function. For consistency with the EDQNM model, $\gamma_u(k,t)$ and $q_i(\boldsymbol{k},t)$ are defined as

$$\gamma_u(k,t) = Re_L^{-1}k^2 + \frac{1}{2}\iint_\Delta \mathrm{d}p\,\mathrm{d}q \frac{k}{pq}\theta(\mu_R^{kpq})\frac{p}{k}(xy+z^3)E(q,t), \tag{3.4}$$

$$q_i(\boldsymbol{k},t) = -\mathrm{i}P_{imn}(\boldsymbol{k})\int a(t)\sqrt{\theta(\mu_R^{kpq})}\hat{v}_m(\boldsymbol{p},t)\hat{w}_n(\boldsymbol{q},t)\,\hat{\mathrm{d}}\boldsymbol{p}, \tag{3.5}$$

where homogeneity requires that $\boldsymbol{k}+\boldsymbol{p}+\boldsymbol{q}=0$, $a(t)$ is a Gaussian random variable that satisfies $\overline{a(t)} = 0$ and $\overline{a(t)a(t')} = \delta(t-t')$, $\hat{v}_i(\boldsymbol{p},t)$ and $\hat{w}_i(\boldsymbol{q},t)$ are statistically independent random Gaussian velocity fields that are chosen to be consistent with the energy spectrum $E(k,t)$, μ_R^{kpq} is the inverse time scale for the energy spectrum (see (4.16) for the definition) and x, y and z are the cosines of the angles between the wavevectors of the (k,p,q) triad defined as

$$x \equiv \frac{\boldsymbol{p}\cdot\boldsymbol{q}}{pq} = \frac{k^2-p^2-q^2}{2pq}, \tag{3.6}$$

$$y \equiv \frac{\boldsymbol{k}\cdot\boldsymbol{q}}{kq} = \frac{p^2-k^2-q^2}{2kq}, \tag{3.7}$$

$$z \equiv \frac{\boldsymbol{k}\cdot\boldsymbol{p}}{kp} = \frac{q^2-k^2-p^2}{2kp}. \tag{3.8}$$

An analogous Langevin equation for the scalar field can be derived by extending the approach taken by Orszag. To simplify the nomenclature, we represent the two scalar spectra in terms of a vector as follows: $\Phi(\boldsymbol{k},t) \equiv (\hat{\phi}_\alpha(\boldsymbol{k},t), \hat{\phi}_\beta(\boldsymbol{k},t))$. The Langevin equation for $\Phi(\boldsymbol{k},t)$ takes the general form

$$\frac{\partial \Phi(\boldsymbol{k},t)}{\partial t} + \boldsymbol{A}(k,t)\cdot\Phi(\boldsymbol{k},t) = \boldsymbol{r}(\boldsymbol{k},t), \tag{3.9}$$

where $\boldsymbol{A}(k,t)$ is a second-order coefficient (i.e. non-stochastic) matrix, which we will define shortly and $\boldsymbol{r}(\boldsymbol{k},t)$ is a random forcing vector defined as

$$\boldsymbol{r}(\boldsymbol{k},t) = -\mathrm{i}k_m \int a'(t)\hat{v}_m(\boldsymbol{p},t)\boldsymbol{C}(q,t)\cdot\boldsymbol{\Psi}(\boldsymbol{q},t)\,\mathrm{d}p. \tag{3.10}$$

$\boldsymbol{C}(q,t)$ is another coefficient matrix, $\boldsymbol{\Psi}(\boldsymbol{q},t)$ is a vector consisting of two independent random Gaussian variables with zero mean and unit variance, and $a'(t)$ is another random Gaussian variable with properties identical to $a(t)$. Multiplying (3.9) by $\boldsymbol{\Phi}(\boldsymbol{k},t)$ – obtained by formal integration – and recombining the result yields a closed equation for the scalar covariance matrix, defined as

$$\boldsymbol{E}_\phi(k,t) \equiv \begin{pmatrix} E_\phi^\alpha(k,t) & E_\phi^{\alpha\beta}(k,t) \\ E_\phi^{\alpha\beta}(k,t) & E_\phi^\beta(k,t) \end{pmatrix}. \tag{3.11}$$

The result is

$$\frac{\partial \boldsymbol{E}_\phi(k,t)}{\partial t} + \boldsymbol{A}(k,t)\cdot\boldsymbol{E}_\phi(k,t) + [\boldsymbol{A}(k,t)\cdot\boldsymbol{E}_\phi(k,t)]^T = \iint_\Delta g_1(k,p,q)E(p)\boldsymbol{D}(q,t)\,\mathrm{d}p\,\mathrm{d}q, \tag{3.12}$$

where $\boldsymbol{D}(q,t) \equiv \boldsymbol{C}(q,t)\cdot\boldsymbol{C}^T(q,t)$. The superscript T refers to the transpose of the matrix. To be consistent with the EDQNM model for the scalar autocorrelation spectra, the matrix $\boldsymbol{A}(k,t)$ must have the following form:

$$\boldsymbol{A}(k,t) \equiv \begin{pmatrix} \gamma_\alpha(k,t) & 0 \\ 0 & \gamma_\beta(k,t) \end{pmatrix}, \tag{3.13}$$

where

$$\gamma_\alpha(k,t) = Pe_\alpha^{-1} + \tfrac{1}{2}\iint_\Delta \mathrm{d}p\,\mathrm{d}q\, g_2(k,p,q)\theta({}^\alpha\mu_M^{pkq})E(p,t) \tag{3.14}$$

and $\gamma_\beta(k,t)$ follows by analogy. Likewise, $\boldsymbol{D}(q,t)$ takes the form

$$\boldsymbol{D}(q,t) \equiv \begin{pmatrix} \theta({}^\alpha\mu_M^{pkq})E_\phi^\alpha(q,t) & \theta_{12}E_\phi^{\alpha\beta}(q,t) \\ \theta_{12}E_\phi^{\alpha\beta}(q,t) & \theta({}^\beta\mu_M^{pkq})E_\phi^\beta(q,t) \end{pmatrix}, \tag{3.15}$$

where θ_{12} is an unspecified constant. At this stage, the definitions given in (3.13) and (3.15) ensure that the equations for $E^\alpha(k,t)$ and $E^\beta(k,t)$ represented in (3.12) are consistent with the EDQNM model. The equation for $E^{\alpha\beta}(k,t)$ will depend on how the coefficient θ_{12} is chosen.

Constructing the Langevin model requires that we solve for the $\boldsymbol{C}(q,t)$ matrix. Using the Cholesky decomposition (Golub & Loan 1983), we can construct a lower-triangular matrix $\boldsymbol{C}(q,t)$; however $\boldsymbol{D}(q,t)$ must be positive definite (i.e. $\mathrm{Det}(\boldsymbol{D}) \geqslant 0$) to make this possible. This implies that the following constraint must hold:

$$\theta({}^\alpha\mu_M^{pkq})\theta({}^\beta\mu_M^{pkq})E_\phi^\alpha(q,t)E_\phi^\beta(q,t) - [\theta_{12}E_\phi^{\alpha\beta}(q,t)]^2 \geqslant 0. \tag{3.16}$$

If we assume that the Schwartz equality is satisfied (i.e. $E_\phi^{\alpha\beta}(q,t) = \sqrt{E_\phi^\alpha(q,t)E_\phi^\beta(q,t)}$), then the worst case becomes

$$\theta_{12} \leqslant \sqrt{\theta({}^\alpha\mu_M^{pkq})\theta({}^\beta\mu_M^{pkq})}, \tag{3.17}$$

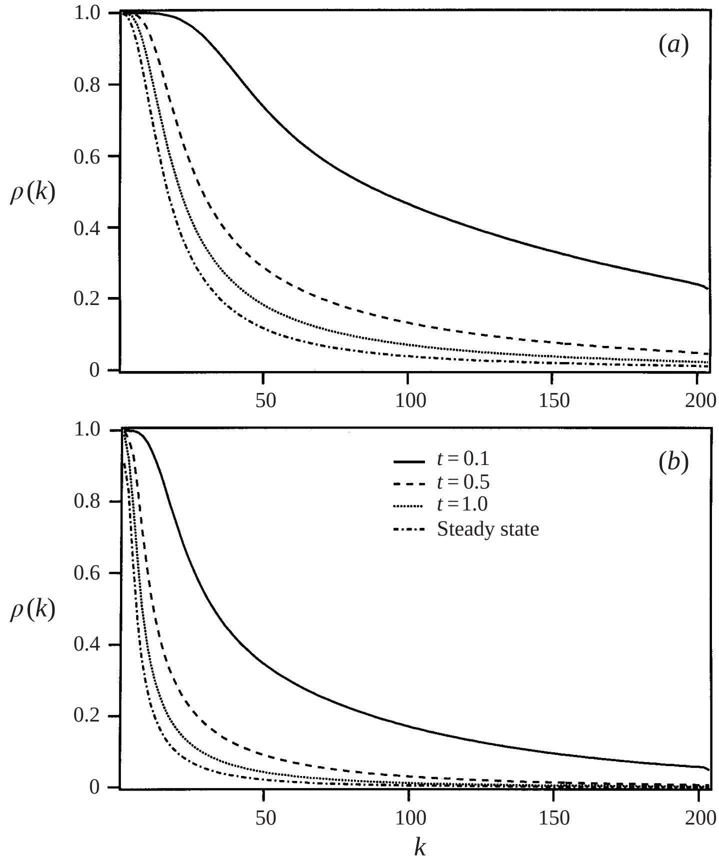

FIGURE 4. Coherency spectrum for the covariance spectrum derived using the Langevin model for the transfer. The result is realizable for all time; however, closer examination shows that the covariance spectrum is not properly conserved by the transfer function $Tr_L^{\alpha\beta}(k)$, i.e. $\int_0^\infty Tr_L^{\alpha\beta}(k)\,\mathrm{d}k \neq 0$.

(i.e. a sufficient condition for the existence of the Langevin model is that θ_{12} is less than the geometric mean of the θ-values for each autocorrelation spectrum). If we satisfy the equality shown in (3.17), the equation for the transfer spectrum, $Tr_L^{\alpha\beta}(k,t)$, becomes

$$Tr_L^{\alpha\beta}(k,t) = \iint_\Delta \Big[g_1(k,p,q)\sqrt{\theta({}^\alpha\mu_M^{pkq})\theta({}^\beta\mu_M^{pkq})}E(p)E_\phi^{\alpha\beta}(q,t) - g_2(k,p,q)\tfrac{1}{2}[\theta({}^\alpha\mu_M^{pkq}) + \theta({}^\beta\mu_M^{pkq})]\, E(p)E_\phi^{\alpha\beta}(k,t)\Big]\,\mathrm{d}p\,\mathrm{d}q. \qquad (3.18)$$

The major difference lies in the definitions of the θ-values, which are now defined in terms of a geometric and arithmetic mean of the autocorrelation values, instead of the mixed coefficient that arises from the EDQNM procedure. As a result, the Langevin model yields an asymmetric transfer of the scalar covariance; that is, transfer into the wavenumber k is controlled by the geometric mean of the autocorrelation θ-values, whereas transfer out of the wavenumber k is controlled by the arithmetic mean of the same.

Figure 4 shows the evolution of the coherency spectrum for cases (i) and (ii) using $Tr_L^{\alpha\beta}(k)$ defined in (3.18). It is apparent that the modified transfer function repairs

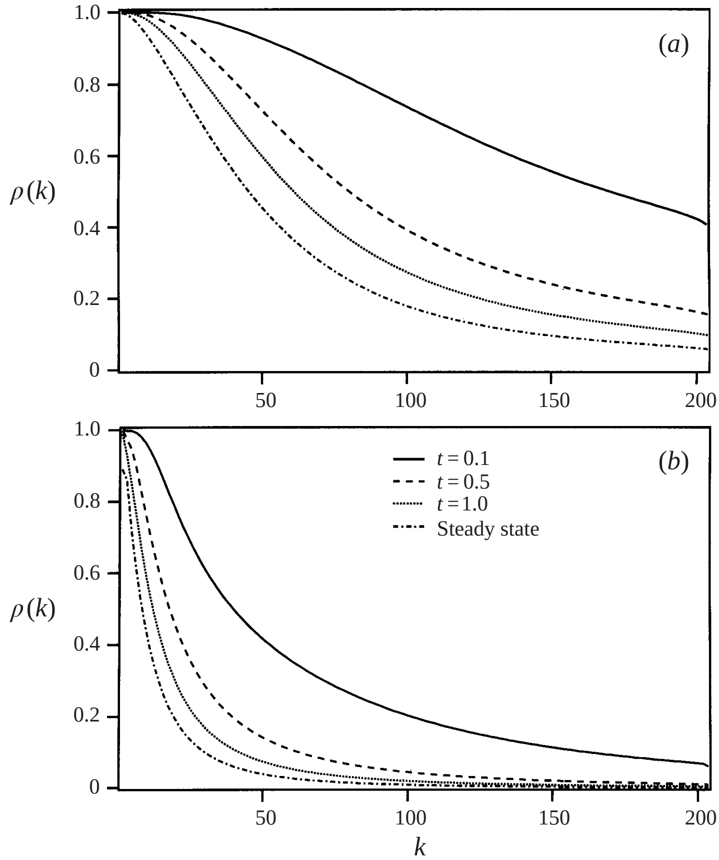

FIGURE 5. Coherency spectrum for the EDQNM model with the modified coefficients for (*a*) case (i) and (*b*) case (ii). The result is realizable and the transfer term properly conserves the scalar covariance.

the problem with realizability found for the standard EDQNM model. Indeed, the resulting spectra are realizable for all time. However, one important problem with (3.18) is that it no longer conserves the scalar covariance (i.e. $\int_0^\infty Tr_L^{\alpha\beta}(k)\,\mathrm{d}k \neq 0$). This unphysical result is directly related to the break in symmetry between transfer into and out of each wavenumber. The use of different θ-functions for the two parts of $Tr_L^{\alpha\beta}(k,t)$ in (3.18) destroys the overall conservation of the scalar covariance. However, under the circumstance that the arithmetic and geometric means of the θ-functions are equal, then both conservation and realizability would be obtained. This condition can occur if and only if the autocorrelation inverse time scales are defined to be independent of the scalar molecular diffusivities. This assumption for the coefficients yields a model that is both conservative and realizable. To accomplish this, we replace the inverse time scales by

$$ {}^{\alpha}\mu_M^{pkq} = {}^{\beta}\mu_M^{pkq} = {}^{\alpha\beta}\mu_M^{pkq} = c_{1M}\mu^p + c_{2M}(\mu^k + \mu^q) + \frac{(k^2+p^2+q^2)}{Re_L} \equiv \mu_M^{pkq}, \qquad (3.19) $$

in which the explicit dependence on the scalar molecular diffusivities (here appearing in the form of Pe_α and Pe_β) is replaced by the kinematic viscosity (here Re_L). The

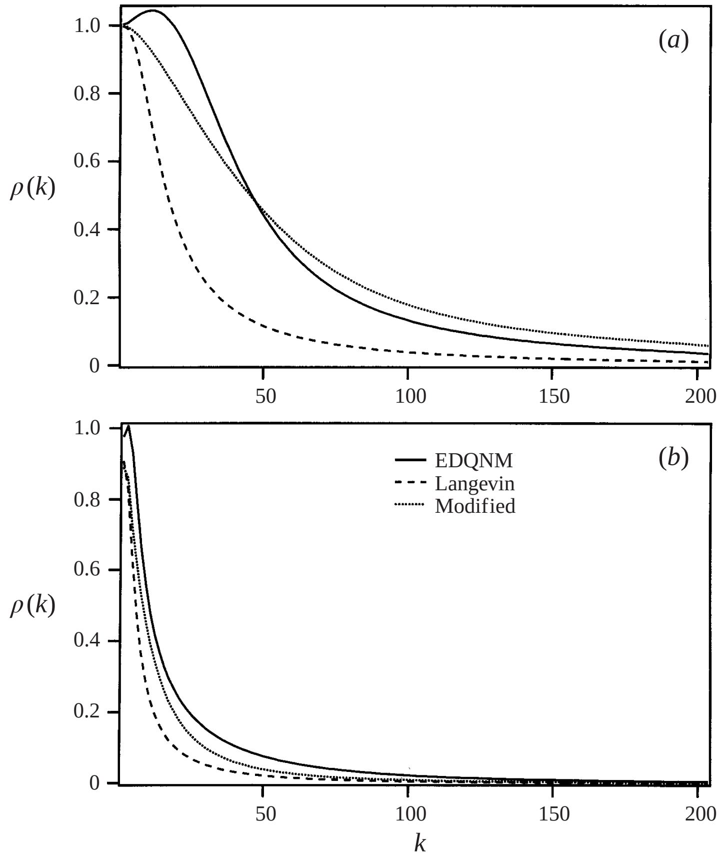

FIGURE 6. Comparison of the steady-state coherency spectra for the original EDQNM model, the Langevin model, and the EDQNM model with modified coefficients for (*a*) case (i) and (*b*) case (ii).

use of these coefficients yields the following modified transfer function:

$$Tr_M^{\alpha\beta}(k,t) = \iint_\Delta [g_1(k,p,q)E(p)E_\phi^{\alpha\beta}(q,t) - g_2(k,p,q)E(p)E_\phi^{\alpha\beta}(k,t)]\theta(\mu_M^{pkq})\mathrm{d}p\,\mathrm{d}q. \tag{3.20}$$

It should be noted that the choice is not unique; however, it is believed to be the best choice because it accomplishes the objective with the least modification of the coefficients.

Figure 5 shows the evolution of the coherency spectrum for the EDQNM model with the revised inverse time scales. Once again, the coherency spectrum remains below unity for all time and wavenumbers. A comparison of the steady-state results from the original EDQNM model, the Langevin model and the newly proposed model using (3.19) for cases (i) and (ii) is shown in figure 6. It is apparent that this modest change has a significant impact on the coherency spectrum at all wavenumbers. As noted earlier, the new model can be considered a physical model, as it satisfies the Cauchy–Schwartz condition and the transfer term properly conserves the scalar covariance; however, its accuracy must be assessed by comparison with either experimental data or numerical simulations. This will be the topic of a future paper.

3.3. *Physical explanation*

In hindsight, it is possible to explain the results presented above without invoking the Langevin analysis. Imagine that scalars α and β are being convected by turbulence, and at some instant in time t, $\phi'_\alpha = \phi'_\beta$ over some portion of the spectrum, corresponding to the Cauchy–Schwartz equality being satisfied. By definition, the triple correlations leading to the respective transfer terms over that same range of the spectrum are identical and so at that instant one expects $Tr^{\alpha}(k,t) = Tr^{\beta}(k,t) = Tr^{\alpha\beta}(k,t)$, when $E^{\alpha}(k,t) = E^{\beta}(k,t) = E^{\alpha\beta}(k,t)$. However, it is readily apparent from (2.18) and (2.26) and the definitions of the inverse time scales ((2.19) and (2.27)) that the EDQNM model will not produce identical transfer terms under these circumstances because the models for the three transfer functions are explicit functions of the respective scalar molecular diffusivities (through the definitions of the θ-functions). Notice that this explicit dependence is eliminated in the modified definition of the inverse time scale given in (3.19), and so the modified model predicts identical transfer functions under these circumstances.

Thus, we conjecture that a sufficient condition for realizability is that the auto- and cross-correlation transfer functions should reduce to identical functions of k whenever the three scalar spectra are equal. There is additional evidence to support this conjecture. For example, the scalar molecular diffusivities (in the dimensionless equations, the Péclet numbers) are multiplied by factors k^2, p^2 and q^2 and thus one expects their effect to be greatest at high wavenumbers. This is consistent with the observation in figure 3 that the coherency spectrum is initially unrealizable at high wavenumbers.

The conjecture is somewhat surprising because it suggests that the terms arising from the exact linear diffusion terms in the original differential equation for the triple correlations are causing the realizability problem. However, the form they take in the final expression for the transfer functions is not exact because of the Markovian approximation. It is at this step that the explicit dependence of the transfer functions on the molecular diffusivities is introduced. Clearly, relaxing the Markovian approximation by integrating a coupled system of ODEs for the θ-functions would relieve this problem; however, such a calculation is extremely numerically intensive, as the number of θ-functions to be integrated scales like N^3 for N grid points. Moreover, relaxing the Markovian approximation jeopardizes the realizability of the scalar autocorrelation spectra (i.e. positive definitness). Thus, the simplest and most robust solution to the problem may be to modify the inverse time scales of the Markovianized model, as suggested here.

4. Equations for scalars with mean gradients

The equation governing the advection and diffusion of a passive scalar with constant physical properties is given in (2.1). In this section, we are concerned with the mixing of passive scalars with a uniform mean gradient in a single direction. The Reynolds decomposition of the scalar concentration into a mean and fluctuating component now becomes $\phi_\alpha = \phi'_\alpha + \Gamma_\alpha x_3$, where Γ_α is the magnitude of the mean gradient of species α and its direction is taken to be x_3 without loss of generality. As a result of the mean gradient, all correlations involving the scalar will be axisymmetric in the (x_1, x_2)-plane instead of being isotropic.

The governing equation for the scalars can be found by imposing the mean gradient on (2.1). If we then non-dimensionalize the resulting equation using the integral scale

L (for x_i), the r.m.s. fluctuating velocity u' (for u'_i), the large-eddy turnover time L/u', $\Gamma_\alpha L$ (for ϕ'_α) and $\Gamma_\beta L$ (for ϕ'_β), we obtain the following:

$$\frac{\partial \phi'_\alpha}{\partial t} + \frac{\partial}{\partial x_i}(u'_i \phi'_\alpha) + u'_3 = \frac{1}{Pe_\alpha}\frac{\partial^2 \phi'_\alpha}{\partial x_i \partial x_i}, \tag{4.1}$$

$$\frac{\partial \phi'_\beta}{\partial t} + \frac{\partial}{\partial x_i}(u'_i \phi'_\beta) + u'_3 = \frac{1}{Pe_\beta}\frac{\partial^2 \phi'_\beta}{\partial x_i \partial x_i}, \tag{4.2}$$

where Pe_α and Pe_β retain their definitions from § 2. As (4.1) and (4.2) are linear with respect to ϕ', the magnitude of the mean gradients effectively scales out of the problem. Once again, the same variables are used to represent the dimensionless variables since all future equations will be written in dimensionless form. Equations (4.1) and (4.2) represent the starting point for deriving transport equations for all of the two- and three-point correlations that follow.

4.1. *Two-point correlations*

In addition to the two-point correlations identified in (2.4)–(2.7) for the isotropic scalar study, the covariance spectrum in the presence of a uniform mean gradient will further depend upon the Fourier transform of the following two-point correlations:

$$Q_i^\alpha(\boldsymbol{x}_1, \boldsymbol{x}_2) \equiv \overline{u'_i(\boldsymbol{x}_1)\phi'_\alpha(\boldsymbol{x}_2)}, \tag{4.3}$$

$$Q_i^\beta(\boldsymbol{x}_1, \boldsymbol{x}_2) \equiv \overline{u'_i(\boldsymbol{x}_1)\phi'_\beta(\boldsymbol{x}_2)}. \tag{4.4}$$

Thus, in addition to the equation for the scalar covariance, a transport equation for $Q_i^\alpha(\boldsymbol{x}_1, \boldsymbol{x}_2)$ (and by analogy $Q_i^\beta(\boldsymbol{x}_1, \boldsymbol{x}_2)$) must be derived. Moreover, the equation for the scalar covariance will be substantially modified to account for the reduction in symmetry from isotropic to axisymmetric and to include additional transfer terms. It should be noted that the derivations in this part of the study closely parallel those performed by Herr *et al.* (1996), in which the EDQNM theory was applied to a single passive scalar with a uniform mean gradient.

The exact transport equation for $B^{\alpha\beta}(\boldsymbol{x}_1, \boldsymbol{x}_2)$, the scalar covariance, takes the following form:

$$\left[\frac{\partial}{\partial t} - \frac{1}{Pe_\alpha}\nabla_1^2 - \frac{1}{Pe_\beta}\nabla_2^2\right] B^{\alpha\beta}(\boldsymbol{x}_1, \boldsymbol{x}_2)$$
$$= \underbrace{-\frac{\partial}{\partial x_{1i}} M_i^{\alpha\beta}(\boldsymbol{x}_1, \boldsymbol{x}_1, \boldsymbol{x}_2) - \frac{\partial}{\partial x_{2i}} M_i^{\alpha\beta}(\boldsymbol{x}_2, \boldsymbol{x}_1, \boldsymbol{x}_2)}_{\text{inertial transfer}} \; \underbrace{-Q_3^\beta(\boldsymbol{x}_1, \boldsymbol{x}_2) - Q_3^\alpha(\boldsymbol{x}_2, \boldsymbol{x}_1)}_{\text{source}}, \tag{4.5}$$

where $M_i^{\alpha\beta}(\boldsymbol{x}_1, \boldsymbol{x}_2, \boldsymbol{x}_3) \equiv \overline{u'_i(\boldsymbol{x}_1)\phi'_\alpha(\boldsymbol{x}_2)\phi'_\beta(\boldsymbol{x}_3)}$, and ∇_1^2 and ∇_2^2 are the Laplacian operators with respect to $\boldsymbol{x}_1$ and $\boldsymbol{x}_2$. Once again, the inertial transfer terms are at higher order and must be modelled via the EDQNM formalism. Before proceeding to the three-point correlations, however, it is necessary to derive transport equations for $Q_i^\alpha(\boldsymbol{x}_1, \boldsymbol{x}_2)$ and $Q_i^\beta(\boldsymbol{x}_1, \boldsymbol{x}_2)$, which constitute the source term for $B^{\alpha\beta}(\boldsymbol{x}_1, \boldsymbol{x}_2)$ in (4.5).

The exact transport equation for $Q_i^\alpha(\boldsymbol{x}_1, \boldsymbol{x}_2)$ the scalar–velocity cross-correlation is given by

$$\left[\frac{\partial}{\partial t} - \frac{1}{Re_L}\nabla_1^2 - \frac{1}{Pe_\alpha}\nabla_2^2\right] Q_i^\alpha(\boldsymbol{x}_1,\boldsymbol{x}_2) = \underbrace{-\frac{1}{2}P_{iab}^1 T_{ab}^\alpha(\boldsymbol{x}_1,\boldsymbol{x}_1,\boldsymbol{x}_2) - \frac{\partial}{\partial x_{2a}} T_{ia}^\alpha(\boldsymbol{x}_1,\boldsymbol{x}_2,\boldsymbol{x}_2)}_{\text{inertial transfer}} \underbrace{-R_{i3}(\boldsymbol{x}_1,\boldsymbol{x}_2)}_{\text{source}}, \tag{4.6}$$

where $T_{ia}^\alpha(\boldsymbol{x}_1,\boldsymbol{x}_2,\boldsymbol{x}_3) \equiv \overline{u_i'(\boldsymbol{x}_1)u_a'(\boldsymbol{x}_2)\phi_\alpha'(\boldsymbol{x}_3)}$. The projection operator P_{iab} is symmetric in the last two indices and is defined as (the superscript 1 implies that all derivatives occur at $\boldsymbol{x}_1$)

$$P_{iab} = \delta_{ia}\frac{\partial}{\partial x_b} + \delta_{ib}\frac{\partial}{\partial x_a} + \frac{2}{\nabla^2}\frac{\partial^3}{\partial x_i x_a x_b}. \tag{4.7}$$

The source term in (4.6) is exact (as was the case for (4.5)), but the triple correlations will have to be modelled.

From the definition of $Q_i^\alpha(\boldsymbol{k},\boldsymbol{p})$ (see (2.8)) it can be shown through vector invariant arguments that tensorial constraints, continuity, and the reality condition will impose the following form (Batchelor 1946; Chandrasekhar 1950):

$$Q_i^\alpha(\boldsymbol{k},\boldsymbol{p}) = \hat{\delta}(\boldsymbol{k}+\boldsymbol{p})P_{i3}(\boldsymbol{k})Q^\alpha(k,\mu), \tag{4.8}$$

where μ is the cosine of the angle between $\boldsymbol{k}$ and the mean gradient vector, $Q^\alpha(k,\mu)$ is a real function of the wavenumber k and angle μ (see (2.14) for the definition of $P_{ij}(\boldsymbol{k})$). Ulitsky & Collins (1997) showed in general that the scalar–velocity correlation can have a second component that is proportional to $\epsilon_{i3j}k_j/k$, where ϵ_{ijk} is the alternating unit tensor. However, under the assumption of mirror-symmetric isotropic turbulence, this term will be identically zero.

Although the velocity field is isotropic, the presence of the uniform mean gradient makes all correlations involving the scalar axisymmetric. From a spectral standpoint, this suggests that the energy spectrum will only be a function of the wavenumber k, but that all scalar spectra will be functions not only of the wavenumber k, but also of the angle μ. In a later section, a discussion will be given on separating the angle dependence from the wavenumber dependence.

Taking the Fourier transform of (4.6) and noting that $P_{i3}(\boldsymbol{k})P_{i3}(\boldsymbol{k}) = (1-\mu^2)$, we obtain the following equation for $Q^\alpha(k,\mu)$:

$$\left[\frac{\partial}{\partial t} + \left(\frac{1}{Re_L} + \frac{1}{Pe_\alpha}\right)k^2\right](1-\mu^2)Q^\alpha(k,\mu) = \underbrace{-k_jP_{i3}(\boldsymbol{k})\iint T_{ij}^\alpha(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})\,\hat{\mathrm{d}}\boldsymbol{p}\,\hat{\mathrm{d}}\boldsymbol{q} - \tfrac{1}{2}P_{3ij}(\boldsymbol{k})\iint T_{ij}^\alpha(\boldsymbol{q},\boldsymbol{p},\boldsymbol{k})\,\hat{\mathrm{d}}\boldsymbol{p}\,\hat{\mathrm{d}}\boldsymbol{q}}_{\text{inertial transfer}} \underbrace{-(1-\mu^2)R(k)}_{\text{source}}. \tag{4.9}$$

The *anisotropic* scalar spectrum now takes the following form (Batchelor 1946; Chandrasekhar 1950):

$$B^{\alpha\beta}(\boldsymbol{k},\boldsymbol{p}) \equiv 2\hat{\delta}(\boldsymbol{k}+\boldsymbol{p})B^{\alpha\beta}(k,\mu), \tag{4.10}$$

where in general, $B^{\alpha\beta}(k,\mu)$ will be a complex function of the wavenumber k and angle μ; however, in accordance with the argument made earlier for $Q_i^\alpha(\boldsymbol{k},\boldsymbol{p})$, the imaginary component will be identically zero as a result of the initial conditions and the assumption of mirror-symmetric turbulence. With this definition, the transport equation for $B^{\alpha\beta}(k,\mu)$ becomes

$$\left[\frac{\partial}{\partial t} + \left(\frac{1}{Pe_\alpha} + \frac{1}{Pe_\beta}\right)k^2\right] B^{\alpha\beta}(k,\mu)$$
$$= \underbrace{-k_i \iint \tfrac{1}{2}[M_i^{\alpha\beta}(\boldsymbol{p},\boldsymbol{k},\boldsymbol{q}) + M_i^{\beta\alpha}(\boldsymbol{p},\boldsymbol{k},\boldsymbol{q})]\,\hat{\mathrm{d}}\boldsymbol{p}\,\hat{\mathrm{d}}\boldsymbol{q}}_{\text{inertial transfer}}$$
$$\underbrace{-\tfrac{1}{2}(1-\mu^2)[Q^\alpha(k,\mu) + Q^\beta(k,\mu)]}_{\text{source}}. \tag{4.11}$$

Note that the triple correlations appearing in (4.9) and (4.11) are purely imaginary; however to simplify the nomenclature, it will be understood that T_{ij}^α and $M_i^{\alpha\beta}$ actually refer to the imaginary parts of T_{ij}^α and $M_i^{\alpha\beta}$ respectively (the real parts of T_{ij}^α and $M_i^{\alpha\beta}$ are identically zero). If the scalar diffusivities were equal, the resulting symmetry of the scalar autocorrelation combined with homogeneity reduces (4.11) to the single-scalar equation (Herr *et al.* 1996)

$$\left[\frac{\partial}{\partial t} + \frac{2}{Pe_\alpha}k^2\right] B^\alpha(k,\mu) = \underbrace{-k_i \iint M_i^\alpha(\boldsymbol{p},\boldsymbol{k},\boldsymbol{q})\,\hat{\mathrm{d}}\boldsymbol{p}\,\hat{\mathrm{d}}\boldsymbol{q}}_{\text{inertial transfer}} \underbrace{-(1-\mu^2)Q^\alpha(k,\mu)}_{\text{source}}. \tag{4.12}$$

4.2. *Three-point correlations*

From (4.9) and (4.11), it is clear that we must derive expressions for $T_{ij}^\alpha(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$ and $M_i^{\alpha\beta}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$ to achieve a closed set of equations. The quasi-normal assumption (Chou 1940; Millionshtchikov 1941) enters at this step, since equations for the temporal evolution of the third-order moments will contain terms involving unknown fourth-order moments. Through the quasi-normal approximation, it is postulated that the joint distributions of all fourth-order moments are nearly Gaussian, and thus, the unknown moments can be expressed as superpositions of products of second-order moments. For example, if $a_i'(\boldsymbol{x}_1)$, $b_j'(\boldsymbol{x}_2)$, $c_k'(\boldsymbol{x}_3)$, and $d_l'(\boldsymbol{x}_4)$ are Gaussian random variables with zero mean, then

$$\overline{a_i'(\boldsymbol{x}_1)b_j'(\boldsymbol{x}_2)c_k'(\boldsymbol{x}_3)d_l'(\boldsymbol{x}_4)} = \overline{a_i'(\boldsymbol{x}_1)b_j'(\boldsymbol{x}_2)}\;\overline{c_k'(\boldsymbol{x}_3)d_l'(\boldsymbol{x}_4)} + \overline{a_i'(\boldsymbol{x}_1)c_k'(\boldsymbol{x}_3)}\;\overline{b_j'(\boldsymbol{x}_2)d_l'(\boldsymbol{x}_4)}$$
$$+\,\overline{a_i'(\boldsymbol{x}_1)d_l'(\boldsymbol{x}_4)}\;\overline{b_j'(\boldsymbol{x}_2)c_k'(\boldsymbol{x}_3)}. \tag{4.13}$$

Although the distributions of the actual fourth-order moments are not identically Gaussian, we can still use (4.13) if we recognize that the equality now becomes an approximation. By applying (4.13) and homogeneity to all the fourth-order moments that appear in the transport equations for $T_{ij}^\alpha(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$ and $M_i^{\alpha\beta}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$, we can obtain time-dependent equations for the third-order moments. If we then use eddy damping to control the fourth-order cumulants and the Markovian approximation to simplify the time history of the third-order moments (Lesieur 1987), then the following closed

$\mathscr{R}_{ij3}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$	$\{-P_{iab}(\boldsymbol{k})P_{ja}(\boldsymbol{p})P_{b3}(\boldsymbol{q})R(p)R(q) - P_{jab}(\boldsymbol{p})P_{ia}(\boldsymbol{k})P_{b3}(\boldsymbol{q})R(k)R(q)$ $-P_{3ab}(\boldsymbol{q})P_{ia}(\boldsymbol{k})P_{jb}(\boldsymbol{p})R(k)R(p)\}\,\hat{\delta}(\boldsymbol{k}+\boldsymbol{p}+\boldsymbol{q})$
$\mathscr{T}^{\alpha}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$	$\{-P_{jab}(\boldsymbol{p})P_{ia}(\boldsymbol{k})P_{b3}(\boldsymbol{q})Q^{\alpha}(q,\mu'')R(k) - P_{iab}(\boldsymbol{k})P_{aj}(\boldsymbol{p})P_{b3}(\boldsymbol{q})Q^{\alpha}(q,\mu'')R(p)$ $-q_nP_{in}(\boldsymbol{k})P_{j3}(\boldsymbol{p})Q^{\alpha}(p,\mu')R(k) - q_nP_{jn}(\boldsymbol{p})P_{i3}(\boldsymbol{k})Q^{\alpha}(k,\mu)R(p)\}\,\hat{\delta}(\boldsymbol{k}+\boldsymbol{p}+\boldsymbol{q})$
$\mathscr{T}^{\beta}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$	$\{-P_{jab}(\boldsymbol{p})P_{ia}(\boldsymbol{k})P_{b3}(\boldsymbol{q})Q^{\beta}(q,\mu'')R(k) - P_{iab}(\boldsymbol{k})P_{aj}(\boldsymbol{p})P_{b3}(\boldsymbol{q})Q^{\beta}(q,\mu'')R(p)$ $-q_nP_{in}(\boldsymbol{k})P_{j3}(\boldsymbol{p})Q^{\beta}(p,\mu')R(k) - q_nP_{jn}(\boldsymbol{p})P_{i3}(\boldsymbol{k})Q^{\beta}(k,\mu)R(p)\}\,\hat{\delta}(\boldsymbol{k}+\boldsymbol{p}+\boldsymbol{q})$
$\mathscr{M}^{\alpha\beta}_{i}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$	$\{-2p_jP_{ij}(\boldsymbol{k})R(k)B^{\alpha\beta}(q,\mu'') - 2q_jP_{ij}(\boldsymbol{k})R(k)B^{\alpha\beta}(p,\mu')$ $-p_jP_{i3}(\boldsymbol{k})P_{j3}(\boldsymbol{q})Q^{\alpha}(k,\mu)Q^{\beta}(q,\mu'') - q_jP_{i3}(\boldsymbol{k})P_{j3}(\boldsymbol{p})Q^{\beta}(k,\mu)Q^{\alpha}(p,\mu')$ $-P_{iab}(\boldsymbol{k})P_{a3}(\boldsymbol{p})P_{b3}(\boldsymbol{q})Q^{\alpha}(p,\mu')Q^{\beta}(q,\mu'')\}\,\hat{\delta}(\boldsymbol{k}+\boldsymbol{p}+\boldsymbol{q})$

TABLE 2. Explicit representation of triple correlations.

expressions can be derived for $T^{\alpha}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$ and $M^{\alpha\beta}_{i}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$:

$$T^{\alpha}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}) = \left[\mathscr{T}^{\alpha}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}) - \frac{\mathscr{R}_{ij3}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})}{\mu_R^{kpq}}\right]\theta({}^{\alpha}\mu_T^{kpq}), \tag{4.14}$$

$$\begin{aligned} M^{\alpha\beta}_{i}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}) = {} & \left[\frac{\mathscr{R}_{i33}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})}{\mu_R^{kpq}} - \mathscr{T}^{\beta}_{i3}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})\right]\xi({}^{\beta}\mu_T^{kpq},{}^{\alpha\beta}\mu_M^{kpq}) \\ & + \left[\frac{\mathscr{R}_{i33}(\boldsymbol{k},\boldsymbol{q},\boldsymbol{p})}{\mu_R^{kqp}} - \mathscr{T}^{\alpha}_{i3}(\boldsymbol{k},\boldsymbol{q},\boldsymbol{p})\right]\xi({}^{\alpha}\mu_T^{kqp},{}^{\alpha\beta}\mu_M^{kpq}) \\ & + \mathscr{M}^{\alpha\beta}_{i}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})\theta({}^{\alpha\beta}\mu_M^{kpq}). \end{aligned} \tag{4.15}$$

The expressions for $\mathscr{R}_{ij3}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$, $\mathscr{T}^{\alpha}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$, $\mathscr{T}^{\beta}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$, and $\mathscr{M}^{\alpha\beta}_{i}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$, can be found in table 2. The new eddy-damping terms are defined as

$$\left.\begin{aligned} \mu_R^{kpq} &= c_R(\mu^k+\mu^p+\mu^q) + \frac{1}{Re_L}(k^2+p^2+q^2), \\ {}^{\alpha}\mu_T^{kpq} &= c_{1T}(\mu^k+\mu^p) + c_{2T}\,\mu^q + \frac{1}{Re_L}(k^2+p^2) + \frac{1}{Pe_\alpha}q^2, \\ {}^{\beta}\mu_T^{kpq} &= c_{1T}(\mu^k+\mu^p) + c_{2T}\,\mu^q + \frac{1}{Re_L}(k^2+p^2) + \frac{1}{Pe_\beta}q^2; \end{aligned}\right\} \tag{4.16}$$

${}^{\alpha\beta}\mu_M^{kpq}$, μ^k and $\theta(\gamma)$ are defined in (2.19)–(2.21). The new time-dependent coefficient, $\xi(\gamma,\delta)$, is given by

$$\xi(\gamma,\delta) = \begin{cases} \dfrac{1}{\gamma}\left[\dfrac{1-\mathrm{e}^{-\delta t}}{\delta} + \dfrac{\mathrm{e}^{-\gamma t}-\mathrm{e}^{-\delta t}}{\gamma-\delta}\right], & \gamma \neq \delta \\ \dfrac{1}{\gamma}\left[\dfrac{1-\mathrm{e}^{-\gamma t}}{\gamma} - t\mathrm{e}^{-\gamma t}\right], & \gamma = \delta. \end{cases} \tag{4.17}$$

There are three additional unknown constants that must be specified to complete the model. The constant c_R is usually assigned the value 0.36 to ensure that the energy spectrum obeys the classical Kolmogorov scaling argument in the limit of infinite Reynolds number (Andre & Lesieur 1977). The other two new coefficients, c_{1T} and c_{2T}, are empirical constants associated with scalar transfer. They cannot be determined theoretically, as $T^{\alpha}_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$ is not conservative, and thus cannot be constrained by some overall conservation principle. Following Herr *et al.* (1996), the constants c_{1T} and c_{2T} were given values of 0 and 1.0 respectively. By using these values for c_{1T} and c_{2T}, we can achieve consistency with their EDQNM model for the limit of equal molecular diffusivities. We note that c_{1M} and c_{2M} were again assigned the value 0.36 to ensure consistency of the proposed model with the earlier isotropic model in the limit of a vanishingly small scalar gradient.

Substituting the third-order moments into (4.9) and (4.11) yields closed expressions for $Q^{\alpha}(k,\mu)$ and $B^{\alpha\beta}(k,\mu)$. The only remaining problem is converting the implicit angle dependence into an explicit one. The method used by Herring (1974) involves a Legendre series expansion for the second-order moments, where the argument for the Legendre polynomial is the cosine of the angle between the mean gradient and $\boldsymbol{k}$, $\boldsymbol{p}$, or $\boldsymbol{q}$ (i.e. μ, μ', or μ'' respectively). For example, the expansions for $B^i(k,\mu)$ and $Q^j(k,\mu)$ take the following form (where i refers to α, β, or $\alpha\beta$, and j refers to α or β):

$$B^i(k,\mu) = \sum_{n=0}^{\infty} B^i_{2n}(k) L_{2n}(\mu),$$

$$Q^j(k,\mu) = \sum_{n=0}^{\infty} Q^j_{2n}(k) L_{2n}(\mu).$$

Similar expressions can be written for $B^i(p,\mu')$, $B^i(q,\mu'')$, $Q^j(p,\mu')$, and $Q^j(q,\mu'')$. Note that because $B^i(k,\mu)$ and $Q^j(k,\mu)$ are even functions of μ, only the even terms of the series are needed. Also, the first term in each infinite series represents the isotropic contribution to the correlation, while the higher-order terms account for the anisotropy due to the presence of the uniform mean gradients. Equations for the component spectra, $B^i_{2n}(k)$ and $Q^j_{2n}(k)$ can be derived from their respective transport equations by taking advantage of the orthogonality properties of the Legendre polynomials; to derive the transport equation for a specific order, multiply the appropriate governing equation ((4.9) or (4.11)) by the Legendre polynomial of that order and integrate over $\mathrm{d}\mu$ from -1 to 1.

It was shown in Herr *et al.* (1996) that if all correlations involving the scalar field are initialized to zero at $t = 0$, then the scalar–velocity cross-correlation will only involve the first term in the infinite series, while the scalar autocorrelation and covariance spectra will require the first two terms in the series. All higher-order terms will be identically zero for all time. As this study uses an identical initialization, the same arguments apply and we find that the separation of the wavenumber from the angle dependence takes the simple form of

$$\left.\begin{aligned} B^i(k,\mu) &= L_0(\mu)B^i_0(k) + L_2(\mu)B^i_2(k), \\ Q^j(k,\mu) &= L_0(\mu)Q^j_0(k). \end{aligned}\right\} \tag{4.18}$$

Note that $L_0(\mu) = 1$ and $L_2(\mu) = (3\mu^2 - 1)/2$.

Upon examination of (4.9) and (4.11), one will find that all the convolution integrals are of the form

$$\iint \hat{\delta}(\boldsymbol{k}+\boldsymbol{p}+\boldsymbol{q}) F(k,p,q,\mu,\mu',\mu'')\, \hat{\mathrm{d}}\boldsymbol{p}\, \hat{\mathrm{d}}\boldsymbol{q}, \tag{4.19}$$

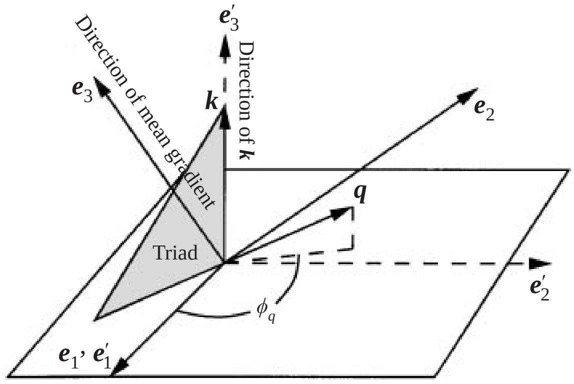

FIGURE 7. Coordinate system for evaluating the convolution integral: $(\boldsymbol{e}_1, \boldsymbol{e}_2, \boldsymbol{e}_3)$ is the natural coordinate system based on the direction of the mean gradient, while $(\boldsymbol{e}_1', \boldsymbol{e}_2', \boldsymbol{e}_3')$ is chosen so that $\boldsymbol{e}_3'$ is aligned with the $\boldsymbol{k}$ vector. The latter coordinate system is used to evaluate the convolution integrals because it simplifies the integrands.

and thus will vanish unless $\boldsymbol{k}$, $\boldsymbol{p}$, and $\boldsymbol{q}$ form a triangle (triad). It can be shown that by switching to a coordinate system with one axis aligned along the $\boldsymbol{k}$ vector of a triad (instead of along the mean gradient direction) μ' and μ'' can be expressed in terms of μ, the cosines of angles between the vectors of the triad, and the spherical angle $\phi_q (0 \leqslant \phi_q \leqslant 2\pi)$ (Nakauchi 1984)

$$\left.\begin{aligned} \mu &\equiv \frac{k_3}{k}, \\ \mu' &\equiv \frac{p_3}{p} = -\frac{\sqrt{1-\mu^2} N \sin\phi_q}{pk} + z\mu, \\ \mu'' &\equiv \frac{q_3}{q} = \frac{\sqrt{1-\mu^2} N \sin\phi_q}{qk} + y\mu. \end{aligned}\right\} \tag{4.20}$$

See figure 7 for a geometric sketch of the transformation. By using the law of cosines, the angles between the vectors of the triad can be expressed in terms of k, p, and q, and the vector integration over $\boldsymbol{p}$ and $\boldsymbol{q}$ can be replaced by a scalar integration over p and q as follows:

$$\frac{1}{(2\pi)^3} \iint_\Delta \left[\int_0^{2\pi} \hat{F}(k, p, q, \mu, \phi_q) \frac{pq}{k} \mathrm{d}\phi_q \right] \mathrm{d}p \, \mathrm{d}q. \tag{4.21}$$

Here, the integration over ϕ_q is done analytically, while the integration over p and q is done numerically.

5. Results and discussion for scalars with mean gradients

The closed equations for $Q_0^\alpha(k)$, $Q_0^\beta(k)$, $B_0^{\alpha\beta}(k)$, $B_0^\alpha(k)$, $B_0^\beta(k)$, $B_2^{\alpha\beta}(k)$, $B_2^\alpha(k)$ and $B_2^\beta(k)$ are solved on a uniform grid with 128 wavenumbers and non-dimensional $\Delta k = 1.59$. The forced energy spectrum is the same as was used in the isotropic scalar study, as are the values of the Schmidt numbers (i.e. case (i) corresponds to $Sc_\alpha = 1$, $Sc_\beta = 0.1$; case (ii) corresponds to $Sc_\alpha = 1$, $Sc_\beta = 0.01$). The definitions of the scalar spectra are

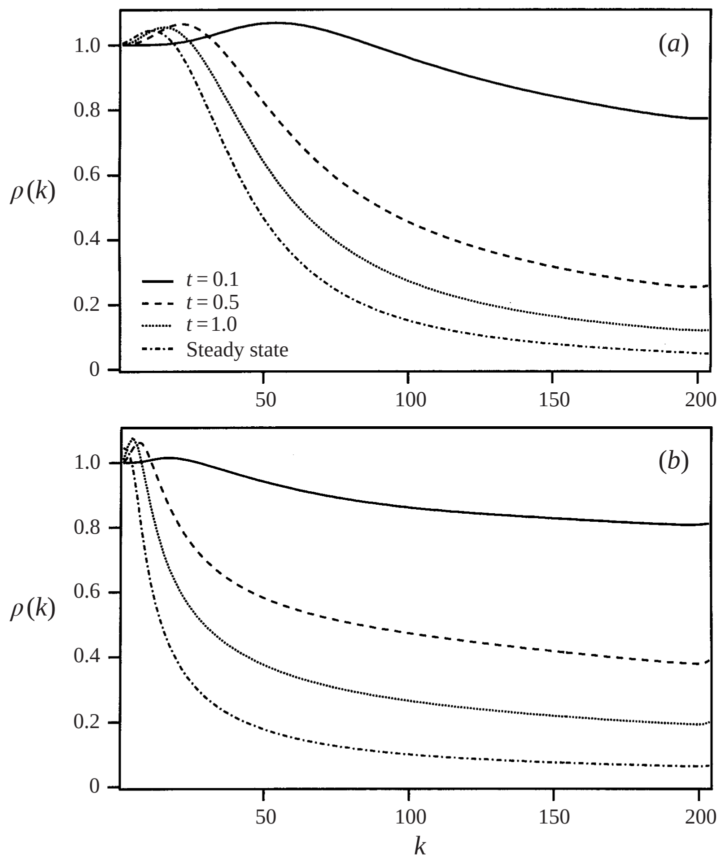

FIGURE 8. Evolution of the coherency spectrum for (*a*) case (i) and (*b*) case (ii). Once again, $\rho(k) > 1$ over a range of wavenumbers in violation of the Cauchy–Schwartz inequality. The violation occurs initially at high wavenumbers and moves to lower wavenumbers with increasing time.

now

$$E^i_\phi(k) = \frac{1}{(2\pi)^3}\iint B^i(\boldsymbol{k},\boldsymbol{p})\,\hat{\mathrm{d}}\boldsymbol{p}k^2\,\mathrm{d}\Omega_k = \frac{1}{2\pi^2}\int_{-1}^{1} k^2 B^i(k,\mu)\,\mathrm{d}\mu = \frac{k^2 B^i_0(k)}{\pi^2}, \qquad (5.1)$$

where the superscript i refers to α, β or $\alpha\beta$.

5.1. *Realizability*

Figure 8 shows the evolution of the coherency spectrum (see (3.1) for definition) for cases (i) and (ii). Once again, it is apparent that $\rho(k)$ exceeds unity over a range of wavenumbers. The phenomenon is similar to the one observed for the isotropic scalar; however, because of the complexity of the transfer terms for the axisymmetric scalar field it is now much more difficult to identify the origin of the partially unrealizable solution.

In the case of the isotropic scalar, it was possible to make progress by considering the Langevin equation for the scalar autocorrelation spectrum. However, such a representation for a scalar with a uniform mean gradient that is consistent with both the scalar–velocity (4.9) and scalar–scalar (4.11) equations is not as easily constructed. The EDQNM closure for $T^\alpha_{ij}(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$ produces interactions of the form RQ^α and RR while the closure for $M^{\alpha\beta}_i(\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})$ produces interactions of the form $RB^{\alpha\beta}$, $Q^\alpha Q^\beta$, RQ^α,

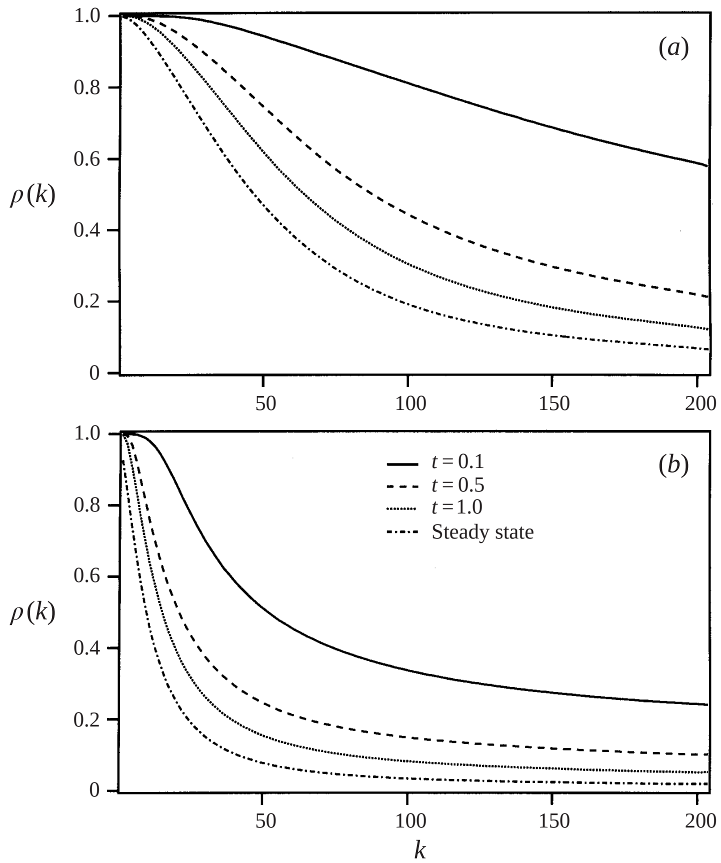

FIGURE 9. Evolution of the coherency spectrum for (*a*) case (i) and (*b*) case (ii) using the EDQNM model with the modified coefficients. The result is realizable and the transfer term properly conserves the scalar covariance.

RQ^{β} and RR. Given the velocity representation shown in (3.3), the equation for the scalar field must generate all of the interactions found in (4.9) and (4.11). The number and complexity of the interactions that must be generated by the scalar Langevin equation makes it prohibitively difficult to determine the appropriate coefficients for each term. Thus, the analysis for the scalar with a uniform mean gradient must be done without the benefit of the Langevin equation.

In the absence of a Langevin equation, we will utilize the conjecture discussed in § 3.3. The idea is to force the autocorrelation and covariance transfer spectra to be equal when the scalar fluctuations are perfectly correlated (i.e. when $E^{\alpha}(k,t) = E^{\beta}(k,t) = E^{\alpha\beta}(k,t)$ and $Q^{\alpha}(k,t) = Q^{\beta}(k,t)$). To accomplish this, the inverse time scales ${}^{\alpha}\mu_T^{kpq}$, ${}^{\beta}\mu_T^{kpq}$, and ${}^{\alpha\beta}\mu_M^{kpq}$ again must be independent of the scalar molecular diffusivities (or Péclet numbers Pe_{α} and Pe_{β}). One way to accomplish this is simply to replace Pe_{α} and Pe_{β} in these expressions by Re_L throughout.

Figure 9 shows the evolution of the covariance spectra for cases (i) and (ii) after substituting the modified eddy-damping coefficients. Notice that all spectra are realizable for all time. Despite the lack of an underlying Langevin model, the previously derived fix for the isotropic spectrum appears to work in the more general case of an anisotropic spectrum. This says nothing about the accuracy of the result, since realizability is only one criterion for a good model. We believe the model will continue to be accurate over the energy-containing eddies and the inertial range, since presumably the effect of molecular properties is small. At very large wavenumbers, there may be some effect of the modified inverse time scales. Finally, we emphasize that eliminating the diffusivity dependence from the inverse time scales does not eliminate the effect of the molecular diffusivity on the scalar spectra themselves. The latter effect (presumably the major effect) is explicitly accounted for by the diffusive terms in the scalar equations. Furthermore, as the triple correlations depend on the scalar spectra, they retain an implicit dependence on the scalar molecular diffusivities.

6. Conclusions

The EDQNM theory has been used to derive closed transport equations for the scalar covariance spectrum advected by isotropic turbulence. Two different cases were considered. In the first example, scalars ϕ_α and ϕ_β were generated by an isotropic forcing that was perfectly correlated. Thereafter, differences in the molecular diffusivities of the two scalars caused the scalars to decorrelate with time, an effect commonly known as differential diffusion. In this study, the focus was on developing a spectral model that was conservative and realizable, where 'realizable' here refers to a covariance spectrum that satisfies the Cauchy–Schwartz inequality.

We observed that the standard EDQNM model does not satisfy this more stringent realizability constraint. By considering the underlying Langevin equation for the scalar autocorrelations, it was possible to construct a spectral equation for the covariance spectrum that was fully realizable. However, this model did not properly conserve the transfer of the covariance spectrum without restricting the definitions of the inverse time scales. In particular, these time scales must be independent of the molecular diffusivities of the two scalars in order to produce a model that properly conserves the covariance spectrum and is realizable for all time. It is important to note that this restriction does not imply that the transfer terms are independent of the scalar diffusivities. Rather, they retain an implicit dependence through the scalar spectra, which in turn depend on the scalar diffusivities. Thus, it is believed that the present model can accurately describe the effects of differential diffusion in passive scalars.

Next, the EDQNM model for an axisymmetric scalar covariance spectrum was considered by introducing uniform mean gradients into the two scalars ϕ_α and ϕ_β. In this case, all spectra are shown to be functions of the wavenumber k and the cosine of the angle between the vector $\boldsymbol{k}$ and the direction of the mean gradient (here called μ). The straightforward application of the EDQNM procedure yields a covariance spectrum that likewise violates the Cauchy–Schwartz inequality. However, in this case it was not possible to construct a Langevin equation that reproduces all of the interactions found in the EDQNM model for the scalar–scalar and scalar–velocity spectra; thus, no formal approach to seeking a realizable spectrum exists. Instead, we applied the results obtained for the isotropic spectrum. By eliminating the dependence of the inverse time scales on the scalar molecular diffusivities, the model for the covariance spectrum was realizable for all times and combinations of the parameters considered.

The applicability of the result from the isotropic scalar study to the mean gradient case suggests that the strategy may have broader implications for Markovian models. For example, it was noted in the isotropic study that under circumstances where ϕ'_α and ϕ'_β are perfectly correlated, we expect the scalar transfer terms Tr^α, Tr^β and $Tr^{\alpha\beta}$ to be equal. The EDQNM model, with the original definitions for the inverse time scales, in general cannot satisfy this constraint because of the explicit dependence of the transfer functions on the molecular diffusivities. It now appears that this may cause the unrealizable covariance spectrum at early times.

The above explanation suggests that any model that introduces an explicit dependence of the triple correlations (transfer spectra) on the scalar diffusivities will likely suffer from the same problem. Models with more accurate evolution equations for the θ-coefficients may avoid this problem at the price of adding substantially to the cost of the calculation. Recall that the number of coefficients to be evaluated corresponds to the number of k–p–q combinations in the calculation, which is of the order of N^3 for N grid points. In the absence of very large computational resources, it may be necessary to modify the coefficients as suggested here to eliminate unrealizable results in the computationally tractable Markovian models.

In closing, we note that this is the first half of a two-part study of differential diffusion using spectral models. In a companion paper (Ulitsky, Vaithianathan & Collins 2000), the second half of the study will consider in greater depth the nature and rate of decorrelation of the two scalar spectra as well as investigate the role of local vs. non-local triadic interactions in scalar transfer. Additionally, comparisons of the proposed EDQNM model with DNS will be presented.

The authors gratefully acknowledge Dr Timothy Clark for suggesting the Langevin equation approach as an alternative to the moment expansion. We also wish to thank the reviewers for several insightful comments on the Langevin formulation. This work was supported by the United State Department of Energy Accelerated Strategic Computing Initiative (ASCI) program and Los Alamos National Laboratory, Group T-3. Partial support for one of the authors (L. R. C.) was provided by the National Science Foundation CTS-9732223.

Appendix. Relationship between the present notation for the transfer spectrum and the classical notation in Lesieur (1987)

In this Appendix, we show how (2.18) is equivalent to the equation for the transfer spectrum given in Lesieur (1987). The EDQNM form for $Tr^{\alpha}(k)$ in Lesieur (1987) is given by

$$\iint_{\Delta} \frac{k}{pq}(1-y^2)E(q)\left[k^2E_T(p,t)-p^2E_T(k,t)\right]\theta^T_{kpq}\,\mathrm{d}p\,\mathrm{d}q. \tag{A 1}$$

Here $E_T(\ )$ is equivalent in our notation to $E^{\alpha}_{\phi}(\ )$ and θ^T_{kpq} is equivalent to $\theta\left({}^{\alpha}\mu^{qkp}_{M}\right)$. If we define a vector $\boldsymbol{N}$ to be normal to the plane formed by $\boldsymbol{k}$, $\boldsymbol{p}$, and $\boldsymbol{q}$, then $\boldsymbol{N}$ can be calculated by

$$\boldsymbol{N}=\boldsymbol{k}\times\boldsymbol{p}=\boldsymbol{p}\times\boldsymbol{q}=\boldsymbol{q}\times\boldsymbol{k}. \tag{A 2}$$

Thus, $N\equiv|\boldsymbol{N}|$ is twice the area of the triangle formed by k, p and q and N^2 takes the value of

$$N^2=k^2p^2(1-z^2)=p^2q^2(1-x^2)=k^2q^2(1-y^2). \tag{A 3}$$

Note that we can also use Heron's formula to express N^2 in terms of k, p, and q as was done in (2.24).

Switching the dummy variables of integration in (2.18) gives the following equivalent expression for $Tr^\alpha(k)$:

$$Tr^\alpha(k,t) = \iint_\Delta \left[g_1(k,q,p)E(q)E^\alpha_\phi(p,t) - g_2(k,q,p)E(q)E^\alpha_\phi(k,t)\right] \theta\left({}^\alpha\mu_M^{qkp}\right) \mathrm{d}p\,\mathrm{d}q. \tag{A 4}$$

If we then substitute $N^2 = k^2q^2(1-y^2)$ into the definitions of $g_1(k,q,p)$ and $g_2(k,q,p)$, we obtain

$$\iint_\Delta \frac{k}{pq}(1-y^2)E(q)\left[k^2E^\alpha_\phi(p,t) - p^2E^\alpha_\phi(k,t)\right]\theta({}^\alpha\mu_M^{qkp})\,\mathrm{d}p\,\mathrm{d}q. \tag{A 5}$$

Thus, the two equations for Tr^α are seen to be equivalent. It should be noted that our definitions of x, y, and z differ from those given in Lesieur (1987). For example, we chose to define y as the cosine of the angle between the $\boldsymbol{k}$ and $\boldsymbol{q}$ vectors. Lesieur, however, defines y as the cosine of the angle in the interior of the triad whose adjacent sides are k and q. Therefore, the two definitions of y differ by a minus sign, as the angles are separated by π radians. The same applies for x and z. In (A 1) or (A 5), this difference in definitions is irrelevant since y is raised to an even power.

REFERENCES

Andre, J. C. & Lesieur, M. 1977 Influence of helicity on the evolution of isotropic turbulence at high Reynolds number. *J. Fluid Mech.* **81**, 187.

Batchelor, G. K. 1946 The theory of axisymmetric turbulence. *Proc. R. Soc. Lond.* A **186**, 480.

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

Bilger, R. W. & Dibble, R. W. 1982 Differential molecular diffusion effects in turbulent mixing. *Combust. Sci. Tech.* **28**, 161.

Chandrasekhar, S. 1950 The decay of axisymmetric turbulence. *Phil. Trans. R. Soc. Lond.* A **242**, 557.

Chen, J.-Y., Bilger, R. W. & Dibble, R. W. 1990 Pdf modeling of nonpremixed $CO/H_2/N_2$ jet flames with reduced mechanisims. In *Twenty-Third Symp. (Intl) on Combustion, Pittsburgh*, p. 775. The Combustion Institute.

Chou, P. Y. 1940 On an extension of Reynolds' method of finding apparent stress and the nature of turbulence. *Chinese J. Phys.* **4**, 1.

Drake, M. C., Pitz, R. W. & Lapp, M. 1986 Laser measurements on nonpremixed H_2-air flames for assessments of turbulent combustion models. *AIAA J.* **24**, 905.

Eswaran, V. & Pope, S. B. 1988 An examination of forcing in direct numerical simulations of turbulence. *Comput. Fluids* **16**, 257.

Golub, G. H. & Loan, C. F. V. 1983 *Matrix Computations*. Johns Hopkins Press.

Herr, S., Wang, L.-P. & Collins, L. R. 1996 EDQNM model of a passive scalar with a uniform mean gradient. *Phys. Fluids* **8**, 1588.

Herring, J. R. 1974 Approach of axisymmetric turbulence to isotropy. *Phys. Fluids* **17**, 859.

Herring, J. R., Schertzer, D., Lesieur, M., Newman, G. R., Chollet, J. P. & Larcheveque, M. 1982 A comparative assessment of spectral closures as applied to passive scalar diffusion. *J. Fluid Mech.* **124**, 411.

Hinze, J. O. 1987 *Turbulence*. McGraw-Hill.

Kerstein, A. R., Dibble, R. W., Long, M. B., Yip, B. & Lyons, K. 1989 Measurement and computation of differential molecular diffusion in a turbulent jet. In *Seventh Symp. on Turbulent Shear Flows*, Paper 14-2.

Kraichnan, R. H. 1991 Stochastic modeling of isotropic turbulence. In *New Perspectives in Turbulence*, pp. 1–54. Springer.

Lesieur, M. 1987 *Turbulence in Fluids*. Nijhoff.

Leslie, D. C. 1973 *Developments in the Theory of Turbulence*. Clarendon.

Millionshtchikov, M. 1941 On the theory of homogeneous isotropic turbulence. *Dokl. Akad. Nauk. SSSR* **32**, 615.

NAKAUCHI, N. 1984 An application of the modified zero-fourth-cumulant approximation to homogeneous axisymmetric turbulance. *J. Phys. Soc. Japan* **53**, 1682.

NAKAUCHI, N., OSHIMA, H. & SAITO, Y. 1989 A passive scalar convected by homogeneous axisymmetric turbulence. *Phys. Fluids* A **1**, 723.

ORSZAG, S. A. 1970 Analytical theories of turbulence. *J. Fluid Mech.* **41**, 363.

ORSZAG, S. A. 1973 Lectures on the Statistical Theory of Turbulence. In *Fluid Dynamics 1973, Les Houches Summer School of Theoretical Physics*, pp. 235–374. Gordon and Breach.

POUQUET, A., LESIEUR, M., ANDRE, J. C. & BASDEVANT, C. 1975 Evolution of high Reynolds number two-dimensional turbulence. *J. Fluid Mech.* **72**, 305.

TATSUMI, T. 1980 Theory of homogeneous turbulence. *Adv. Appl. Mech.* **20**, 39.

ULITSKY, M. & COLLINS, L. R. 1997 Application of the eddy-damped quasi-normal Markovian spectral transport theory to premixed turbulent flame propagation. *Phys. Fluids* **9**, 3410–3430.

ULITSKY, M., VAITHIANATHAN, T. & COLLINS, L. R. 2000 A spectral study of differential diffusion with and without uniform mean scalar gradients. *J. Fluid Mech.* in preparation.

VRANOS, A., KNIGHT, B. A., PROSCIA, W. M., CHIAPPETTA, L. & SMOOKE, M. D. 1992 Nitric oxide formation and differential diffusion in a turbulent methane-hydrogen diffusion flame. In *Twenty-Fourth Symp. (Intl) on Combustion, Pittsburgh*, p. 377. The Combustion Institute.

WILLIAMS, F. A. 1985 *Combustion Theory*, 2nd Edn. Addison-Wesley.

YEUNG, P. K. 1996 Multi-scalar triadic interactions in differential diffusion with and without mean scalar gradients. *J. Fluid Mech.* **321**, 235.

YEUNG, P. K. & POPE, S. B. 1993 Differential diffusion of passive scalars in isotropic turbulence. *Phys. Fluids* A **5**, 2467.