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## Passive scalar spectrum in isotropic turbulence: Prediction by the Lagrangian direct-interaction approximation

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A closed set of equations for the passive scalar correlation function in isotropic turbulence is formulated by the Lagrangian direct-interaction approximation developed previously by the present authors [Kida and Goto, J. Fluid Mech. 345, 307 (1997)]. We examine the behavior of solutions to the resultant equations for arbitrary values of the Schmidt number, and show systematically that this closure theory is completely consistent with the phenomenological theories on the scalar spectrum function by Obukhov (1949), Corrsin (1951), Batchelor et al. (1959) and Batchelor (1959). The universal form of the function in the statistically stationary state is obtained by solving the closure equations numerically in the whole wavenumber range for each case of moderate, extremely large and small values of the Schmidt number.

KEYWORDS: Turbulence, Passive scalar, Direct-interaction approximation

#### I. INTRODUCTION

One of the fundamental challenges in turbulence research is the prediction of the statistical properties of field quantities on the bases of the first principle, i.e. the equations of fluid motion. The so-called closure theories of turbulence, which concern the lower-order statistics such as the first and second order moments of velocity, have been developed under various approximations by many authors over fifty years, which includes the EDQNM (eddy damped quasi-normal Markovian) by Orszag [1], a series of DIA (direct-interaction approximation) by Kraichnan [2,3], the LET (local-energy-transfer) theory by McComb et al. [4], the LRA (Lagrangian renormalized approximation) by Kaneda [5] as well as the Lagrangian DIA by ourselves [6]. (The last two theories yield an identical set of equations though their working assumptions are different in an important respect [7].) All of these theories are successful to describe the energy spectrum function reasonably well; they are consistent with the -5/3 power law in the inertial range. It turned out that the Lagrangian DIA (and LRA) in particular yields an excellent prediction of the energy spectrum function in the whole universal range of wavenumber for stationary isotropic turbulence as well as for decaying one, while it has no adjustable free parameters and it is much simpler than Kraichnan's LHDIA (Lagrangian history DIA) both in the procedure and in the final form.

It should be interesting therefore to apply this Lagrangian DIA to a passive scalar field, such as temperature, contaminant, particle concentration, dye, smoke, etc, which is advected and mixed in isotropic turbulence. The statistics of a scalar field is quite different depending upon the ratio s of the kinematic viscosity  $\nu$  of a fluid and the diffusion coefficient  $\kappa$  of a passive scalar. Here, we name s the Schmidt number (or the Prandtl number on speaking of temperature). The behavior of the scalar power spectrum  $\Theta(k)$ , where k stands for the wavenumber, has been derived by phenomenological arguments (see Tennekes & Lumley [8], Lesieur [9]). It depends upon the relative magnitude of several characteristic wavenumbers, which include the peak wavenumbers,  $k_{\rm v}$  and  $k_{\rm s}$ , of the energy and scalar power spectra, the Kolmogorov wavenumber,

$$k_{\rm K} = (\epsilon/\nu^3)^{1/4},$$
 (1.1)

at which the inertial-range turbulent diffusion time  $(k^{2/3}\epsilon^{1/3})^{-1}$  and the viscous dissipation time  $(\nu k^2)^{-1}$  are comparable, the Obukhov-Corrsin wavenumber,

$$k_{\rm C} = (\epsilon/\kappa^3)^{1/4} \ (= s^{3/4} k_{\rm K}),$$
 (1.2)

at which the inertial-range turbulent diffusion time and the scalar dissipation time  $(\kappa k^2)^{-1}$  are comparable (meaningful only for s < 1 when  $k_c$  lies in the inertial range), and the Batchelor wavenumber,

$$k_{\rm B} = (\epsilon/\nu\kappa^2)^{1/4} \ (= s^{1/2} k_{\rm K} = s^{-1/4} k_{\rm C}),$$
 (1.3)

at which the scalar dissipation time and the shearing time  $(\nu/\epsilon)^{1/2}$  of vortices of the Kolmogorov scale are comparable (meaningful only for s > 1 when  $k_B$  lies in the viscous dissipative range). Here,  $\epsilon$  is the energy dissipation rate.

Three kinds of power laws are known: (i) the inertial-advective (inertial-convective) range, in which

$$\Theta(k) = C_1 \chi \epsilon^{-1/3} k^{-5/3} \qquad (\max\{k_{\rm v}, k_{\rm s}\} \ll k \ll \min\{k_{\rm K}, k_{\rm c}\}), \tag{1.4}$$

and neither the molecular viscosity nor the scalar dissipation is effective [10,11], (ii) the viscous-advective (viscous-convective) range, in which

$$\Theta(k) = C_2 \chi \nu^{1/2} \epsilon^{-1/2} k^{-1} \qquad (k_{\rm K} \ll k \ll k_{\rm B} \ s \gg 1), \tag{1.5}$$

and the scalar field is deformed by the shearing motion induced by vortices of the Kolmogorov scale [12], and (iii) the inertial-diffusive (inertial-conductive) range, in which

$$\Theta(k) = C_3 \chi \kappa^{-3} \epsilon^{2/3} k^{-17/3} \qquad (k_{\rm C} \ll k \ll k_{\rm K}, \ s \ll 1), \tag{1.6}$$

and the passive scalar is rapidly diffusing while being mixed by turbulence [13]. Here,  $\chi$  denotes the scalar dissipation rates. These three power laws have been observed either in the real turbulence [14–16] or in the numerically simulated turbulence [17]. They are also consistent with the abridged LHDIA [18].

In this paper we formulate the Lagrangian DIA for a passive scalar field advected in isotropic turbulence, and solve the resultant equations analytically (in asymptotic limits) and numerically. We will not only confirm all of the above phenomenological results but also present the following three new findings. First, we derive all of the three power laws systematically, and determine the boundaries of the respective power regions by an asymptotic analysis at large or small values of the wavenumber and of the Schmidt number. Second, we determine the universal functional forms of the scalar power spectrum with high accuracy by solving the stationary equations. (An estimation of the functional form from a late state in a decaying numerical simulation of the closure equations, as was done in refs. [19,20], should not be so accurate. It is not easy to know the time when it has approached the universal state and there is no guarantee that the functional forms of the spectrum in the decaying and stationary turbulence ever coincide with each other. For example, in Fig.12 of [19] or in Fig.13 of [20] we hardly observe the universality of the constants even when an identical closure equation is solved.) Third, we describe the behavior of the scalar power spectrum in the whole universal range of wavenumber and for all ranges of the Schmidt number.

This paper is organized as follows. Based upon the basic equations described in Sec. II, we formulate in Sec. III the Lagrangian DIA for a passive scalar field to derive an integro-differential equation for the correlation function. We make in Sec. IV a detailed analysis of the resultant closure equation to find the universal form of the passive scalar spectrum. Three kinds of scaling laws (1.4)—(1.5) are shown to be consistent with the closure equation and all the universal constants are evaluated. In addition, the universal form of the function is determined numerically for several finite values of s as well as for  $s \gg 1$  and  $s \ll 1$ . Section V is devoted to a summary and concluding remarks.

### II. BASIC EQUATIONS

We consider the statistical behavior of a passive scalar field  $\theta(x,t)$  which obeys the advection-diffusion equation

$$\frac{\partial}{\partial t} \theta(\mathbf{x}, t) + u_i(\mathbf{x}, t) \frac{\partial}{\partial x_i} \theta(\mathbf{x}, t) = \kappa \frac{\partial^2}{\partial x_i \partial x_i} \theta(\mathbf{x}, t) , \qquad (2.1)$$

where  $u_i(\boldsymbol{x},t)$  is an incompressible turbulent velocity field governed by the Navier-Stokes equation

$$\frac{\partial}{\partial t} u_i(\boldsymbol{x}, t) + u_j(\boldsymbol{x}, t) \frac{\partial}{\partial x_j} u_i(\boldsymbol{x}, t) = -\frac{1}{\rho} \frac{\partial}{\partial x_i} p(\boldsymbol{x}, t) + \nu \frac{\partial^2}{\partial x_j \partial x_j} u_i(\boldsymbol{x}, t) \quad (i = 1, 2, 3)$$
(2.2)

and the equation of continuity

$$\frac{\partial}{\partial x_i} u_i(\boldsymbol{x}, t) = 0 \ . \tag{2.3}$$

In the Lagrangian closure formulation, it is convenient to introduce the Lagrangian position function [5] defined by

$$\psi(\mathbf{x}, t | \mathbf{x}', t') = \delta^3(\mathbf{x} - \mathbf{y}(t | \mathbf{x}', t')) , \qquad (2.4)$$

where y(t|x',t') is the position at time t of a fluid element which passed x' at  $t' \leq t$ . The position function  $\psi(x,t|x',t')$  describes the probability density that a fluid element which passed x' at t' will pass x at t, and it is governed by

$$\frac{\partial}{\partial t} \psi(\mathbf{x}, t | \mathbf{x}', t') = -u_i(\mathbf{x}, t) \frac{\partial}{\partial x_i} \psi(\mathbf{x}, t | \mathbf{x}', t')$$
(2.5)

with the initial condition

$$\psi(\boldsymbol{x}, t'|\boldsymbol{x}', t') = \delta^{3}(\boldsymbol{x} - \boldsymbol{x}') . \tag{2.6}$$

The Lagrangian velocity and scalar fields are written in terms of the respective Eulerian counterparts and the position function, as

$$v_i(t|\mathbf{x}',t') = \int d^3\mathbf{x} \ u_i(\mathbf{x},t) \ \psi(\mathbf{x},t|\mathbf{x}',t')$$
(2.7)

and

$$\theta^{(L)}(t|\boldsymbol{x},t') = \int d^3\boldsymbol{x}' \; \psi(\boldsymbol{x}',t|\boldsymbol{x},t') \; \theta(\boldsymbol{x}',t) \; . \tag{2.8}$$

The Lagrangian correlation functions for the velocity and scalar fields are then defined respectively by

$$V_{ij}(\mathbf{r},t,t') = \overline{v_i(t|\mathbf{x}+\mathbf{r},t') u_j(\mathbf{x},t')}$$
(2.9)

and

$$Z(\mathbf{r},t,t') = \overline{\theta^{(L)}(t|\mathbf{x}+\mathbf{r},t')} \overline{\theta(\mathbf{x}',t')}, \qquad (2.10)$$

where the homogeneity of the fields has been assumed, and an overbar denotes an ensemble average.

In order to make the following analysis easier, we assume that the fluid is confined in a periodic cube of side L, and at the final stage of calculation we will take the limit  $L \to \infty$ . We can then decompose the Eulerian velocity field into the Fourier series as

$$u_i(\boldsymbol{x},t) = \left(\frac{2\pi}{L}\right)^3 \sum_{\boldsymbol{k}} \tilde{u}_i(\boldsymbol{k},t) \exp\left[i\boldsymbol{k}\cdot\boldsymbol{x}\right], \qquad (2.11)$$

where  $\mathbf{k} = (2\pi/L)(n_1, n_2, n_3)$ ,  $(n_1, n_2, n_3 = 0, \pm 1, \pm 2, \cdots)$  is the wavenumber vector. Other Eulerian and Lagrangian fields are decomposed in the same manner. Then, (2.1)—(2.3), (2.5) and (2.6) are respectively written as

$$\left[\frac{\partial}{\partial t} + \kappa k^2\right] \widetilde{\theta}(\mathbf{k}, t) = -i \, k_j \, \left(\frac{2\pi}{L}\right)^3 \sum_{\substack{\mathbf{p} \\ (\mathbf{k} + \mathbf{p} + \mathbf{q} = \mathbf{o})}} \widetilde{u}_j(-\mathbf{p}, t) \, \widetilde{\theta}(-\mathbf{q}, t) , \qquad (2.12)$$

$$\left[\frac{\partial}{\partial t} + \nu k^2\right] \widetilde{u}_i(\mathbf{k}, t) = -\frac{\mathrm{i}}{2} \left(\frac{2\pi}{L}\right)^3 \widetilde{P}_{ijm}(\mathbf{k}) \sum_{\substack{\mathbf{p} \ (\mathbf{k}+\mathbf{p}+\mathbf{q}=\mathbf{o})}} \widetilde{u}_j(-\mathbf{p}, t) \widetilde{u}_m(-\mathbf{q}, t) , \qquad (2.13)$$

$$k_i \, \widetilde{u}_i(\mathbf{k}, t) = 0 \,, \tag{2.14}$$

$$\frac{\partial}{\partial t} \widetilde{\psi}(\mathbf{k}, t | \mathbf{k}', t') = -i k_j \left( \frac{2\pi}{L} \right)^3 \sum_{\substack{\mathbf{p} \\ (\mathbf{k} + \mathbf{p} + \mathbf{q} = \mathbf{o})}} \widetilde{u}_j(-\mathbf{p}, t) \widetilde{\psi}(-\mathbf{q}, t | \mathbf{k}', t')$$
(2.15)

and

$$\widetilde{\psi}(\mathbf{k}, t'|\mathbf{k}', t') = \frac{L^3}{(2\pi)^6} \, \delta^3_{\mathbf{k}+\mathbf{k}'} \,, \tag{2.16}$$

where  $\tilde{P}_{ijm}(\mathbf{k}) = k_m \tilde{P}_{ij}(\mathbf{k}) + k_j \tilde{P}_{im}(\mathbf{k})$  and  $\tilde{P}_{ij}(\mathbf{k}) = \delta_{ij} - k_i k_j / k^2$ . The relations (2.8) and (2.7) between the Lagrangian and Eulerian fields are written as

$$\widetilde{v}_{i}(t|\boldsymbol{k},t') = \frac{(2\pi)^{6}}{L^{3}} \sum_{\boldsymbol{k}'} \widetilde{u}_{i}(\boldsymbol{k}',t) \, \widetilde{\psi}(-\boldsymbol{k}',t|\boldsymbol{k},t')$$
(2.17)

and

$$\widetilde{\theta}^{(L)}(t|\mathbf{k},t') = \frac{(2\pi)^6}{L^3} \sum_{\mathbf{k'}} \widetilde{\theta}(\mathbf{k'},t) \, \widetilde{\psi}(-\mathbf{k'},t|\mathbf{k},t') . \qquad (2.18)$$

The time derivatives of (2.17) and (2.18) yield respectively the governing equations for the Fourier transforms of the Lagrangian fields as

$$\frac{\partial}{\partial t} \widetilde{v}_{i}(t|\mathbf{k}',t') = -\frac{(2\pi)^{6}}{L^{3}} \nu \sum_{\mathbf{p}} p^{2} \widetilde{u}_{i}(\mathbf{p},t) \widetilde{\psi}(-\mathbf{p},t|\mathbf{k}',t')$$

$$-i \frac{(2\pi)^{9}}{L^{6}} \sum_{\mathbf{p}} \sum_{\mathbf{q}} \sum_{\mathbf{r}} \frac{r_{i}r_{m}r_{n}}{r^{2}} \widetilde{u}_{m}(\mathbf{p},t) \widetilde{u}_{n}(\mathbf{q},t) \widetilde{\psi}(\mathbf{r},t|\mathbf{k}',t')$$
(2.19)

and

$$\left[\frac{\partial}{\partial t} + \kappa k^2\right] \widetilde{\theta}^{(L)}(t|\mathbf{k}, t') = 0.$$
 (2.20)

The evolution equations for the Fourier transform of the Lagrangian scalar correlation function

$$\widetilde{Z}(\boldsymbol{k}, t, t') = \left(\frac{1}{2\pi}\right)^{3} \int d^{3}\boldsymbol{r} \, Z(\boldsymbol{r}, t, t') \, \exp\left[-i\boldsymbol{k} \cdot \boldsymbol{r}\right] = \left(\frac{2\pi}{L}\right)^{3} \, \overline{\widetilde{\theta}^{(L)}(t|\boldsymbol{k}, t')} \, \widetilde{\theta}(-\boldsymbol{k}, t')$$
(2.21)

are derived, from (2.12) and (2.20), for the single time correlation as

$$\left[\frac{\partial}{\partial t} + 2\kappa k^{2}\right] \widetilde{Z}(\mathbf{k}, t, t) = -i k_{\jmath} \left(\frac{2\pi}{L}\right)^{6} \sum_{\substack{\mathbf{p} \\ (\mathbf{k} + \mathbf{p} + \mathbf{q} = \mathbf{o})}} \overline{\widetilde{u}_{\jmath}(-\mathbf{p}, t) \, \widetilde{\theta}(-\mathbf{q}, t) \, \widetilde{\theta}(-\mathbf{k}, t)} + (\mathbf{k} \to -\mathbf{k}) \tag{2.22}$$

and for the two-time correlation as

$$\left[\frac{\partial}{\partial t} + \kappa k^2\right] \widetilde{Z}(\mathbf{k}, t, t') = 0.$$
 (2.23)

The evolution equations for the Fourier transform of the Lagrangian velocity correlation function

$$\widetilde{V}_{ij}(\mathbf{k},t,t') = \left(\frac{1}{2\pi}\right)^3 \int d^3\mathbf{r} \, V_{ij}(\mathbf{r},t,t') \, \exp\left[-i\mathbf{k}\cdot\mathbf{r}\right] = \left(\frac{2\pi}{L}\right)^3 \, \overline{\widetilde{v}_i(t|\mathbf{k},t')} \, \widetilde{u}_j(-\mathbf{k},t')$$
(2.24)

are derived similarly from (2.13) and (2.19) (see eqs. (2.26) and (2.27) in ref. [6]).

For a later use, we introduce here the incompressible part of the Lagrangian velocity correlation function

$$\widetilde{Q}_{ij}(\mathbf{k},t,t') = \widetilde{P}_{im}(\mathbf{k}) \, \widetilde{V}_{mj}(\mathbf{k},t,t') , \qquad (2.25)$$

which has already been determined in the Lagrangian DIA for the velocity field [6], and the passive scalar power spectrum function

$$\Theta(k,t) = k^2 \oint d\Omega \, \widetilde{Z}(k,t,t) , \qquad (2.26)$$

which will be considered in detail in the following sections. Here,  $\oint d\Omega$  denotes a solid angle integration in the Fourier space. The scalar transfer function T(k) is defined by a solid angle integration of the right-hand side of (2.22) as

$$\left[\frac{\partial}{\partial t} + 2\kappa k^2\right] \Theta(k, t) = T(k) , \qquad (2.27)$$

and the flux function  $\Pi(k)$  is defined by

$$\Pi(k) = \int_{k}^{\infty} \mathrm{d}k' \, T(k') \ . \tag{2.28}$$

#### III. LAGRANGIAN DIA FOR A PASSIVE SCALAR FIELD

In this section, a closed equation for the Lagrangian correlation function of the passive scalar field is derived by DIA. We define the response functions of  $\tilde{\theta}(k,t)$  and  $\tilde{\theta}^{(L)}(t|k,t')$  by

$$\widetilde{G}(\mathbf{k}, t | \mathbf{k}', t') = \frac{\delta \widetilde{\theta}(\mathbf{k}, t)}{\delta \widetilde{\theta}(\mathbf{k}', t')}$$
(3.1)

and

$$\widetilde{G}^{(L)}(t|\mathbf{k},\mathbf{k}',t') = \frac{\delta\widetilde{\theta}^{(L)}(t|\mathbf{k},t')}{\delta\widetilde{\theta}(\mathbf{k}',t')},$$
(3.2)

respectively, where  $\delta$  denotes a functional derivative. The evolution equations for these response functions are respectively derived by taking functional derivatives of (2.12) and (2.20) as

$$\left[\frac{\partial}{\partial t} + \kappa k^2\right] \widetilde{G}(\mathbf{k}, t | \mathbf{k}', t') = -i k_j \left(\frac{2\pi}{L}\right)^3 \sum_{\substack{\mathbf{p} \\ (\mathbf{k} + \mathbf{p} + \mathbf{q} = \mathbf{o})}} \widetilde{u}_j(-\mathbf{p}, t) \widetilde{G}(-\mathbf{q}, t | \mathbf{k}', t')$$
(3.3)

and

$$\left[\frac{\partial}{\partial t} + \kappa k^2\right] \widetilde{G}^{(L)}(t|\mathbf{k}, \mathbf{k}', t') = 0.$$
(3.4)

The initial conditions are given by

$$\widetilde{G}(\mathbf{k}, t' | \mathbf{k}', t') = \widetilde{G}^{(L)}(t' | \mathbf{k}, \mathbf{k}', t') = \frac{L^3}{(2\pi)^6} \, \delta_{\mathbf{k} + \mathbf{k}'}^3 \,. \tag{3.5}$$

#### A. Direct-interaction decomposition

The DIA is based upon the direct-interaction decomposition [2], in which  $\widetilde{\theta}$  and  $\widetilde{G}$  are respectively written as

$$\widetilde{\theta}(k,t) = \widetilde{\theta}^{(0)}(k,t||t_0,k_0,p_0,q_0) + \widetilde{\theta}^{(1)}(k,t||t_0,k_0,p_0,q_0)$$
(3.6)

and

$$\widetilde{G}(\mathbf{k}, t) = \widetilde{G}^{(0)}(\mathbf{k}, t | \mathbf{k}', t' | \mathbf{k}_0, \mathbf{p}_0, \mathbf{q}_0) + \widetilde{G}^{(1)}(\mathbf{k}, t | \mathbf{k}', t' | \mathbf{k}_0, \mathbf{p}_0, \mathbf{q}_0) .$$
(3.7)

Here,  $k_0$ ,  $p_0$  and  $q_0$  ( $k_0 + p_0 + q_0 = o$ ) are a triplet of wavenumbers, the direct interaction between which has been removed in the non-direct-interaction (NDI) fields designated by superscript  $^{(0)}$ . (We call  $\widetilde{\theta}^{(1)}$  and  $\widetilde{G}^{(1)}$  the direct-interaction (DI) fields.) These decompositions for  $\widetilde{\theta}$  and  $\widetilde{G}$  are made after  $t_0$  and t', respectively. Hereafter, the argument  $t_0$  in  $\widetilde{\theta}^{(0)}$  and  $\widetilde{\theta}^{(1)}$  will be omitted for simplicity of notation. The initial conditions for  $\widetilde{\theta}^{(0)}$ ,  $\widetilde{\theta}^{(1)}$ ,  $\widetilde{G}^{(0)}$  and  $\widetilde{G}^{(1)}$  are given by

$$\widetilde{\theta}^{(0)}(\mathbf{k}, t_0 || \mathbf{k}_0, \mathbf{p}_0, \mathbf{q}_0) = \widetilde{\theta}(\mathbf{k}, t_0) , \qquad \widetilde{\theta}^{(1)}(\mathbf{k}, t_0 || \mathbf{k}_0, \mathbf{p}_0, \mathbf{q}_0) = 0 ,$$
 (3.8)

$$\widetilde{G}^{(0)}(\mathbf{k}, t'|\mathbf{k}', t'|\mathbf{k}_0, \mathbf{p}_0, \mathbf{q}_0) = \frac{L^3}{(2\pi)^6} \, \delta_{\mathbf{k}+\mathbf{k}'}^3 \quad \text{and} \quad \widetilde{G}^{(1)}(\mathbf{k}, t'|\mathbf{k}', t'|\mathbf{k}_0, \mathbf{p}_0, \mathbf{q}_0) = 0 \,\,, \tag{3.9}$$

respectively. The decompositions for  $\tilde{u}_i$  and  $\tilde{\psi}$  are similarly performed (see (3.1)—(3.4), (A2), (A8)—(A11) and (B2) in ref. [6]). Since the NDI fields  $\tilde{\theta}^{(0)}$  and  $\tilde{G}^{(0)}$  are fictitious fields in which there is no direct interaction between the particular modes of wavenumbers  $k_0$ ,  $p_0$  and  $q_0$ , they are respectively governed by

$$\left[\frac{\partial}{\partial t} + \kappa k^{2}\right] \widetilde{\theta}^{(0)}(\boldsymbol{k}, t || \boldsymbol{k}_{0}, \boldsymbol{p}_{0}, \boldsymbol{q}_{0}) = -i k_{j} \left(\frac{2\pi}{L}\right)^{3} \sum_{\boldsymbol{p}} \sum_{\boldsymbol{q}} \widetilde{u}_{j}(-\boldsymbol{p}, t) \widetilde{\theta}^{(0)}(-\boldsymbol{q}, t || \boldsymbol{k}_{0}, \boldsymbol{p}_{0}, \boldsymbol{q}_{0}) \tag{3.10}$$

and

$$\left[\frac{\partial}{\partial t} + \kappa k^2\right] \widetilde{G}^{(0)}(\boldsymbol{k}, t | \boldsymbol{k}', t' | \boldsymbol{k}_0, \boldsymbol{p}_0, \boldsymbol{q}_0) = -i k_j \left(\frac{2\pi}{L}\right)^3 \sum_{\substack{\boldsymbol{p} \ (\boldsymbol{k} + \boldsymbol{p} + \boldsymbol{q} = \boldsymbol{o})}} \widetilde{G}^{(0)}(-\boldsymbol{q}, t | \boldsymbol{k}', t' | \boldsymbol{k}_0, \boldsymbol{p}_0, \boldsymbol{q}_0), \quad (3.11)$$

where  $\sum \sum'$  denotes the summation without the direct interactions between the three modes of wavenumbers  $k_0$ ,  $p_0$  and  $q_0$ . The evolution equation for the DI field  $\tilde{\theta}^{(1)}$  is then obtained by subtracting (3.10) from (2.12) as

$$\left[\frac{\partial}{\partial t} + \kappa k^{2}\right] \widetilde{\theta}^{(1)}(\mathbf{k}, t || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) = -i k_{j} \left(\frac{2\pi}{L}\right)^{3} \sum_{\mathbf{p}} \sum_{\mathbf{q}} '\widetilde{u}_{j}(-\mathbf{p}, t) \, \widetilde{\theta}^{(1)}(-\mathbf{q}, t || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) 
-i \delta_{\mathbf{k} - \mathbf{k}_{0}}^{3} k_{0j} \, \widetilde{u}_{j}(-\mathbf{p}_{0}, t) \, \widetilde{\theta}^{(0)}(-\mathbf{q}_{0}, t || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) 
-i \delta_{\mathbf{k} - \mathbf{k}_{0}}^{3} k_{0j} \, \widetilde{u}_{j}(-\mathbf{p}_{0}, t) \, \widetilde{\theta}^{(0)}(-\mathbf{q}_{0}, t || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) 
+i \delta_{\mathbf{k} + \mathbf{k}_{0}}^{3} k_{0j} \, \widetilde{u}_{j}(\mathbf{p}_{0}, t) \, \widetilde{\theta}^{(0)}(\mathbf{q}_{0}, t || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) 
+i \delta_{\mathbf{k} + \mathbf{k}_{0}}^{3} k_{0j} \, \widetilde{u}_{j}(\mathbf{q}_{0}, t) \, \widetilde{\theta}^{(0)}(\mathbf{p}_{0}, t || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) 
+i k_{0} \rightarrow \mathbf{p}_{0} \rightarrow \mathbf{q}_{0} \rightarrow \mathbf{k}_{0}) .$$
(3.12)

It is easily shown from (3.8), (3.11) and (3.12) that the DI field  $\widetilde{\theta}^{(1)}$  is expressed in terms of  $\widetilde{G}^{(0)}$  and  $\widetilde{\theta}^{(0)}$  as

$$\widetilde{\theta}^{(1)}(\mathbf{k}, t || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) = -i \, k_{J} \, \frac{(2\pi)^{9}}{L^{6}} \, \int_{t_{0}}^{t} dt'' \, \widetilde{G}^{(0)}(\mathbf{k}, t || -\mathbf{k}, t'' || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) \\
\times \, \left[ \, \delta_{\mathbf{k} - \mathbf{k}_{0}}^{3} \, \widetilde{u}_{j}(-\mathbf{p}_{0}, t'') \, \widetilde{\theta}^{(0)}(-\mathbf{q}_{0}, t'' || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) \right. \\
+ \delta_{\mathbf{k} - \mathbf{k}_{0}}^{3} \, \widetilde{u}_{j}(-\mathbf{q}_{0}, t'') \, \widetilde{\theta}^{(0)}(-\mathbf{p}_{0}, t'' || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) \\
+ \delta_{\mathbf{k} + \mathbf{k}_{0}}^{3} \, \widetilde{u}_{j}(\mathbf{p}_{0}, t'') \, \widetilde{\theta}^{(0)}(\mathbf{q}_{0}, t'' || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) \\
+ \delta_{\mathbf{k} + \mathbf{k}_{0}}^{3} \, \widetilde{u}_{j}(\mathbf{q}_{0}, t'') \, \widetilde{\theta}^{(0)}(\mathbf{p}_{0}, t'' || \mathbf{k}_{0}, \mathbf{p}_{0}, \mathbf{q}_{0}) \\
+ (\mathbf{k}_{0} \to \mathbf{p}_{0} \to \mathbf{q}_{0} \to \mathbf{k}_{0}) \, \right] . \tag{3.13}$$

In the same manner, the DI field  $\widetilde{u}_{i}^{(1)}$  is also expressed in terms of the NDI fields (see [6]).

## B. Closed equation for passive scalar spectrum

Here, we will derive an approximate expression of the third order correlation in the evolution equation (2.22) for the scalar correlation function  $\tilde{Z}$  in terms of  $\tilde{Z}$  itself and the Lagrangian velocity correlation function by the Lagrangian DIA. This approximation is based upon the following three assumptions: (I) The DI filed is much smaller in magnitude than the NDI field as long as  $t-t_0$  (for  $\tilde{\theta}$ ) or t-t' (for  $\tilde{G}$ ) is limited within the order of the correlation time scale of the velocity field. (II) Any two Fourier components without direct interaction are statistically independent of each other. For example, any two of  $\tilde{\theta}^{(0)}(\mathbf{k}_0,t||\mathbf{k}_0,\mathbf{p}_0,q_0)$ ,  $\tilde{\theta}^{(0)}(\mathbf{p}_0,t'||\mathbf{k}_0,\mathbf{p}_0,q_0)$  and  $\tilde{u}_k(q_0,t'')$  are statistically independent (see (3.10)). (III) The NDI field of the position function  $\tilde{\psi}^{(0)}$  is statistically independent of those of the Eulerian quantities such as  $\tilde{\psi}^{(0)}$  itself,  $\tilde{\theta}^{(0)}$ ,  $\tilde{G}^{(0)}$  and  $\tilde{u}_k^{(0)}$ . See ref. [7] for detailed discussions on assumptions (I) and (II).

First, we consider the one-time scalar correlation function  $\tilde{Z}(\boldsymbol{k},t,t)$  which is governed by (2.22). By substituting the direct-interaction decompositions into the right-hand side of (2.22) and by neglecting the higher-order terms of the DI fields (the assumption (I)), we obtain

(Nonlinear term of (2.22))
$$= -i k_{j} \left(\frac{2\pi}{L}\right)^{6} \sum_{\boldsymbol{p}} \sum_{\boldsymbol{q}} \left[ \overline{\widetilde{u}_{j}^{(0)}(-\boldsymbol{p},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \, \widetilde{\theta}^{(0)}(-\boldsymbol{q},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}) \, \widetilde{\theta}^{(0)}(-\boldsymbol{k},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \right. \\ \left. + \overline{\widetilde{u}_{j}^{(1)}(-\boldsymbol{p},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \, \widetilde{\theta}^{(0)}(-\boldsymbol{q},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}) \, \widetilde{\theta}^{(0)}(-\boldsymbol{k},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \right. \\ \left. + \overline{\widetilde{u}_{j}^{(0)}(-\boldsymbol{p},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \, \widetilde{\theta}^{(1)}(-\boldsymbol{q},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}) \, \widetilde{\theta}^{(0)}(-\boldsymbol{k},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \right. \\ \left. + \overline{\widetilde{u}_{j}^{(0)}(-\boldsymbol{p},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \, \widetilde{\theta}^{(0)}(-\boldsymbol{q},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}) \, \widetilde{\theta}^{(1)}(-\boldsymbol{k},t||\boldsymbol{k},\boldsymbol{p},\boldsymbol{q})} \right] .$$

$$(3.14)$$

Note that this approximation is valid as long as  $t-t_0$  is within the order of the time-scale of the velocity correlation function. The first term of the above equation vanishes under the assumption that  $\widetilde{u}_{j}^{(0)}(\boldsymbol{p},t||\boldsymbol{k},\boldsymbol{p},q), \ \widetilde{\theta}^{(0)}(\boldsymbol{p},t||\boldsymbol{k},\boldsymbol{p},q))$  and  $\widetilde{\theta}^{(0)}(\boldsymbol{q},t||\boldsymbol{k},\boldsymbol{p},q)$  are statistically independent of each other (the assumption (II)). Since the other three terms are evaluated similarly, we describe it here only for the third term. Substitution of the solution (3.13) of  $\widetilde{\theta}^{(1)}$  into the third term yields

(Third term of (3.14)) = 
$$-i k_{j} \left(\frac{2\pi}{L}\right)^{6} \sum_{\substack{p \ (k+p+q=o)}} \sum_{q} -i \left(-q_{m}\right) \frac{(2\pi)^{9}}{L^{6}} \int_{t_{0}}^{t} dt''$$

$$\times \left[\overline{\widetilde{G}^{(0)}(-q,t|q,t''|k,p,q)} \, \widetilde{u}_{m}^{(0)}(p,t''|k,p,q) \, \widetilde{u}_{j}^{(0)}(-p,t|k,p,q) \, \widetilde{\theta}^{(0)}(k,t''|k,p,q) \, \widetilde{\theta}^{(0)}(-k,t|k,p,q) \right] + \overline{\widetilde{G}^{(0)}(-q,t|q,t''|k,p,q)} \, \widetilde{u}_{m}^{(0)}(k,t''|k,p,q) \, \widetilde{u}_{j}^{(0)}(-p,t|k,p,q) \, \widetilde{\theta}^{(0)}(p,t''|k,p,q) \, \widetilde{\theta}^{(0)}(-k,t|k,p,q) \right] . \tag{3.15}$$

The second term of the above equation vanishes, because

$$\overline{\widetilde{\theta}^{(0)}(\boldsymbol{k}, t'' || \boldsymbol{k}_0, \boldsymbol{p}_0, \boldsymbol{q}_0) \ \widetilde{u}_i(-\boldsymbol{k}, t)} = 0$$
(3.16)

if the flow field is statistically isotropic.

The first term, on the other hand, is converted, under the assumption (II), into

(First term of (3.15)) = 
$$k_j \frac{(2\pi)^9}{L^6} \sum_{\substack{p \ (k+p+q=o)}} q_m \int_{t_0}^t dt'' \, \overline{\widetilde{G}^{(L)}(t|-q,q,t'')} \, \widetilde{Q}_{jm}(-p,t,t'') \, \widetilde{Z}(-k,t,t'') \, .$$
 (3.17)

Here, we have used the relations that

$$\overline{\widetilde{u}_{i}^{(0)}(\boldsymbol{k},t)} \ \widetilde{u}_{j}^{(0)}(-\boldsymbol{k},t') = \left(\frac{2\pi}{L}\right)^{3} \ \widetilde{Q}_{ij}(\boldsymbol{k},t,t') , \qquad (3.18)$$

$$\overline{\widetilde{\theta}^{(0)}(\mathbf{k},t)}\,\widetilde{\theta}^{(0)}(-\mathbf{k},t') = \left(\frac{2\pi}{L}\right)^3\,\widetilde{Z}(\mathbf{k},t,t') \tag{3.19}$$

and

$$\overline{\widetilde{G}^{(0)}(\mathbf{k},t|-\mathbf{k},t')} = \overline{\widetilde{G}^{(L)}(t|\mathbf{k},-\mathbf{k},t')}. \tag{3.20}$$

See ref. [6] and Appendix A for the derivations of (3.18) and (3.19)—(3.20), respectively. In a similar manner, we can calculate the second and fourth terms on the right-hand side of (2.22) to obtain

$$\left[\frac{\partial}{\partial t} + 2\kappa k^{2}\right] \widetilde{Z}(\boldsymbol{k}, t, t) = k_{j} k_{m} \frac{(2\pi)^{9}}{L^{6}} \sum_{\boldsymbol{p}} \sum_{\boldsymbol{q}} \int_{t_{0}}^{t} dt'' \, \widetilde{Q}_{jm}(-\boldsymbol{p}, t, t'') \\
\times \left[-\overline{\widetilde{G}^{(L)}(t|-\boldsymbol{q}, \boldsymbol{q}, t'')} \, \widetilde{Z}(-\boldsymbol{k}, t, t'') + \overline{\widetilde{G}^{(L)}(t|-\boldsymbol{k}, \boldsymbol{k}, t'')} \, \widetilde{Z}(-\boldsymbol{q}, t, t'')\right] \\
+ (\boldsymbol{k} \to -\boldsymbol{k}), \tag{3.21}$$

where use has been made of the incompressible condition of the velocity correlation function, i.e.,  $k_j \widetilde{Q}_{ij}(\boldsymbol{k}) = 0$ .

Turning now to the two-time correlation function  $\widetilde{Z}(k,t,t')$  and the response function  $\widetilde{G}^{(L)}(k,t,t')$ , we can integrate the governing equations (2.23) and (3.4) as

$$\widetilde{Z}(\mathbf{k}, t, t') = \widetilde{Z}(\mathbf{k}, t', t') \exp\left[-\kappa k^2 (t - t')\right]$$
(3.22)

and

$$\widetilde{G}^{(L)}(t|\mathbf{k}, -\mathbf{k}, t') = \frac{L^3}{(2\pi)^6} \exp\left[-\kappa k^2(t - t')\right]$$
 (3.23)

under the initial condition (3.5).

The combination of (3.21), (3.22) and (3.23) then yields

$$\left[\frac{\partial}{\partial t} + 2\kappa k^{2}\right] \widetilde{Z}(\boldsymbol{k}, t, t) = 2 k_{j} k_{m} \left(\frac{2\pi}{L}\right)^{3} \sum_{\boldsymbol{p}} \sum_{\boldsymbol{q}} \int_{t_{0}}^{t} dt'' \, \widetilde{Q}_{jm}(-\boldsymbol{p}, t, t'') \, \exp\left[-\kappa (k^{2} + q^{2})(t - t'')\right] \\
\times \left[-\widetilde{Z}(-\boldsymbol{k}, t'', t'') + \widetilde{Z}(-\boldsymbol{q}, t'', t'')\right]. \tag{3.24}$$

By taking the limit  $L \to \infty$ , we may convert (3.24) into

$$\left[ \frac{\partial}{\partial t} + 2 \kappa k^{2} \right] \widetilde{Z}(\boldsymbol{k}, t, t) = 2 k_{j} k_{m} \int d^{3}\boldsymbol{p} \int d^{3}\boldsymbol{q} \, \delta_{\boldsymbol{k}+\boldsymbol{p}+\boldsymbol{q}}^{3} \int_{t_{0}}^{t} dt'' \, \widetilde{Q}_{jm}(-\boldsymbol{p}, t, t'') \\
\times \exp \left[ -\kappa (k^{2} + \boldsymbol{q}^{2})(t - t'') \right] \left[ -\widetilde{Z}(-\boldsymbol{k}, t'', t'') + \widetilde{Z}(-\boldsymbol{q}, t'', t'') \right]. \tag{3.25}$$

This gives a closed system of equations for  $\widetilde{Z}(k,t,t)$  because the function  $\widetilde{Q}$  has already been known under the Lagrangian DIA [6]. Let us stress again that the present formulation of the Lagrangian DIA is quite simple and clear. From now on, we will confine ourselves to a stationary and isotropic field, so that  $\widetilde{Z}$  and  $\widetilde{Q}$  may be expressed as

$$\widetilde{Z}(\mathbf{k}, t, t) = Z^{\dagger}(k) , \qquad (3.26)$$

$$\widetilde{Q}_{ij}(\mathbf{k}, t, t') = \frac{1}{2} \widetilde{P}_{ij}(\mathbf{k}) Q^{\dagger}(\mathbf{k}, t - t') . \tag{3.27}$$

Equation (3.25) then reduces to

$$2\kappa Z^{\dagger}(k) = \iint_{\triangle_k} \mathrm{d}p \,\mathrm{d}q \,\frac{2\pi pq}{k} \,\sigma(k, p, q) \left[ -Z^{\dagger}(k) + Z^{\dagger}(q) \right] \int_0^{t-t_0} \mathrm{d}t' \,Q^{\dagger}(p, t') \,\exp\left[ -\kappa \left( k^2 + q^2 \right) t' \right]$$
$$= T(k)/4\pi k^4 \,\,, \tag{3.28}$$

where

as

$$\sigma(k, p, q) = \frac{k_j k_m}{k^2} \widetilde{P}_{jm}(\mathbf{p}) = \frac{(k + p + q)(k + p - q)(k - p + q)(-k + p + q)}{4p^2k^2} , \qquad (3.29)$$

the second equality is due to (2.26), (2.27) and (3.26). Equation (3.28) describes the balance between the scalar fluctuation transfer and its dissipation. Recall that (3.28) is valid as long as  $t - t_0$  does not exceed the order of the velocity correlation time (see the paragraph below (3.14)). The exponential decay of  $Q^{\uparrow}(k,t)$  with respect to t, however, permits us to replace  $t - t_0$  by infinity. The resultant closed equation (3.28) for the scalar correlation function may be derived also by LRA [5], although in ref. [20,21] only the LRA equation for  $\kappa = 0$  is given. Note, however, that there is an essential difference between LRA and the Lagrangian DIA in the underlying approximations (see ref. [7]). In the following, we call (3.28) the LRA-DIA equation.

#### IV. UNIVERSAL FORMS OF PASSIVE SCALAR SPECTRUM

We discuss in this section the functional form of the scalar spectrum  $\Theta(k)$  in the universal range for arbitrary values of the Schmidt number. To make the following analysis clear, we non-dimensionalize the wavenumber and the time

$$k = \hat{k} k_{\kappa} \tag{4.1}$$

and

$$t = \tau \, \epsilon^{-1/3} \, k^{-2/3} \, \,, \tag{4.2}$$

respectively, since the velocity correlation function  $Q^{\dagger}(k,t)$  is expressed in terms of these normalized variables as

$$Q^{\dagger}(k,t) = \frac{1}{2\pi} K \epsilon^{2/3} (k_{\kappa} \hat{k})^{-11/3} Q(\hat{k},\tau) \quad \text{with} \quad Q(0,0) = 1 , \qquad (4.3)$$

where K denotes the Kolmogorov constant. We define a non-dimensional scalar spectrum  $\widehat{\Theta}$  by

$$\Theta(k) = C_1 \chi \epsilon^{-1/3} k_{\kappa}^{-5/3} \widehat{\Theta}(\widehat{k}) . \tag{4.4}$$

It follows from (2.26), (3.26) and (4.4) that

$$Z^{\dagger}(k) = \frac{1}{4\pi k^2} \Theta(k) = \frac{1}{4\pi} C_1 \chi \epsilon^{-1/3} k_{\kappa}^{-11/3} \widehat{k}^{-2} \widehat{\Theta}(\widehat{k}) . \tag{4.5}$$

Then, the LRA-DIA equation (3.28) is written as

$$\widehat{\Theta}(\widehat{k}) = \frac{K}{2} s \widehat{k}^{-4/3} \iint_{\Delta_{1}} dp \, dq \, \sigma(1, p, q) \, p^{-8/3} \, q \left[ -\widehat{\Theta}(\widehat{k}) + \frac{1}{q^{2}} \, \widehat{\Theta}(\widehat{k}q) \right]$$

$$\times \int_{0}^{\infty} dt \, Q(\widehat{k}p, tp^{2/3}) \, \exp\left[ -s^{-1} \, \widehat{k}^{4/3} \, (1 + q^{2}) \, t \right]$$

$$= T(k) / \left[ 2\kappa \, k^{2} \, C_{1} \, \chi \, \epsilon^{-1/3} \, k_{\kappa}^{-5/3} \right] ,$$

$$(4.6)$$

where the second equality follows from (2.27) and (4.4).

In the following subsections we will describe the solution for various values of k and s: the  $k^{-5/3}$  power spectrum at  $k \ll \min\{k_{\rm K}, k_{\rm C}\}$  for arbitrary s in Sec. A, numerical solutions for finite s in Sec. B, the asymptotic forms for  $s \gg 1$  in Sec. C, and for  $s \ll 1$  in Sec. D, and bumps in the spectrum at the end of power law regions in Sec. E.

## A. Inertial-advective range

Assuming that both the kinematic viscosity  $\nu$  and the scalar diffusivity  $\kappa$  are so small that  $\max\{k_v, k_s\} \ll \min\{k_{\kappa}, k_{\kappa}\}$ , we consider the inertial-advective range

$$\max\{k_v, k_s\} \ll k \ll \min\{k_{\rm K}, k_{\rm C}\} \quad \Longrightarrow \quad \widehat{k} \ll \min\{1, s^{3/4}\} \ . \tag{4.7}$$

Then, since  $s^{-1} \widehat{k}^{4/3} \ll 1$  and  $\widehat{k} \ll 1$ , (4.6) may be written as

$$0 = \iint_{\Delta_1} \mathrm{d}p \,\mathrm{d}q \,\sigma(1, p, q) \,p^{-10/3} \,q \left[ -\widehat{\Theta}(\widehat{k}) + \frac{1}{q^2} \,\widehat{\Theta}(\widehat{k}q) \right] W_1 \qquad (\forall s, \ k \ll k_{\kappa}, k_{c})$$

$$(4.8)$$

at the leading order, where

$$W_1 = \int_0^\infty dt \, Q(0, t) \ . \tag{4.9}$$

Since no characteristic scales appear in (4.8), it allows a power form of the spectrum function. Substitution of

$$\widehat{\Theta}(\widehat{k}) \propto \widehat{k}^{-a} \tag{4.10}$$

into (4.8) leads to

$$0 = \left[ \int_0^1 dq \int_{1-q}^{1+q} dp + \int_1^\infty dq \int_{q-1}^{q+1} dp \right] \sigma(1,p,q) p^{-10/3} q (q^{-a-2} - 1) , \qquad (4.11)$$

which is rewritten as

$$0 = \int_0^1 dq \int_{1-a}^{1+q} dp \, \sigma(1, p, q) \, p^{-10/3} \, q \, (q^{-a-2} - 1) \, (1 - q^{a-5/3})$$

$$(4.12)$$

by changing the integral variables in the second term on the right-hand side of (4.11) as p = p'/q' and q = 1/q'. This equation has two apparent scaling laws of a = 5/3 and -2. The former corresponds to the Obukhov-Corrsin spectrum (1.4) with finite flux  $\Pi(k) = \chi(\neq 0)$ . The latter, on the other hand, represents a state of equipartition of the fluctuation of the passive scalar field with vanishing flux. Since we are interested in a statistically stationary state with finite flux through the advective range toward the diffusive range, we will not consider this solution in the following.

The Obukhov-Corrsin constant  $C_1$  in (1.4) is shown to be expressed by  $W_1$  and the Kolmogorov constant K as

$$C_1 = \frac{910\sqrt{3}}{729\pi \, KW_1} \tag{4.13}$$

(see Appendix B for derivation). A similar relation was derived in the LHDIA [18,22]. A numerical integration of (4.9) gives  $W_1 = 1.19$  and then  $C_1 = 0.34$  [21]. This is about a half of the experimental values which scatter around  $(5/3) \times 0.4 = 0.67$  [14]. The reason of this discrepancy is not known. Incidentally, the abridged LHDIA yields  $C_1 = 0.208$  [22].

#### B. Finite Schmidt number

We describe here numerical solutions of the LRA-DIA equation (4.6) for finite Schmidt numbers. We search for a solution by an iteration method that approaches the -5/3 power form in the inertial-advective range  $k \ll \min\{k_{\text{K}}, k_{\text{C}}\}$  discussed in the preceding subsection.

The scalar spectra for  $s \ge 1$  are shown in Fig.1 together with the asymptotic form in the limit  $s \gg 1$  (see Sec. IV C). The wavenumber is normalized by  $k_{\rm K}$  in (a) and by  $k_{\rm B}$  in (b). In these figures we can see that the -5/3 power law range extends up to  $k_{\rm K}$ , which is consistent with the argument in the preceding subsection because  $k_{\rm K} < k_{\rm C}$  if s > 1, and that the function obeys the -1 power law in the larger-wavenumber range  $k_{\rm K} \ll k \ll k_{\rm B}$ . This -1 power law range widens with the Schmidt number, and is followed by an exponential decay at  $k \gg k_{\rm B}$ . These behaviors are consistent

with the phenomenology for large Schmidt numbers by Batchelor [12] (see Sec. IV C for detailed discussions). We also observe a bump in the spectrum around  $k_{\rm B}$ , which will be discussed in Sec. IV E.

In Fig.2, we plot the numerical solutions in the cases of  $s \leq 1$  together with the asymptotic form in the small Schmidt number limit (see Sec. IV D). Since the upper limit of the inertial-advective range is  $k_{\rm C}$  for s < 1 (see 1.4), the wavenumber is normalized by  $k_{\rm C}$  instead of  $k_{\rm K}$  in (a). The -5/3 power law is actually established at  $k \ll k_{\rm C}$ . Moreover we observe that as s decreases the spectrum seems to approach the -17/3 power law at  $k \gg k_{\rm C}$ , which was phenomenologically predicted by Batchelor *et al.* [13]. The wavenumber in (b) is normalized by  $k_{\rm K}$  in order to focus the spectrum around  $k_{\rm K}$ . We can see in (a) and (b) that the -17/3 power law range extends between  $k_{\rm C}$  and  $k_{\rm K}$  and widens as s decreases.

#### C. Large Schmidt number limit

We consider here the universal form of the scalar spectrum in the large Schmidt number limit. To do it we introduce variable normalizations of the wavenumber and the spectrum such that

$$k = \check{k} k_{\kappa} s^{\alpha} \tag{4.14}$$

and

$$\Theta(k) = C_1 \chi \epsilon^{-1/3} k_{\kappa}^{-5/3} s^{\beta} \check{\Theta}(\check{k})$$

$$(4.15)$$

with undetermined parameters  $\alpha$  and  $\beta$ . Note that  $\alpha$  indicates the reference wavenumber which we focus on. The reference wavenumbers for  $\alpha = 0$ , 1/2 and 3/4, for example, are  $k_{\rm K}$ ,  $k_{\rm B}$  and  $k_{\rm C}$ , respectively. It should be mentioned that in the large (or small) Schmidt number limit the characteristic wavenumbers  $\{k_{\rm K}, k_{\rm C}, k_{\rm B}\}$  are separated infinitely far from each other on a logarithmic scale.

On substitution of (4.14) and (4.15) into (4.6), we obtain

$$\tilde{\Xi}(\tilde{k}) = \frac{K}{2} s^{1-4\alpha/3} \tilde{k}^{-4/3} \iint_{\Delta_{1}} dp \, dq \, \sigma(1, p, q) \, p^{-8/3} \, q \left[ -\tilde{\Xi}(\tilde{k}) + \tilde{\Xi}(\tilde{k}q) \right] \\
\times \int_{0}^{\infty} dt \, Q(s^{\alpha} \tilde{k} p, t p^{2/3}) \, \exp\left[ -s^{-(1-4\alpha/3)} \, \tilde{k}^{4/3} \, (1+q^{2}) \, t \right], \tag{4.16}$$

where

$$\check{\Xi}(\check{k}) = \check{\Theta}(\check{k})/\check{k}^2 \ . \tag{4.17}$$

Since this equation depends upon the Schmidt number only through  $s^{\alpha}$  and  $s^{1-4\alpha/3}$ , we consider the cases of  $\alpha < 0$ ,  $\alpha = 0$  and  $\alpha > 0$  in turn. (It will be shown in subsection [3] below that an apparent critical value  $\alpha = 3/4$  is actually irrelevant.)

#### [1] Inertial-advective range ( $\alpha < 0$ )

For  $\alpha < 0$  we are in the wavenumber range below  $k_{\rm K}$  because  $s^{\alpha} < 1$ . Since  $s^{\alpha} \ll 1$  and  $s^{-(1-4\alpha/3)} \ll 1$ , (4.16) leads to

$$0 = \iint_{\Delta_1} dp \, dq \, \sigma(1, p, q) \, p^{-10/3} \, q \, \left[ -\tilde{\Xi}(\check{k}) + \tilde{\Xi}(\check{k}q) \, \right] W_1 \qquad (s \gg 1, \alpha < 0) \, . \tag{4.18}$$

This is identical to the LRA-DIA equation (4.8) in the inertial-advective range, which yields the spectrum  $\tilde{\Theta}(\tilde{k})$  proportional to  $\tilde{k}^{-5/3}$ .

#### [2] Around Kolmogorov wavenumber ( $\alpha = 0$ )

For  $\alpha = 0$   $(k = O(k_K))$ , (4.16) is deduced to

$$0 = \iint_{\Delta_1} dp \, dq \, \sigma(1, p, q) \, p^{-10/3} \, q \, \left[ - \check{\Xi}(\check{k}) + \check{\Xi}(\check{k}q) \, \right] \, \int_0^\infty dt \, Q(\check{k}p, t) \qquad (s \gg 1, \alpha = 0) \, . \tag{4.19}$$

This equation has an asymptotic solution proportional to  $\tilde{k}^{-5/3}$  for  $\tilde{k} \ll 1$  because it coincides with (4.8). In the opposite limit  $\tilde{k} \gg 1$ , on the other hand, we find that the contribution from the region  $p \ll 1$  is dominant in the integral, since Q(k,t) decays exponentially with k as  $Q(k,t) \propto \exp(-ck)$  (see the paragraph below (4.62)). Thus, by changing the integral variable as q = 1 + px, we may rewrite (4.19) as

$$0 = \int_0^\infty dp \, p^{-3} \int_{-1}^1 dx \, \sigma(1, p, 1 + px) \, (1 + px) \left[ -\tilde{\Xi}(\check{k}) + \tilde{\Xi}(\check{k}(1 + px)) \right] \int_0^\infty dt \, Q(\check{k}p, t)$$
 (4.20)

Substituting the expansions

$$\sigma(1, p, 1 + px) (1 + px) = (1 - x^2) + 2 (1 - x^2) px + O(p^2)$$
(4.21)

 $\quad \text{and} \quad$ 

$$-\check{\Xi}(\check{k}) + \check{\Xi}(\check{k}(1+px)) = \check{k}px \frac{\mathrm{d}\check{\Xi}}{\mathrm{d}\check{k}} + \frac{1}{2} (\check{k}px)^2 \frac{\mathrm{d}^2\check{\Xi}}{\mathrm{d}\check{k}^2} + O(p^3)$$

$$(4.22)$$

into (4.19) and carrying out the integration with respect to x, we obtain

$$0 = \int_0^\infty \mathrm{d}p \, p^{-1} \, \frac{\mathrm{d}}{\mathrm{d}\tilde{k}} \left[ \, \check{k}^4 \, \frac{\mathrm{d}\tilde{\Xi}}{\mathrm{d}\tilde{k}} \, \right] \, \int_0^\infty \mathrm{d}t \, Q(\check{k}p, t) \tag{4.23}$$

at the leading order of p. Hence, the leading order of (4.19) for  $\check{k}\gg 1$  is

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{k}} \left[ \check{k}^4 \frac{\mathrm{d}\tilde{\Xi}}{\mathrm{d}\tilde{k}} \right] = 0 \qquad (s \gg 1, \alpha = 0, \check{k} \gg 1) , \qquad (4.24)$$

which gives  $\check{\Xi}(\check{k}) \propto \check{k}^{-3}$  (i.e.  $\check{\Theta}(\check{k}) \propto \check{k}^{-1}$ ).

Thus, a solution of (4.19) behaves as  $\check{\Theta}(\check{k}) \propto \check{k}^{-5/3}$  for  $\check{k} \ll 1$  and  $\check{k}^{-1}$  for  $\check{k} \gg 1$ . We now solve (4.19) numerically so that the solution may satisfy these asymptotic forms. The result is drawn in Fig.3(a), in which the transition from the -5/3 to the -1 power laws occurs around the Kolmogorov wavenumber (see also Fig.1(a)).

In the cases of  $\alpha > 0$   $(k \gg k_{\rm K})$ , the contribution from  $p \ll 1$  is dominant in the integral of (4.16) because Q(k,t) decays exponentially with k at large k. Therefore, we may carry out the integration with respect to q by putting q = 1 + px and by expanding the integrand into power series of p up to  $O(p^2)$  (see (4.21) and (4.22)) to obtain

$$\tilde{\Xi}(\tilde{k}) = \frac{K}{15} s^{1-4\alpha/3} \, \tilde{k}^{-4/3} \, \int_0^\infty \mathrm{d}p \, p^{1/3} \int_0^\infty \mathrm{d}t \, Q(s^\alpha \tilde{k}p, tp^{2/3}) \, \exp\left[-2 \, s^{-(1-4\alpha/3)} \, \tilde{k}^{4/3} \, t \right] \\
\times \left[ 4 \left( \tilde{k} - \tilde{k}^{7/3} \, s^{-(1-4\alpha/3)} \, t \right) \, \frac{\mathrm{d}\tilde{\Xi}}{\mathrm{d}\tilde{k}} + \tilde{k}^2 \, \frac{\mathrm{d}^2 \tilde{\Xi}}{\mathrm{d}\tilde{k}^2} \right]$$
(4.25)

at the leading order. By changing the integral variables as  $(p,t) \to (s^{-\alpha}p,s^{2\alpha/3}t)$ , this equation is converted into

$$\tilde{\Xi}(\check{k}) = \frac{K}{15} s^{1-2\alpha} \check{k}^{-4/3} \int_0^\infty dp \, p^{1/3} \int_0^\infty dt \, Q(\check{k}p, tp^{2/3}) \, \exp\left[-2 \, s^{-(1-2\alpha)} \, \check{k}^{4/3} \, t\right] \\
\times \left[ 4 \left(\check{k} - \check{k}^{7/3} \, s^{-(1-2\alpha)} \, t\right) \frac{d\check{\Xi}}{d\check{k}} + \check{k}^2 \, \frac{d^2 \check{\Xi}}{d\check{k}^2} \right] \qquad (s \gg 1, \alpha > 0) \, . \tag{4.26}$$

Since this equation depends upon s through only  $s^{1-2\alpha}$ , we will examine three cases  $0 < \alpha < 1/2$ ,  $\alpha = 1/2$  and  $1/2 < \alpha$  separately in the following subsections.

[3-1] Viscous-advective range  $(0 < \alpha < 1/2)$ 

If  $0 < \alpha < 1/2$   $(k_{\rm K} \ll k \ll k_{\rm B})$ , (4.26) reduces to

$$0 = \frac{K}{15} s^{1-2\alpha} W_2 \check{k}^{-1} \left[ 4 \frac{d\tilde{\Xi}}{d\check{k}} + \check{k} \frac{d^2 \check{\Xi}}{d\check{k}^2} \right] , \qquad (4.27)$$

where

$$W_2 = \int_0^\infty dx \, \int_0^\infty dy \, x^{-1/3} \, Q(x, y) \,. \tag{4.28}$$

Since all the factors outside the brackets in (4.27) are non-zero, we have

$$0 = 4 \frac{\mathrm{d}\check{\Xi}}{\mathrm{d}\check{k}} + \check{k} \frac{\mathrm{d}^2 \check{\Xi}}{\mathrm{d}\check{k}^2} \qquad (s \gg 1, 0 < \alpha < 1/2) , \qquad (4.29)$$

which yields a power law solution as  $\check{\Xi}(\check{k}) = \propto \check{k}^{-3}$  (that is,  $\check{\Theta}(\check{k}) \propto \check{k}^{-1}$ ). This implies that we see only the -1 power law (1.5) of the spectrum function in the viscous-advective range  $(k_{\kappa} \ll k \ll k_{\rm B})$ .

In order to estimate the universal constant  $C_2$  in (1.5) we express the transfer function (2.27) in this range in terms of  $\Theta(k)$  as

$$T(k) = \frac{2}{15} K \nu W_2 k_K^2 \frac{d}{dk} \left[ k^4 \frac{d}{dk} \left[ \frac{\Theta(k)}{k^2} \right] \right] , \qquad (4.30)$$

which follows from (2.27), (4.15), (4.17) and the right-hand side of (4.27). Then, the flux function  $\Pi(k)$ , defined by (2.28), is written as

$$\Pi(k) = -\frac{2}{15} K \nu W_2 k_{\kappa}^2 k^4 \frac{\mathrm{d}}{\mathrm{d}k} \left[ \frac{\Theta(k)}{k^2} \right] , \qquad (4.31)$$

since the contribution from the diffusive range to the integration in (2.28) is negligible. By substituting the power law (1.5) into the above equation, we obtain

$$\Pi(k) = \frac{2}{5} \chi K W_2 C_2 , \qquad (4.32)$$

which yields

$$C_2 = \frac{5}{2KW_2} = 1.30 , (4.33)$$

because  $\Pi(k) = \chi$  in the advective range and the numerical value of  $W_2$  is 1.11. This value of  $C_2$  should be compared with  $3.9 \pm 1.5$  [15],  $3.7 \pm 1.5$  [16], which were measured in tidal channel flows, as well as  $1.5 \sim 2.5$  (LRA-DIA), 1.5 (a modified LRA) [20],  $0.6 \sim 1.0$  (abridged LHDIA; the deviation is too large) and  $1.9 \sim 2.0$  (strain-based abridged LHDIA) [19], which were determined numerically from various Lagrangian closure equations. All estimations by these closure theories are quite small compared with the experimental data. It should be mentioned here an important difference in the methods of evaluation of  $C_2$  used in the above closure theories and the present one; they estimated it from a late state of a freely decaying solution whereas we did it from a stationary solution. This is the reason why the numerical value obtained in ref. [20] is different from ours, \* though the same LRA-DIA equation is solved. Since their results themselves show large deviations, such a method may not be appropriate to evaluate the universal form. In the above measurements [15,16] the universal constant  $C_2$  in the viscous-advective range is evaluated by fitting the temperature spectrum function with the Batchelor form ((4.44) below) in the whole viscous range, in which the Schmidt number is about 10. It should be pointed out here a reservation that the -1 power range is not so wide at this value of the Schmidt number (see Fig.1(b)).

A comment on Gibson's bounds [23] may be in order. He derived that  $\sqrt{3} \le C_2 \le 2\sqrt{3}$  for a homogeneous dissipation field by making use of the Batchelor form (4.44) of the spectrum function with the relation  $C_2 = -(\epsilon/\nu)^{1/2}/\gamma$  ( $\gamma$  is the least eigenvalue of the rate-of-strain tensor). Since the Batchelor form (and then Gibson's bounds) is not a solution to the LRA-DIA equation but a phenomenology, it is not unnatural that the present estimation of  $C_2$  violates these bounds.

[3-2] Around Batchelor wavenumber ( $\alpha = 1/2$ )

For  $\alpha = 1/2 \ (k = O(k_{\rm B}))$ , (4.26) reduces to

$$\tilde{\Xi}(\tilde{k}) = 4 \left[ f_1(\tilde{k}) \tilde{k}^{-1/3} - f_2(\tilde{k}) \tilde{k} \right] \frac{d\tilde{\Xi}}{d\tilde{k}} + f_1(\tilde{k}) \tilde{k}^{2/3} \frac{d^2 \tilde{\Xi}}{d\tilde{k}^2} \qquad (s \gg 1, \alpha = 1/2) , \tag{4.34}$$

<sup>\*</sup>Factor 2 is missing in Sec.4.4 of [20].

where

$$f_1(k) = \frac{K}{15} \int_0^\infty \mathrm{d}p \, p^{1/3} \int_0^\infty \mathrm{d}t \, Q(kp, tp^{2/3}) \, \exp\left[-2k^{4/3}t\right] \tag{4.35}$$

and

$$f_2(k) = \frac{K}{15} \int_0^\infty dp \, p^{1/3} \int_0^\infty dt \, t \, Q(kp, tp^{2/3}) \, \exp\left[-2k^{4/3}t\right] \,. \tag{4.36}$$

It is easy to show that (4.34) has the asymptotic solution of  $\check{\Xi}(\check{k}) \propto \check{k}^{-3}$  (i.e.  $\check{\Theta}(\check{k}) \propto \check{k}^{-1}$ ) at  $\check{k} \ll 1$  because (4.34) coincides with (4.29) in this limit. (Note that  $f_1(\check{k}) \propto \check{k}^{-2/3}$  and  $f_2(\check{k}) \propto \log \check{k}$  for  $\check{k} \ll 1$ .)

In the opposite limit  $\check{k} \gg 1$ , on the other hand, we have the asymptotic expressions,

$$f_1(\check{k}) = c_1 \, \check{k}^{-8/3} + c_2 \, \check{k}^{-14/3} + O(\check{k}^{-20/3}) \quad \text{and} \quad f_2(\check{k}) = \frac{1}{2} \, c_1 \, \check{k}^{-4} + c_2 \, \check{k}^{-6} + O(\check{k}^{-8}) \quad (\check{k} \to \infty) \,,$$
 (4.37)

where

$$c_1 = \frac{K}{30} \int_0^\infty \mathrm{d}p \, p^{1/3} \, Q(p,0) = \frac{1}{60} \tag{4.38}$$

and

$$c_2 = \frac{K}{60} \int_0^\infty dp \, p \, \left. \frac{\partial}{\partial t} \, Q(p, t) \, \right|_{t=0} = \frac{7S}{360\sqrt{60}} \, . \tag{4.39}$$

The second equalities of (4.38) and (4.39) are respectively derived from the relation  $\epsilon = 2\nu \int_0^\infty \mathrm{d}k k^2 E(k) = 2\nu K \epsilon^{2/3} \int_0^\infty \mathrm{d}k k^{1/3} Q(k/k_K, 0)$  and the Lagrangian DIA equation for Q(k, t) together with the expression of the skewness factor S of the velocity derivative (see (4.19) and (4.23) in ref. [6]). Hence, in the limit  $\check{k} \to \infty$ , (4.34) reduces to

$$\check{k}^2 \, \check{\Xi}(\check{k}) = 2c_1 \check{k}^{-1} \, \frac{\mathrm{d}\check{\Xi}}{\mathrm{d}\check{k}} + \left[ c_1 + c_2 \, \check{k}^{-2} \, \right] \frac{\mathrm{d}^2 \check{\Xi}}{\mathrm{d}\check{k}^2} \tag{4.40}$$

at the leading order, from which the asymptotic form of  $\check{\Xi}$  is derived to be

$$\check{\Xi}(\check{k}) \propto \check{k}^a \exp\left[-\check{k}^2/(2\sqrt{c_1})\right] \qquad (s \gg 1, \alpha = 1/2, \check{k} \gg 1)$$
(4.41)

or

$$\Theta(k) \propto k^{a+2} \exp \left[ -\frac{\sqrt{60}}{2} \kappa \left( \frac{\nu}{\epsilon} \right)^{1/2} k^2 \right]$$
 (4.42)

with

$$a = \frac{1}{2} \left[ \frac{c_2}{(c_1)^{3/2}} - 3 \right] = \frac{7}{12} S - \frac{3}{2} \approx -1.9 ,$$
 (4.43)

where we used  $S \approx -0.66$  which had been already determined by the Lagrangian DIA [24]. Thus we have found asymptotic solutions of (4.34) in both of small and large wavenumber limits. The numerical solution to this equation

integrated from the large wavenumber limit is shown in Fig.3(b), in which we can clearly see the -1 power law and the exponential asymptotes at small and large wavenumbers.

The asymptotic form (4.42) is similar to the one derived phenomenologically by Batchelor [12], which is

$$\Theta(k) = C_2 \chi \nu^{1/2} \epsilon^{-1/2} k^{-1} \exp \left[ -C_2 \kappa \left( \frac{\nu}{\epsilon} \right)^{1/2} k^2 \right]. \tag{4.44}$$

A comparison of the arguments of the exponential function in (4.42) and (4.44) would give  $C_2 = \sqrt{60}/2 = 3.87$ . This value is in a quite good agreement with experimental values  $3.9 \pm 1.5$  [15],  $3.7 \pm 1.5$  [16]. Recall that  $C_2$  is determined experimentally by the use of the Batchelor form (4.44). Another analytical form of the spectrum in the viscous range,

$$\Theta(k) = C_2 \chi (\epsilon/\nu)^{1/2} \check{k}^{-1} \left[ 1 + \sqrt{6C_2} \check{k} \right] \exp \left[ -\sqrt{6C_2} \check{k} \right], \tag{4.45}$$

was derived by Mjolsness [25] based upon Kraichnan's LHDIA equation under the assumption that the transfer function is proportional to  $\check{k}^{-1}\mathrm{d}\check{\Xi}/\mathrm{d}\check{k}+\mathrm{d}^2\check{\Xi}/\mathrm{d}\check{k}^2$ . Recently Bogucki et al. [26] have shown that (4.45) agrees with a direct numerical simulation with a fitting parameter  $C_2=5.26\pm0.25$ . However, we would like to note two points to be considered. First, (4.45) should not be precise at wavenumbers larger than  $k_{\mathrm{B}}$  because the above expression of the transfer function can be applied only at  $k\ll k_{\mathrm{B}}$  under Kraichnan's formulation of Lagrangian DIA just like the present one (see the paragraph below (4.36)). Second, the value of  $C_2=5.26$  suggested by Bogucki et al. is much larger than the experimental data [15,16].

[3-3] Far viscous-diffusive range ( $\alpha > 1/2$ )

For  $\alpha > 1/2$   $(k \gg k_{\scriptscriptstyle \rm B})$ , it follows from (4.26) that

$$\tilde{\Xi}(\tilde{k}) = 0 \qquad (s \gg 1, \alpha > 1/2) \ . \tag{4.46}$$

This is consistent with the exponential decay of the spectrum function at wavenumbers larger than  $k_{\rm B}$  discussed in the preceding subsection.

## D. Small Schmidt number limit

In order to examine the small Schmidt number limit we write the LRA-DIA equation in terms of the normalized wavenumber  $\check{k}$  and the spectrum  $\check{\Theta}$  as

$$\tilde{\Theta}(\check{k}) = \frac{K}{2} s^{1-4\alpha/3} \check{k}^{-4/3} \iint_{\Delta_1} dp \, dq \, \sigma(1, p, q) \, p^{-8/3} \, q \left[ -\tilde{\Theta}(\check{k}) + \frac{1}{q^2} \, \tilde{\Theta}(\check{k}q) \right] \\
\times \int_0^\infty dt \, Q(s^\alpha \check{k}p, tp^{2/3}) \, \exp\left[ -s^{-(1-4\alpha/3)} \, \check{k}^{4/3} \, (1+q^2) \, t \right]. \tag{4.47}$$

This equation, which is equivalent to (4.16), is more convenient in the present subsection because  $\check{\Xi}$  is not necessary to be dealt with. Since (4.47) depends upon s only through  $s^{\alpha}$  and  $s^{1-4\alpha/3}$ , we will examine three cases  $\alpha > 3/4$ ,  $\alpha = 3/4$ ,  $0 < \alpha < 3/4$ ,  $\alpha = 0$  and  $\alpha < 0$  separately in the following subsections.

For  $\alpha > 3/4$   $(k \ll k_c)$ , since  $s^{\alpha} \ll 1$  and  $s^{-(1-4\alpha/3)} \ll 1$ , (4.47) reduces to

$$0 = \iint_{\Delta_1} dp \, dq \, \sigma(1, p, q) \, p^{-10/3} \, q \left[ -\check{\Theta}(\check{k}) + \frac{1}{q^2} \, \check{\Theta}(\check{k}q) \right] \qquad (s \ll 1, \alpha > 3/4) \,, \tag{4.48}$$

which is identical to (4.8). Hence, the spectrum obeys the -5/3 power law. This is consistent with the argument in Sec. IV A because  $k_{\rm C} \ll k_{\rm K}$  for  $s \ll 1$ .

[2] Around Obukhov-Corrsin wavenumber ( $\alpha = 3/4$ )

In the case of  $\alpha = 3/4$   $(k = O(k_c))$ , (4.47) leads to

$$\tilde{\Theta}(\check{k}) = \frac{K}{2} \check{k}^{-4/3} \iint_{\triangle_{1}} dp \, dq \, \sigma(1, p, q) \, p^{-8/3} \, q \left[ -\check{\Theta}(\check{k}) + \frac{1}{q^{2}} \, \check{\Theta}(\check{k}q) \right] \\
\times \int_{0}^{\infty} dt \, Q(0, tp^{2/3}) \, \exp\left[ -\check{k}^{4/3} \, (1 + q^{2}) \, t \right] \qquad (s \ll 1, \alpha = 3/4) \, .$$
(4.49)

Since this equation coincides with (4.48) at  $\check{k} \ll 1$ , its asymptotic solution is proportional to  $\check{k}^{-5/3}$  in this limit. At  $\check{k} \gg 1$ , on the other hand, the exponential factor in the integrand allows us to replace  $Q(0, tp^{2/3})$  by Q(0,0) = 1. Then, we obtain

$$\check{\Theta}(\check{k}) = \frac{K}{2} \, \check{k}^{-8/3} \, \iint_{\Delta_1} \mathrm{d}p \, \mathrm{d}q \, \sigma(1, p, q) \, \frac{p^{-8/3} \, q}{1 + q^2} \left[ -\check{\Theta}(\check{k}) + \frac{1}{q^2} \, \check{\Theta}(\check{k}q) \right] \, . \tag{4.50}$$

In order to estimate the limiting behavior for  $\check{k}\gg 1$  of this integral we divide it into three parts as

$$\check{\Theta}(\check{k}) = \frac{K}{2} \check{k}^{-8/3} \int_{0}^{\varepsilon} dp \int_{1-p}^{1+p} dq \, \sigma(1, p, q) \, \frac{p^{-8/3} \, q}{1+q^2} \left[ -\check{\Theta}(\check{k}) + \frac{1}{q^2} \, \check{\Theta}(\check{k}q) \right] 
- \frac{K}{2} \check{k}^{-8/3} \int_{\varepsilon}^{\infty} dp \int_{|1-p|}^{1+p} dq \, \sigma(1, p, q) \, \frac{p^{-8/3} \, q}{1+q^2} \, \check{\Theta}(\check{k}) 
+ \frac{K}{2} \check{k}^{-8/3} \int_{\varepsilon}^{\infty} dp \int_{|1-p|}^{1+p} dq \, \sigma(1, p, q) \, \frac{p^{-8/3} \, q}{(1+q^2)q} \, \check{\Theta}(\check{k}q) ,$$
(4.51)

where  $\varepsilon$  ( $\ll$  1) is a constant. The first and second terms are respectively proportional to  $\varepsilon^{4/3} \, \check{k}^{-2/3} \, \frac{\mathrm{d}}{\mathrm{d}\check{k}} [\check{k}^2 \frac{\mathrm{d}}{\mathrm{d}\check{k}} (\check{\Theta}/\check{k}^2)]$  and  $\varepsilon^{-2/3} \check{k}^{-8/3} \check{\Theta}(\check{k})$ , both of which will be shown to be smaller than the third term (see (4.52) below). The asymptotic behavior of the third term may be obtained by noting that a dominant contribution to the integral comes from the vicinity of  $q \approx 0$  if  $\check{\Theta}$  is a decreasing function. Thus, we may convert (4.50) into

$$\check{\Theta}(\check{k}) = \frac{2K}{3} \check{k}^{-17/3} \int_0^\infty dq \, q^2 \, \check{\Theta}(q) \qquad (s \ll 1, \alpha = 3/4, \check{k} \gg 1) \,. \tag{4.52}$$

Hence, the spectrum obeys the -17/3 power law at  $\check{k} \gg 1$ . The numerical solution of (4.49) shown in Fig.4(a) actually exhibits a transition from the -5/3 to the -17/3 power laws around the Obukhov-Corrsin wavenumber  $k_{\rm C}$ .

[3] Diffusive range (
$$\alpha < 3/4$$
)

For  $\alpha < 3/4$   $(k \gg k_c)$ , since  $s^{-1+4\alpha/3} \gg 1$ , the exponential factor in (4.47) is a rapidly decreasing function and the contribution from the vicinity of the origin is dominant in the integral with respect to t. It then reduces to

$$\check{\Theta}(\check{k}) = \frac{K}{2} s^{2(1-4\alpha/3)} \check{k}^{-8/3} \iint_{\Delta_1} dp \, dq \, \sigma(1, p, q) \, \frac{p^{-8/3} q}{1+q^2} \left[ -\check{\Theta}(\check{k}) + \frac{1}{q^2} \, \check{\Theta}(\check{k}q) \right] \, Q(s^{\alpha} \check{k}p, 0) \, . \tag{4.53}$$

This is further simplified by dividing the integral with respect to q into two regions,  $I_1$  ( $0 \le q \le \varepsilon$ ) and  $I_2$  ( $q \ge \varepsilon$ ), where  $\varepsilon$  ( $\ll$  1) is a constant independent of s. Then, (4.53) is written as

$$\check{\Theta}(\check{k}) = \frac{K}{2} \, s^{2(1-4\alpha/3)} \, \check{k}^{-8/3} \, \Big[ \, I_1 + I_2 \, \Big] \, . \tag{4.54}$$

It is easy to show that  $I_2$  and the first term of  $I_1$  are bounded irrespective of the value of s. As for the second term of  $I_1$ , we make a change of integral variables as p = 1 + qx, expand the integrand around q = 0 and carry out the integral with respect to x to obtain

(Second term of 
$$I_1$$
) =  $\frac{4}{3} Q(\check{k}s^{\alpha}, 0) \check{k}^{-3} \left[ \int_0^{\infty} dq \, q^2 \, \check{\Theta}(q) - \int_{\varepsilon}^{\infty} dq \, q^2 \, \check{\Theta}(q) \right]$   
=  $\frac{4}{3} Q(\check{k}s^{\alpha}, 0) \check{k}^{-3} \left[ \frac{1}{2C_1} s^{-\beta - 3\alpha + 1} - \int_{\varepsilon}^{\infty} dq \, q^2 \, \check{\Theta}(q) \right],$  (4.55)

where use has been made of  $\chi = 2 \kappa \int_0^\infty dk \, k^2 \, \Theta(k)$ . The second term of this equation is also bounded and neglected compared with the first term in the limit  $s \to 0$  (because (4.57) and  $\alpha < 3/4$ ). On substitution of the first term of (4.55) into (4.54), we find

$$\check{\Theta}(\check{k}) = \frac{K}{3C_1} \, s^{3-17\alpha/3-\beta} \check{k}^{-17/3} \, Q(s^{\alpha} \check{k}, 0) \ . \tag{4.56}$$

In order that (4.56) may have a nontrivial solution, we must set

$$\beta = 3 - \frac{17}{3} \alpha \tag{4.57}$$

to obtain

$$\check{\Theta}(\check{k}) = \frac{K}{3C_1} \check{k}^{-17/3} Q(s^{\alpha} \check{k}, 0) \qquad (s \ll 1, \alpha < 3/4) .$$
(4.58)

Since this equation depends upon s through  $s^{\alpha}$ , we will examine three cases  $0 < \alpha < 3/4$ ,  $\alpha = 0$  and  $\alpha < 0$  separately in the following.

[3-1] Inertial-diffusive range  $(0 < \alpha < 3/4)$ 

For  $0 < \alpha < 3/4$   $(k_C \ll k \ll k_K)$ , we find, because of Q(0,0) = 1, that

$$\check{\Theta}(\check{k}) = \frac{K}{3C_1} \check{k}^{-17/3} \qquad (s \ll 1, 0 < \alpha < 3/4) . \tag{4.59}$$

The dimensional form of the scalar spectrum corresponding to (4.59) is written, using (4.15) and (4.57), as

$$\Theta(k) = \frac{1}{3} K \chi \epsilon^{2/3} \kappa^{-3} k^{-17/3} . \tag{4.60}$$

Thus, we have obtained the -17/3 power law in the inertial-diffusive range. By comparing it with (1.6), we get

$$C_3 = \frac{1}{3} K = 0.572 . (4.61)$$

This relation between two universal constants  $C_3$  and K was obtained before by Batchelor et al. [13]. Qian [27] derived  $C_3 = 1.2 K$  by a statistical mechanics theory, whereas Canuto et al. [28] proposed a relation  $C_3 = 8/(27 C_1^2)$  by a turbulence model. There seems no experimental data available because of difficulty of measurements in the inertial-diffusive range. The direct and kinematic numerical simulations by Chasnov et al. [17] strongly support the relation (4.61).

[3-2] Around Kolmogorov wavenumber ( $\alpha = 0$ )

In the case of  $\alpha = 0$   $(k = O(k_{\kappa}))$ , (4.58) is written as

$$\check{\Theta}(\check{k}) = \frac{K}{3C_1} \check{k}^{-17/3} Q(\check{k}, 0) \qquad (s \ll 1, \alpha = 0) . \tag{4.62}$$

Since the function  $Q(\tilde{k},0)$  is known [6], we can draw the scalar spectrum function around  $k_{\kappa}$  (Fig.4(b)). The asymptotic form of energy spectrum  $E(k) = K\epsilon^{2/3}k^{-5/3}Q(k/k_{\kappa},0)$  in the large wavenumber limit may be proportional to  $k^3 \exp(-ck)$ , as shown from the Lagrangian DIA equation for the velocity correlation function (eq.(4.20) in ref. [6]) by the procedure described in ref. [29]. Hence, we have

$$\Theta(k) \propto k^{-1} \exp(-ck) \qquad (s \ll 1, k \gg k_{\text{K}}) . \tag{4.63}$$

[3-3] Far viscous-diffusive range ( $\alpha < 0$ )

Finally, for  $\alpha < 0 \ (k \gg k_{\scriptscriptstyle K})$ , (4.58) yields

$$\check{\Theta}(\check{k}) = 0 \qquad (s \ll 1, \alpha < 0) , \qquad (4.64)$$

which is consistent with (4.63) that the spectrum function is exponentially small at  $k \gg k_{\rm K}$ .

## E. Bump in the spectrum

Here, we discuss the bump structure which is observed around the ends of the viscous-advective range for  $s \ge 1$  (see Fig.1(b) and Fig.3(b)) and of the inertial-advective range for  $s \le 1$  (Fig.2(a) and Fig.4(a)). These may be seen more clearly in their compensated spectra in Figs.5(a) and (b). Since the end wavenumbers ( $k_B$  for  $s \ge 1$  and  $k_C$  for

 $s \leq 1$ ) of these advective ranges correspond to the beginning of the scalar dissipation, this may be understood as a bottleneck phenomenon [30] for the scalar fluctuation transfer. The scalar fluctuation cascades down throughout the advective range toward smaller scales by the interaction with the turbulent velocity field. The cascade is less effective at the end of this range because the scalar fluctuation damps in the diffusive range. This results in a pile up of the scalar fluctuation around the end of the advective range. Actually, such a bump in the scalar spectrum is observed in measurements of atmospheric boundary layer ( $s \sim 0.7$ ) by Williams and Paulson [31] and Champagne *et al.* [32], and of tidal flow ( $s \sim 9.2$ ) by Grant *et al.* [15]; the results of these measurements are collected by Hill [33].

On the other hand, we can hardly observe any bump in the scalar spectrum at the end of the inertial-diffusive range (Fig.4(b)) nor in the compensated spectrum (Fig.5(c)). Recall that the functional form of the scalar spectrum in this range is similar to that of the energy spectrum in a logarithmic scale (see Sec.IVD), and that the end wavenumber  $k_{\rm K}$  represents the beginning of the dissipation range of the velocity field but not of the scalar field. Hence, if there is a bump around  $k_{\rm K}$ , it should be due to the bottleneck effect of the energy cascade. A bump in the energy spectrum is, however, not so clearly observed in experiments (see Fig.2 in [6], for example) if it exists. The bottleneck phenomenon seems to be more effective in the passive scalar fluctuation cascade than in the energy cascade. More detailed quantitative discussions would demand a scrutiny of the three component transfer functions. Anyway, the present results on the bump structures of the spectra are qualitatively consistent with experiments.

It may be worth mentioning, in passing, that the energy spectrum of the Burgers equation with a random forcing over the whole wavenumber range and with a hyperviscosity which enhances the bottleneck effect [30], exhibits a clear bump structure [34]. The governing equation of a passive scalar and the Burgers equation have a similar property in the sense that they do not have a pressure term.

#### V. SUMMARY AND CONCLUDING REMARKS

We have shown that a closed set of equations for the passive scalar correlation function in the Lagrangian DIA formulation [6] is identical to that by LRA [5,20,21], and that it is completely consistent with the phenomenological theories in the inertial-advective range by Obukhov-Corrsin [10,11], in the inertial-diffusive range by Batchelor et al. [13] and in the viscous-advective range by Batchelor [12]. This quite simple Lagrangian closure is, therefore, excellently successful in making a bridge between the phenomenological theories and the basic equations in describing the power spectra of both the velocity and passive scalar fields in isotropic turbulence. In addition, the universal forms of the scalar spectrum in the statistically stationary state have been determined numerically for moderate Schmidt numbers s, and analytically for  $s \ll 1$  and  $s \gg 1$ . The appearance of the bump structure in these universal forms of the spectrum is qualitatively consistent with experimental data. The universal constant  $C_3$  in the inertial-diffusive range is in a good agreement with the numerical simulations by Chasnov et al. [17]. However, the constants in the advective ranges are only about half the experimental data, although the present predictions are closer to the experiments than

the ones by the abridged LHDIA.

This failure in the estimation of the universal constants in the advective range of the scalar spectrum, despite of many successful predictions in other aspects of both the scalar and velocity spectra, should be accepted as a severe manifestation of the incompleteness of the present Lagrangian closure. A reconsideration of the structure of the theory itself, especially, a systematic check of the underlying assumptions should be necessary. This theory is based upon several working assumptions which are summarized as (I)—(III) in Sec. III B for the passive scalar field and in Sec.3.2 of [6] for the velocity field. Some of the assumptions, which correspond to (I) and (II) in Sec. III B, have been checked positively for a model equation [7]. Needless to say, however, it is necessary to check them for the Navier-Stokes system itself. By taking account of this partial support of the assumptions (I) and (II), we currently speculate the propriety of the third assumption of the independency between the NDI fields of the position function and of the Euler fields.

A thorough examination of all the assumptions employed in the present Lagrangian DIA for the Navier-Stokes system and a reformulation with a possible correlation between the position function and the Eulerian quantities still remain as important future works.

#### APPENDIX A

Derivations of (3.19) and (3.20) are described here. First, by substituting (2.18) into (2.21), we obtain

$$\widetilde{Z}(\boldsymbol{k},t,t') = \frac{(2\pi)^9}{L^6} \sum_{\boldsymbol{k'}} \overline{\widetilde{\theta}(\boldsymbol{k'},t)} \, \widetilde{\psi}(-\boldsymbol{k'},t|\boldsymbol{k},t') \, \widetilde{\theta}(-\boldsymbol{k},t') \,, \tag{A1}$$

which is further converted, by substitutions of the direct-interaction decompositions, into

$$\widetilde{Z}(\boldsymbol{k},t,t') = \frac{(2\pi)^9}{L^6} \sum_{\boldsymbol{k'}} \overline{\widetilde{\theta}^{(0)}(\boldsymbol{k'},t||\boldsymbol{k}_0,\boldsymbol{p}_0,\boldsymbol{q}_0)} \, \overline{\widetilde{\psi}^{(0)}(-\boldsymbol{k'},t||\boldsymbol{k},t'||\boldsymbol{k}_0,\boldsymbol{p}_0,\boldsymbol{q}_0)} \, \overline{\widetilde{\theta}^{(0)}(-\boldsymbol{k},t'||\boldsymbol{k}_0,\boldsymbol{p}_0,\boldsymbol{q}_0)}$$
(A2)

(see assumption (III) in Sec.III B). Then, a combination of this equation and

$$\overline{\widetilde{\psi}^{(0)}(\boldsymbol{k},t|\boldsymbol{k}',t')} = \frac{L^3}{(2\pi)^6} \,\delta^3_{\boldsymbol{k}+\boldsymbol{k}'} \tag{A3}$$

(see (3.6) in [6] and discussions in §2 of ref. [5]) yields (3.19). As for (3.20), we take the functional derivative of (2.18) to obtain

$$\overline{\widetilde{G}^{(L)}(t|\boldsymbol{k}, -\boldsymbol{k}, t')} = \frac{(2\pi)^6}{L^3} \sum_{\boldsymbol{k}'} \overline{\widetilde{G}(\boldsymbol{k}', t| -\boldsymbol{k}, t')} \, \widetilde{\psi}(-\boldsymbol{k}', t|\boldsymbol{k}, t') . \tag{A4}$$

By substituting the direct-interaction decompositions into the right-hand side of the above equation and by taking (A3) into account, we find (3.20).

#### APPENDIX B

We prove (4.13) based upon the LRA-DIA equation. The right-hand side of (4.8) gives the transfer function (2.27) in the inertial-advective range as

$$T(k) = KW_1 \nu k_K^{4/3} k \iint_{\Delta_k} dp \, dq \, \sigma(k, p, q) \, p^{-10/3} \, q \left[ -\Theta(k) + \left(\frac{k}{q}\right)^2 \, \Theta(q) \right]. \tag{B1}$$

The scalar flux function (2.28) is then written as

$$\Pi(k) = KW_1 \nu k_{\kappa}^{4/3} \int_{k}^{\infty} dk' k' \iint_{\triangle_{k'}} dp \, dq \, \sigma(k', p, q) \, p^{-10/3} \, q \left[ -\Theta(k') + \left(\frac{k'}{q}\right)^2 \, \Theta(q) \right]$$

$$= KW_1 \nu k_{\kappa}^{4/3} \int_{k}^{\infty} dk' k' \iint_{\substack{p < k \text{ or } q < k}} dp \, dq \, \sigma(k', p, q) \, p^{-10/3} \, q \left[ -\Theta(k') + \left(\frac{k'}{q}\right)^2 \, \Theta(q) \right] , \tag{B2}$$

where we have used the property of the detailed balance of the nonlinear transfer of the scalar fluctuation, i.e.,

$$\int_{k}^{\infty} dk' k' \iint_{\substack{D_{\Delta_{k'}} \\ p,q > k}} dp \, dq \, \sigma(k', p, q) \, p^{-10/3} \, q \left[ -\Theta(k') + \left(\frac{k'}{q}\right)^2 \, \Theta(q) \right] = 0 . \tag{B3}$$

Substitution of the inertial-advective power spectrum (1.4) into (B2) gives

$$\Pi(k) = KW_1 C_1 \chi \int_{k}^{\infty} dk' \iint_{\substack{p < k \text{ or } q < k}} dp \, dq \, \sigma(k', p, q) \, k' \, p^{-10/3} \, q \, \left[ k'^{-5/3} - k'^2 \, q^{-11/3} \right] 
= KW_1 C_1 \chi \int_{1}^{\infty} dk' \iint_{\substack{p < 1 \text{ or } q < 1}} dp \, dq \, \sigma(k', p, q) \, k' \, p^{-10/3} \, q \, \left[ k'^{-5/3} - k'^2 \, q^{-11/3} \right] 
= KW_1 C_1 \chi I ,$$
(B4)

where

$$I = \left[ \int_{1}^{\infty} dk' \int_{0}^{1} dp \int_{k'-p}^{k'+p} dq - \int_{1}^{2} dk' \int_{k'/2}^{1} dp \int_{k'-p}^{p} dq \right]$$

$$\times \left[ \sigma(k', p, q) \, k' \, p^{-10/3} \, q \left[ k'^{-5/3} - k'^{2} \, q^{-11/3} \right] + (\text{similar term } p \leftrightarrow q) \right]$$

$$= \frac{729}{910} \int_{0}^{\infty} \frac{dx}{x^{1/3}(x+1)} - \frac{2187}{910} \int_{0}^{1} \frac{dx}{x^{2} + x + 1} = \frac{729\pi}{910\sqrt{3}}.$$
(B5)

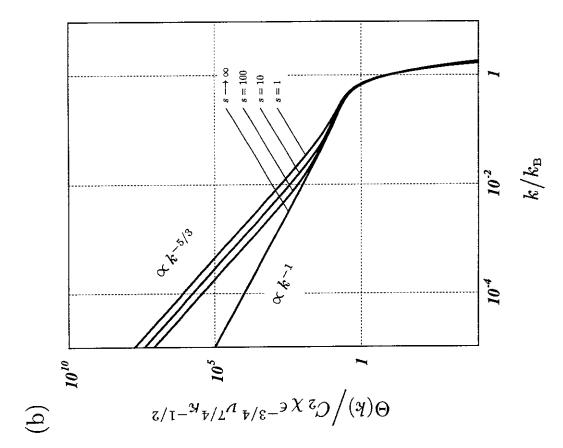
Since  $\Pi = \chi$  in the advective range, we obtain (4.13).

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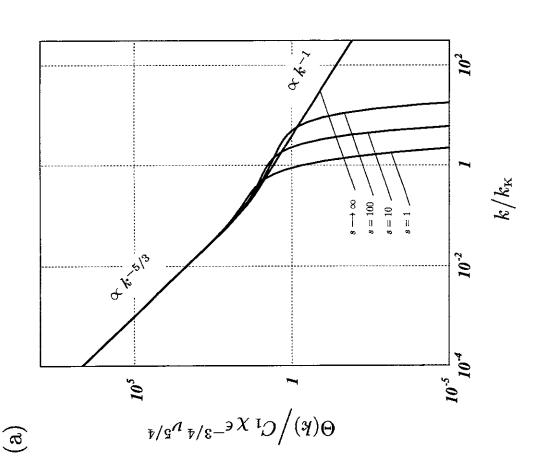
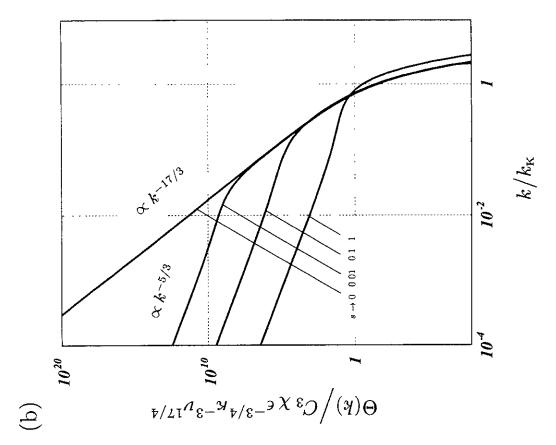


Fig.1 Passive scalar spectra in stationary isotropic turbulence for  $s \ge 1$ . The wavenumber is normalized by (a) the Kolmogorov wavenumber  $k_{\rm k}$  and (b) the Batchelor wavenumber  $k_{\rm b}$ .



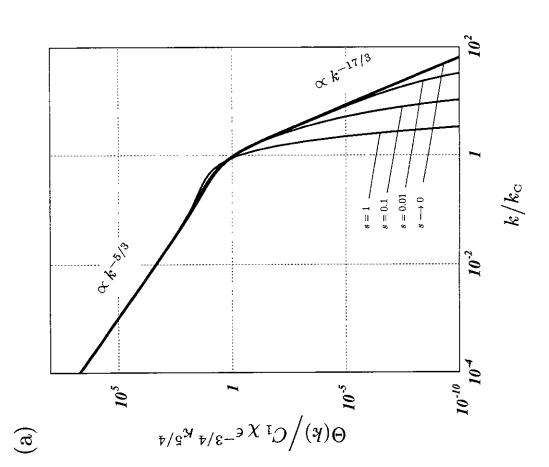


Fig.2 Same as Fig.1 for  $s \le 1$ . The wavenumber is normalized by (a) the Obukhov-Corrsin wavenumber  $k_c$  and (b) the Kolmogorov wavenumber  $k_\kappa$ .

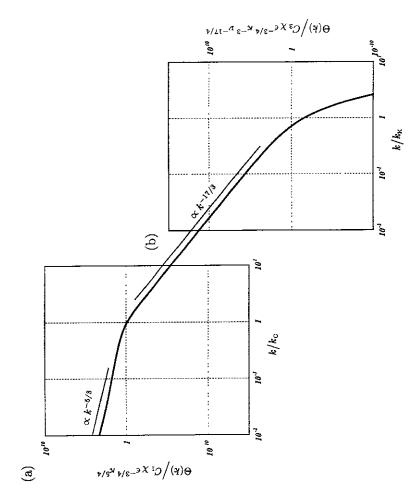


Fig.4 Passive scalar spectrum in stationary isotropic turbulence around (a)  $k_{\rm c}$  and (b)  $k_{\rm k}$  for  $s\ll 1$ .

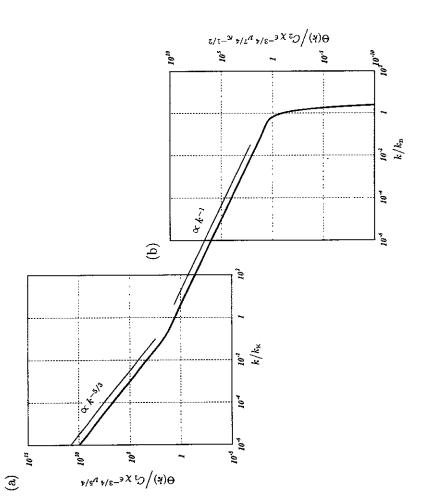


Fig.3 Passive scalar spectrum in stationary isotropic turbulence around (a)  $k_{\rm R}$  and (b)  $k_{\rm B}$  for  $s\gg 1$ .

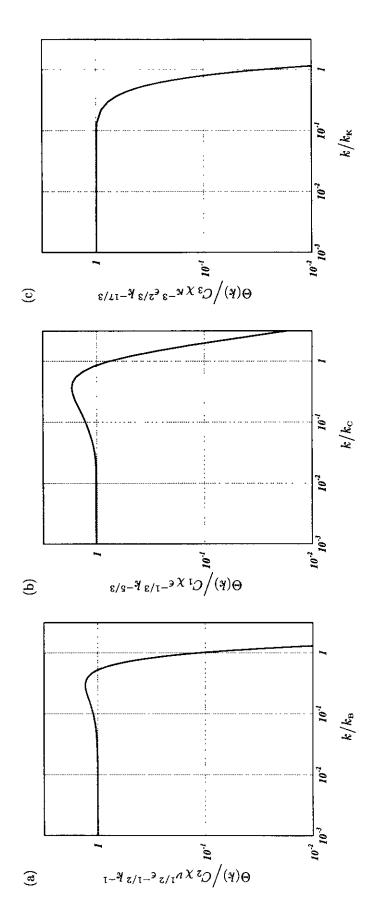


Fig.5 Compensated passive scalar spectra in stationary isotropic turbulence (a) around  $k_{\rm B}$  for  $s \gg 1$ , (b) around  $k_{\rm C}$  for  $s \ll 1$  and (c) around  $k_{\rm K}$  for  $s \ll 1$ .

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