Nonlinear Lagrangian equations for turbulent motion and buoyancy in inhomogeneous flows

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Linear and nonlinear Lagrangian equations are derived for stochastic processes that appear as solutions of the averaged hydrodynamic equations, since their moments satisfy the budgets given by these equations. These equations include the potential temperature, so that non-neutral flows can be described. They will be compared with nonlinear and non-Markovian equations that are obtained using concepts of nonequilibrium statistical mechanics. This approach permits the description of turbulent motion and buoyancy, where memory effects and driving forces with arbitrary colored noise may occur. The equations depend on assumptions that concern the dissipation and pressure redistribution. In the approximations of Kolmogorov and Rotta for these terms, the dissipation time scale remains open, which can be determined by the calculation of the production–dissipation ratio of turbulent kinetic energy. The features of these equations are illustrated by the calculation of turbulent states in the space of invariants.
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I. INTRODUCTION

The averaged hydrodynamic equations (AHE) define budgets at points of the flow for moments of different order of the distribution function of the fluctuating hydrodynamic variables. Lagrangian models for the motion of fluid particles in high-Reynolds number turbulent flows may determine stochastic processes that represent solutions of the AHE, when the dissipation and pressure redistribution terms are modelled in these budget equations. This means that the moments of these stochastic processes obey these budgets, e.g., up to second order without the need for any assumptions about the spatial gradients of the third moments (the turbulent transport terms). Such Lagrangian models are used to study the realizability of solutions of second-order models,[1,2] and they are successfully applied to calculate the turbulent diffusion of passive tracers in complex flows.[3–6] The turbulent dispersion can be described in non-neutral flows, if the potential temperature is included in a stochastic Lagrangian description of particle motion.[7] Such Lagrangian equations, which are linear in the particle velocities and potential temperature (but nonlinear in the particle position), can be derived completely consistent with the AHE up to second order,[8] where the dissipation and pressure redistribution terms are taken in the approximations of Kolmogorov[9] and Rotta,[10] respectively. But here the problem of nonuniqueness arises (which is known for neutral flows),[1,2,4,5] which means the consistency with the AHE does not uniquely determine the stochastic Lagrangian equations. The assessment of consequences of differences in Lagrangian models satisfying the same AHE poses a difficult problem, but it may be expected that these differences influence considerably the calculated features of particle motion in complex flows (i.e., under conditions where the effects of inhomogeneities and anisotropy have to be considered). Moreover, when the approximations of Kolmogorov and Rotta are used for the dissipation and pressure redistribution, a dissipation time scale appears in the Lagrangian equations, which has to be estimated. The calculation of this time scale is an important problem, because it determines the production–dissipation ratio for the turbulent kinetic energy (TKE) and therefore the local character of the energy transfer.

The consistency between stochastic Lagrangian equations and the AHE is considered at first with respect to linear equations for the particle velocity and potential temperature, where, in particular, the description of the potential temperature by a stochastic differential equation is discussed. The same problem is considered for nonlinear Markovian equations in the third section, where the nonuniqueness problem is discussed for non-neutral flows. This investigation of the suitability of stochastic processes to appear as solutions of the AHE is compared in the fourth section with another approach, where stochastic differential equations are derived by applying concepts of nonequilibrium statistical mechanics. This explains in which way memory effects enter into the Lagrangian equations. As an example, Sawford's equation[5] for the acceleration of a fluid particle is derived, which takes colored noise in the velocity equation into account. The influence of (vertical) gradients of the mean (horizontal) flow velocity and potential temperature on particle motion is considered in the fifth section. This is done by considering the relation between the unknown time scale, which arises by the parametrization of the dissipation (next section), and the wind shear and stratification. Through this relation, the Lagrangian equations that are derived in the second section depend (for balanced ratios of production and dissipation of TKE and heat) only upon three flow numbers. These equations reflect nonaveraged hydrodynamic equations in a scale, where the Lagrangian acceleration correlation is small (for time lags much longer than the Kolmogorov time scale)[11]

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and the three flow number play the role of the molecular constants as parameters.

II. LINEAR MARKOVIAN EQUATIONS

Let us first consider linear Markovian equations for the description of fluid particle motion and the change of the potential temperature of particles. Whereas there are good reasons to treat the velocity of a fluid particle as a Markov process in high-Reynolds number flows because of the structure of the acceleration correlation,[3,4,11] the description of the potential temperature by a stochastic process is not comparably investigated. To describe the important part of buoyancy effects on the turbulent dispersion, it was supposed by Zanetti and Al-Madani[12] and Cogan[13] that the potential temperature also satisfies a stochastic differential equation. Van Dop investigated this approach in a more fundamental way.[7] He pointed out the conceptional problem of assigning a potential temperature to a fluid particle. Nevertheless, this approach is continued here, which means a stochastic differential equation is assumed for the potential temperature of a particle. According to inertial subrange theory, the structure function and the autocorrelation of the temperature would be expected as proportional to the time lag and exponential, respectively.[7] The reproduction of these inertial subrange relations can be found, if the potential temperature is described by a stochastic differential equation. As shown below, this approach guarantees, e.g., that the AHE are fulfilled up to second order. These equations are able to explain well the basic features of buoyancy effects in the turbulent dispersion.[7,14] The available experimental evidence tends to indicate that the probability density function of an inert dynamically passive scalar convected by homogeneous turbulence evolves asymptotically toward a Gaussian shape.[15] This is supported by results of direct numerical simulations.[16] At least for an approximately neutral stratification with a negligible influence of the potential temperature on the turbulence, this quantity may be described by a linear stochastic differential equation providing a Gaussian distribution. The idea of a fluid particle with a potential temperature represents a reference picture for the real process in correspondence with the AHE and permits the description of the basic characteristics of buoyancy processes. This is equivalent to approaches employed in the theory of turbulent mixing, where models are used that are not directly related to a physical process, but are a good characterization of essential features of the considered phenomena.[15,17,18] Turbulent motion and buoyancy are described by the same structure of equation in this way. This is particularly advantageous for the comparison with the AHE, as discussed below.

The turbulent flow is regarded as a whole of fluid particles, each having a constant mass. The (time-dependent) total mass of the fluid results from particle mass times the (time-dependent) total number of fluid particles. Neglecting chemical reactions, each particle is characterized at the time t by its position $\mathbf{x}_L(t)$, velocity $\mathbf{U}_L(t)$ [vectors with components $x_L^I(t)$ and $U_L^I(t)$, where $I=1,2,3$ and the subscript L denotes a Lagrangian quantity] and potential temperature $\Theta_L(t)$. Particle density and volume change in time according to a state equation. With respect to comparisons with the AHE, it is advantageous to combine the particle velocity $\mathbf{U}_L(t)$ and potential temperature $\Theta_L(t)$ to the four-dimensional state vector $\mathbf{Z}_L(t)=[\mathbf{U}_L(t),\Theta_L(t)]$. Assuming $[\mathbf{x}_L(t), \mathbf{Z}_L(t)]$ as a Markov process and only linear fluctuations of the state $\mathbf{Z}_L(t)$, these quantities change according to[19,20]

$$\frac{d}{dt}\, x_L^I(t)=Z_L^I(t), \tag{1a}$$

$$\frac{d}{dt}\, Z_L^i(t)=\langle a^i\rangle+G^{ij}(Z_L^j-\langle Z_E^j\rangle)+b^{ij}\,\frac{dW^j}{dt}, \tag{1b}$$

where the small superscripts run from 1 to 4 in contrast to capitals which run from 1 to 3 and summation over repeated superscripts is assumed. The first two terms in (1b) give the systematic particle motion with unknown coefficients $\langle a^i\rangle$ and G^{ij}, where the ensemble average is denoted by $\langle\cdots\rangle$. The ensemble averages of Eulerian quantities (subscript E) are estimated at fixed positions $\mathbf{x}$, which are replaced by $\mathbf{x}=\mathbf{x}_L(t)$ in the equations. Consequently, these equations are linear in the state $\mathbf{Z}_L$ but may be nonlinear in the position $\mathbf{x}_L$. The last term of (1b) describes the influence of a stochastic force, characterized by the white noise dW^j/dt and a matrix b with elements b^{ij}. Here, dW^j/dt is a Gaussian process having a vanishing mean and uncorrelated values to different times, $\langle dW^i/dt\rangle=0$ and, $\langle dW^i/dt(t)\cdot dW^j/dt(t')\rangle=\delta_{ij}\delta(t-t')$, δ_{ij} denotes the Kronecker delta, and $\delta(t-t')$ is the delta function. Instead of considering the equations (1a) and (1b) for the stochastic transport of particles and their changing properties, the equivalent Fokker–Planck equation can be considered for the probability density to find given values of particle properties at given locations and times. The Lagrangian joint mass density function will be denoted by F_L. This function is similar to the corresponding probability density function, but normalized to the mean concentration $\langle c(\mathbf{x},t)\rangle$ of considered particles (which are emitted, e.g., by a source),

$$\int d\mathbf{Z}\; F_L(\mathbf{Z},\mathbf{x},t)=\langle c(\mathbf{x},t)\rangle. \tag{2}$$

The transport equation of F_L can be derived by different methods[17] and is given in correspondence with the stochastic differential equations (1a) and (1b) by the equation

$$\frac{\partial F_L}{\partial t}+\frac{\partial}{\partial x^I}\, Z^I F_L=-\frac{\partial}{\partial Z^i}\,[\langle a^i\rangle+G^{ij}(Z^j-\langle Z_E^j\rangle)]F_L+\frac{\partial^2}{\partial Z^i\,\partial Z^j}\,B^{ij}F_L, \tag{3}$$

where $B^{ij}=1/2b^{ik}b^{kj}$ is introduced. This coefficient B^{ij} is given by[7]

$$B=\frac{1}{4\tau}\begin{pmatrix} C_0q^2 & 0 & 0 & 0\\ 0 & C_0q^2 & 0 & 0\\ 0 & 0 & C_0q^2 & 0\\ 0 & 0 & 0 & C_1\langle(Z_E^4-\langle Z_E^4\rangle)^2\rangle \end{pmatrix}, \tag{4}$$

if Kolmogorov's theory[9] is adopted for a high-Reynolds number flow and the time scale for the dissipation of the potential temperature variance is assumed to be of the same order as that for the dissipation of TKE, which means the unknown constants C_0 and C_1 are considered as being of the same order (which is discussed below). In (4), $q^2 = \langle (Z_E^I - \langle Z_E^I \rangle)(Z_E^I - \langle Z_E^I \rangle) \rangle$ is twice the TKE (with $I=1, 2, 3$), and $\tau = q^2/(2\langle \epsilon \rangle)$ is the (unknown) time scale of dissipation of TKE relating q^2 and the mean dissipation rate $\langle \epsilon \rangle$ of TKE. By F_L, the statistical properties of an ensemble of observed particles are determined at a fixed point. The corresponding mass density function of all fluid particles is denoted by the (Eulerian) function F, which is normalized to the averaged fluid density ρ,

$$\int d\mathbf{Z}\, F(\mathbf{Z},\mathbf{x},t) = \langle \rho(\mathbf{x},t) \rangle. \tag{5}$$

This mass density also has to fulfill the transport equation (3). This relation of F with the unknown coefficients $\langle a^i \rangle$ and G^{ij} can be used for deriving consistency constraints between these coefficients and Eulerian means and variances of the velocity and potential temperature fields. The transport equations for the mean values of the wind and potential temperature fields can be derived by replacing F_L by F in (3), multiplying this relation with Z^i, and integrating over $\mathbf{Z}$. Then, $\langle a^i \rangle$ is determined by

$$\frac{D\langle Z_E^i \rangle}{Dt} + \frac{\partial V^{iL}}{\partial x^L} = \langle a^i \rangle, \tag{6}$$

where the abbreviation $D/Dt = \partial/\partial t + \partial/\partial x^K \cdot \langle Z_E^K \rangle$ is used and the matrix of second moments of the coupled wind and potential temperature field is written by

$$V = \begin{pmatrix} \langle u^1 u^1 \rangle & \langle u^1 u^2 \rangle & \langle u^1 u^3 \rangle & \langle u^1 \theta \rangle \\ \langle u^2 u^1 \rangle & \langle u^2 u^2 \rangle & \langle u^2 u^3 \rangle & \langle u^2 \theta \rangle \\ \langle u^3 u^1 \rangle & \langle u^3 u^2 \rangle & \langle u^3 u^3 \rangle & \langle u^3 \theta \rangle \\ \langle \theta u^1 \rangle & \langle \theta u^2 \rangle & \langle \theta u^3 \rangle & \langle \theta^2 \rangle \end{pmatrix}, \tag{7}$$

with $z^k = Z_E^k - \langle Z_E^k \rangle$ for the fluctuations. Accordingly, by multiplication of (3) (with F instead of F_L) with $Z^i Z^j$ and integration over $\mathbf{Z}$, the transport equations for the second moments can be derived, which read as

$$\frac{DV^{ij}}{Dt} + R^{ij} = -P^{ij} + G^{ik}V^{kj} + G^{jk}V^{ki} + \frac{C_0 q^2}{2\tau}\,\delta_{ij} - \frac{C_0 q^2 - C_1 V^{44}}{2\tau}\,\delta_{i4}\delta_{j4}. \tag{8}$$

The gradients of triple correlations are denoted by $R^{ij} = \partial\langle z^K z^i z^j \rangle/\partial x^K$ and the production is written as $P^{ij} = \langle z^K z^i \rangle \partial\langle Z_E^j \rangle/\partial x^K + \langle z^K z^j \rangle \partial \langle Z_E^i \rangle/\partial x^K$. These equations (6) and (8) will be compared with the corresponding Eulerian budget equation of first and second order. Using the Boussinesq approximation and the incompressibility constraint, $\partial Z_E^K/\partial x^K = 0$, the conservation equations of momentum and potential temperature read as

$$\frac{\tilde{D} Z_E^i}{Dt} = \nu \frac{\partial^2 Z_E^i}{\partial x^K\, \partial x^K} + (\alpha - \nu)\frac{\partial^2 Z_E^4}{\partial x^K\, \partial x^K}\,\delta_{i4} - \langle \rho \rangle^{-1} \frac{\partial p}{\partial x^K}\,\delta_{Ki} - g[1 - \beta(Z_E^4 - \langle Z_E^4 \rangle)]\delta_{i3}, \tag{9}$$

where $\tilde{D}/Dt = \partial/\partial t + \partial/\partial x^K \cdot Z_E^K$, ν is the kinematic viscosity, α is the coefficient of molecular heat transfer, β is the thermal expansion coefficient, p is the pressure, and g is the acceleration due to gravity. Consequently, $\langle a^i \rangle$ is determined by the averaged right-hand side of (9),

$$\langle a^i \rangle = \nu \frac{\partial^2 \langle Z_E^i \rangle}{\partial x^K\, \partial x^K} + (\alpha - \nu)\frac{\partial^2 \langle Z_E^4 \rangle}{\partial x^K\, \partial x^K}\,\delta_{i4} - \langle \rho \rangle^{-1}\frac{\partial \langle p \rangle}{\partial x^K}\,\delta_{Ki} - g\,\delta_{i3}. \tag{10}$$

By modelling the dissipation according to Kolmogorov's theory[9] and supposing a return-to isotropy pressure redistribution according to Rotta,[10] the transport equations of second order can be derived from the conservation equations. These equations read[21] as

$$\frac{DV^{ij}}{Dt} + R^{ij} = -P^{ij} + \left(-\frac{k_1}{4\tau}\,\delta_{ik} + \frac{k_1 - k_3}{2\tau}\,\delta_{i4}\delta_{k4} + \beta g\,\delta_{i3}\delta_{k4}\right)V^{kj} + \left(-\frac{k_1}{4\tau}\,\delta_{jk} + \frac{k_1 - k_3}{2\tau}\,\delta_{j4}\delta_{k4} + \beta g\,\delta_{j3}\delta_{k4}\right)V^{ki} + k_2 q^2 \frac{\partial \langle Z_E^L \rangle}{\partial x^K}(\delta_{Li}\delta_{Kj} + \delta_{Lj}\delta_{Ki}) + \frac{q^2}{2\tau} \cdot \frac{k_1 - 2}{3}\,\delta_{ij} - \frac{(k_1 - 2)/3 \cdot q^2 - (2k_3 - 2k_4 - k_1)V^{44}}{2\tau}\delta_{i4}\delta_{j4}, \tag{11}$$

where the closure parameters k_1, k_2, k_3, and k_4 are introduced. The parameters k_1 k_2 and k_3 arise from the return-to-isotropy theory of Rotta. Because of its simplicity, this approximation is well suited for the illustration of the approach. Whereas the Kolmogorov approximation $\nu\langle \partial z^I/\partial x^K \cdot \partial z^J/\partial x^K \rangle = \frac{1}{3}\langle \epsilon \rangle \delta_{IJ}$ for the dissipation of TKE is widely accepted, some remarks are needed to the formally corresponding assumption[21,22] for $\alpha\langle \partial z^4/\partial x^K \cdot \partial z^4/\partial x^K \rangle = \langle \epsilon_{\theta\theta} \rangle = k_4 \langle \epsilon \rangle q^{-2} V^{44}$ for the dissipation of the potential temperature variance. In investigations of decaying grid turbulence it was shown that the closure parameter k_4 introduced in this way cannot be considered as a universal constant. In dependence on initial conditions, a range of $0.6 < k_4 < 3.1$ was found for this quantity, and variations were observed over the length of the wind tunnel.[23–25] When a time scale $\tau_\theta = V^{44}/(2\langle \epsilon_{\theta\theta} \rangle)$ is introduced analogous to $\tau = q^2/(2\langle \epsilon \rangle)$, this closure supposes the proportionality of both time scales, $\tau_\theta = \tau/k_4$. As stated by Pope,[17] this assumed proportionality can be expected to be less unrealistic for shear flows in which potential temperature and wind fields share a common history and common boundary conditions. This argument supports the parametrization (4) of the coefficient B (which requires the assumption of time scales of the same order for the dissipation of TKE and the potential temperature variance, i.e. C_0 and C_1 of order unity), but this proportionality cannot be expected generally.[23] We see by comparing the transport equations (8) and (11) for the variances V^{ij} to estimate the unknown coefficients G^{ij}, that in this way only conclusions to the symmetric component $\frac{1}{2}[(GV)^{ij}+(GV)^{ji}]$ of $(GV)^{ij}$ can be drawn. This comparison reveals that the equations (11) are consistent with the budget equations (8) for the variances, if

$$G^{ij}=-\frac{k_1}{4\tau}\,\delta_{ij}+\frac{k_1-k_3}{2\tau}\,\delta_{i4}\delta_{j4}+\beta g\,\delta_{i3}\delta_{j4}+\left[\frac{k_2 q^2}{2}\cdot\frac{\partial\langle Z_{\mathrm{E}}^{L}\rangle}{\partial x^K}\,[\delta_{\mathrm{L}\,i}\delta_{Km}+\delta_{\mathrm{L}\,j}\delta_{Km}]\right.$$
$$\left.+\frac{1}{2}\cdot A^{im}+\frac{q^2}{4\tau}\cdot\left(\frac{k_1-2}{3}-C_0\right)\cdot\delta_{im}-\frac{[(k_1-2)/3-C_0]\cdot q^2-(2k_3-2k_4-k_1-C_1)\cdot V^{44}}{4\tau}\delta_{i4}\delta_{m4}\right]\cdot(V^{-1})^{mj}, \tag{12}$$

where A is any antisymmetric matrix. The simplest choice of G^{ij} is obtained by setting $A^{im}=0$, $k_2=0$ (Sec. V), $C_0=(k_1-2)/3$, and $C_1=2k_3-2k_4-k_1$. This brings for the matrix G,

$$G=-\frac{1}{4\tau}\cdot\begin{pmatrix} k_1 & 0 & 0 & 0\\ 0 & k_1 & 0 & 0\\ 0 & 0 & k_1 & -4\beta g\tau\\ 0 & 0 & 0 & 2k_3-k_1\end{pmatrix}, \tag{13}$$

which means G becomes independent of the variances V. The nonuniqueness of the estimation of the coefficient matrix G (by the appearance of the unknown antisymmetric matrix A) is well known for neutral flows.[1,2] This problem will be discussed in the next section with respect to nonlinear equations. Instead, let us come back to the justification of the description of the potential temperature by a stochastic differential equation at the beginning of this section. When $C_1=0$ is set such that no stochastic forcing appears in the Lagrangian equation (1b) for the potential temperature, we find from (12) nonvanishing contributions for the coefficients G^{4K} with $K=1$, 2, 3 caused by the last term on the right-hand side of (12). This means that the change of potential temperature is influenced by velocity fluctuations in general and consequently also by stochastic influences. Such stochastic forces explicitly appear by setting $C_1=2k_3-2k_4-k_1$, but the advantage of this choice is the possibility to describe G^{ij} by the simple expression (13), which is independent of the variances V^{ij}. Then, G^{ij} depends only over the time scale τ on the state of the flow. The estimation of τ is considered in the fifth section, where its relations with wind shear and temperature stratification are investigated.

III. NONLINEAR MARKOVIAN EQUATIONS

Lagrangian equations can be found that are consistent with the AHE up to second order, as shown in the previous section. These equations are linear in the particle state, but may be nonlinear in the particle position. It remains open under which conditions nonlinear velocity and potential temperature fluctuations have to be considered in the Lagrangian equations and how the consistency between the Lagrangian equations and the AHE can be guaranteed with respect to higher moments. This is investigated now by assuming a Markovian process $(\mathbf{x}_{\mathrm{L}}(t),\mathbf{Z}_{\mathrm{L}}(t))$ as above. Through these equations, the relation emerges more distinct between the above considered linear equations and more complicated non-Markovian equations, which are derived in the next section. Additionally, the nonuniqueness problem appears here for non-neutral flows in a more general formulation as before. The particle motion and the potential temperature are described by the nonlinear (Ito) stochastic differential equation,[19,20]

$$\frac{d}{dt}\,x_{\mathrm{L}}^{I}(t)=Z_{\mathrm{L}}^{I}(t), \tag{14a}$$

$$\frac{d}{dt}\,Z_{\mathrm{L}}^{i}(t)=-\gamma^{i}(\mathbf{Z}_{\mathrm{L}},\mathbf{x}_{\mathrm{L}},t)+b^{ij}(\mathbf{Z}_{\mathrm{L}},\mathbf{x}_{\mathrm{L}},t)\,\frac{dW^{j}}{dt}\,(t). \tag{14b}$$

The transport equation for F_{L} that replaces (3) for the nonlinear equation system (14a) and, (14b) reads as

$$\frac{\partial F_{\mathrm{L}}}{\partial t}+\frac{\partial}{\partial x^K}\,Z^K F_{\mathrm{L}}=\frac{\partial}{\partial Z^i}\,\gamma^i(\mathbf{Z},\mathbf{x},t)F_{\mathrm{L}}+\frac{\partial^2}{\partial Z^i\,\partial Z^j}\,B^{ij}(\mathbf{Z},\mathbf{x},t)F_{\mathrm{L}}, \tag{15}$$

where again $B^{ij}=1/2b^{ik}b^{kj}$, which is determined by (4) as above in the linear model, so that the systematic transport coefficient γ remains as a unknown function to be estimated. In the previous section this coefficient was assumed to be a linear function in $\mathbf{Z}-\langle\mathbf{Z}_{\mathrm{E}}\rangle$ and the drift parameter $\langle\mathbf{a}\rangle$ as well as the coefficient G of linear fluctuations had been estimated by the consistency constraint with the AHE up to second order in the approximations of Kolmogorov and Rotta. The hydrodynamic relations for higher moments can be included into this consistency investigation by considering an equation for the mass density function F, from which all moments can be calculated. The most general transport equation for F was derived from Pope,[17] which has the structure of (15) on the left-hand side (with $F_{\mathrm{L}}=F$) and a right-hand side, which can be written as $\partial[h^i(\mathbf{Z},\mathbf{x},t)\cdot F]/\partial Z^i$. The function $h_i(\mathbf{Z},\mathbf{x},t)$ represents a conditional ensemble average at fixed $\mathbf{Z}$ of the gradients of the mean pressure and pressure fluctuations and of the molecular stress tensor and gravity acceleration. This structure of the equation for F is simply a consequence of the hydrodynamic conservation equations for mass, momentum, and heat. Thus, by adopting the approximation of Kolmogorov, the nonlinear equation (15) corresponds with the general equation structure of the mass density function F, and the coefficient γ is a function to be determined in dependence on the gravity acceleration, the mean pressure gradient, and in particular the correlations of pressure fluctuations. Consequently, the model (15) is consistent with the infinite hierarchy of AHE in the approximation of Kolmogorov, if F appears as a possible solution of (15). This condition was proposed by Thomson[4] and provides a relationship between the systematic transport coefficient γ and the Eulerian mass density F. It may be written as

$$\gamma^i=-F^{-1}\left(\frac{\partial}{\partial Z^j}\,B^{ij}F+\Phi^i\right), \tag{16a}$$

where a function $\mathbf{\Phi}$ is introduced that satisfies

$$\frac{\partial\Phi^i}{\partial Z^i}=-\frac{\partial F}{\partial t}-\frac{\partial}{\partial x^K}\,Z^K F, \tag{16b}$$

and $\Phi^i\rightarrow 0$ for $|Z|\rightarrow\infty$. By the relations (16a), (16b), and (4), the coefficients γ and B are determined and the nonlinear Lagrangian equations are found to be consistent with the AHE in the Kolmogorov approximation. However, for inhomogeneous flows not much is known about the mass density function F, which is required for these estimation of γ and B. Only partial information is available for real flows in terms of the moments of lower order of this distribution function. How this information can be applied to construct F as a maximum missing information density function was investigated, e.g., by Du *et al.*[26] But even for given F, the rotation of $\mathbf{\Phi}$, rot $\mathbf{\Phi}$, remains open according to (16b), as the antisymmetric component of $G^{ik}V^{kj}$. To get more insight into this finding, let us consider as an example the mass density function F in a simple approximation as local Gaussian in the state $\mathbf{Z}$,[27] which means

$$F=\frac{\langle\rho\rangle}{(2\pi)^2\det V}\exp\left(-\frac{1}{2}\,(Z^k-\langle Z_{\mathrm{E}}^k\rangle)(V^{-1})^{kl}\right.$$
$$\left.\times(Z^1-\langle Z_{\mathrm{E}}^1\rangle)\right), \tag{17}$$

where $\det V$ is the determinant of V. Taking B^{ij} in correspondence with (4) as independent of $\mathbf{Z}$, we obtain for the equation (14b),

$$\frac{d}{dt}\,Z_{\mathrm{L}}^i(t)=\langle a_{\mathrm{NL}}^i\rangle+G_{\mathrm{NL}}^{ij}(Z_{\mathrm{L}}^j-\langle Z_{\mathrm{E}}^j\rangle)+H^{ijk}(Z_{\mathrm{L}}^j-\langle Z_{\mathrm{E}}^j\rangle)$$
$$\times(Z_{\mathrm{L}}^k-\langle Z_{\mathrm{E}}^k\rangle)+b^{ij}\cdot\frac{dW^j}{dt}, \tag{18}$$

where (16a), and (16b) are used, $\partial\langle Z_{\mathrm{E}}^K\rangle/\partial x^K=0$ is applied, and the subscript NL of the first two terms on the right-hand side denotes quantities in this nonlinear approach, in contrast to the corresponding ones in the linear approach. Here,

$$\langle a_{\mathrm{NL}}^i\rangle=\frac{D\langle Z_{\mathrm{E}}^i\rangle}{Dt}+(1-\lambda)\,\frac{\partial V^{iK}}{\partial x^K}-\frac{1-2\lambda}{2}$$
$$\cdot V^{iK}\,\frac{\partial V^{jl}}{\partial x^K}\,(V^{-1})^{lj}, \tag{19a}$$

$$G_{\mathrm{NL}}^{ij}=\left\{-B^{ik}+\frac{1}{2}\cdot\frac{DV^{ik}}{Dt}+\frac{1}{2}\cdot\left(\frac{\partial\langle Z_{\mathrm{E}}^i\rangle}{\partial x^{\mathrm{L}}}\,V^{\mathrm{L}\,k}\right.\right.$$
$$\left.\left.+\frac{\partial\langle Z_{\mathrm{E}}^k\rangle}{\partial x^{\mathrm{L}}}\,V^{\mathrm{L}\,i}\right)+\frac{1}{2}\cdot A_{\mathrm{NL}}^{ik}\right\}(V^{-1})^{kj}, \tag{19b}$$

$$H^{ijk}=\frac{\lambda}{2}\cdot\left(\frac{\partial V^{in}}{\partial x^M}\,(V^{-1})^{nk}\delta_{Mj}+\frac{\partial V^{in}}{\partial x^M}\,(V^{-1})^{nj}\delta_{Mk}\right)$$
$$+\frac{1-2\lambda}{2}\cdot V^{iM}\,\frac{\partial V^{nl}}{\partial x^M}\,(V^{-1})^{lj}(V^{-1})^{nk}, \tag{19c}$$

are introduced, where A_{NL}^{ik} is any antisymmetric matrix, λ is any scalar quantity, and a constant density $\langle\rho\rangle$ is assumed for simplicity. The expression (19c) shows that nonlinear terms appear proportional to spatial gradients of the variances V^{ij} in dependence on an unknown parameter λ. The equation (18) is a generalization of the linear equations (1a) and (1b). The latter one is deduced by averaging the quadratic terms in the nonlinear equation (18), so that the third term on the right-hand side becomes $H^{ijk}V^{jk}$. The created linear term is equal to $\langle a^i\rangle$ in the linear model, which means $\langle a_{\mathrm{NL}}^i\rangle + H^{ijk}V^{jk}=\langle a^i\rangle$ is fulfilled independent of λ. Instead of considering the matrix G_{NL}^{ij} itself, let us consider $G_{\mathrm{NL}}^{ik}V^{kj}$ separated into the sum of its symmetric part $\frac{1}{2}\,[(G_{\mathrm{NL}}V)^{ij}+(G_{\mathrm{NL}}V)^{ji}]$ and antisymmetric part $\frac{1}{2}\,[(G_{\mathrm{NL}}V)^{ij}-(G_{\mathrm{NL}}V)^{ji}]$. These two summands follow from (19b):

$$G_{\mathrm{NL}}^{ik}V^{kj}+G_{\mathrm{NL}}^{jk}V^{ki}=\frac{DV^{ij}}{Dt}+\frac{\partial\langle Z_{\mathrm{E}}^i\rangle}{\partial x^K}\,V^{Kj}$$
$$+\frac{\partial\langle Z_{\mathrm{E}}^j\rangle}{\partial x^K}\,V^{Ki}-2B^{ij}, \tag{20a}$$

independent of λ as well as

$$G_{\mathrm{NL}}^{ik}V^{kj}-G_{\mathrm{NL}}^{jk}V^{ki}=A_{\mathrm{NL}}^{ij}. \tag{20b}$$

The symmetric component of $G_{\mathrm{NL}}^{ik}V^{kj}$ is given by the same expression as the symmetric component of $G^{ik}V^{kj}$ [which is determined by (8)] of the linear model, up to the gradients of third moments R^{ij} that vanish here. The antisymmetric component of $G_{\mathrm{NL}}^{ik}V^{kj}$ cannot be derived within this approach. This fact as well as the unknown λ reflect the uncertainty of rot $\mathbf{\Phi}$ for the considered Gaussian turbulence. This nonuniqueness was considered by Thomson,[4] Sawford,[5] and Borgas and Sawford[28] with respect to neutral flows. The appearance of λ shows that this problem has not only geometrical aspects. If the third term on the right-hand side of (18) is interpreted as a fluctuating drift (it contributes in the average to $\langle a^i_{\mathrm{NL}}\rangle$), we see that different intensive fluctuations of this drift may occur. However, the drift is not affected by the distribution parameter λ in the ensemble average, since $\langle a^i_{\mathrm{NL}}\rangle + H^{ijk}V^{jk} = \langle a^i\rangle$. For homogeneous and stationary turbulence, we find by means of (18) and (19a) and (19c) that $\langle dZ^i_{\mathrm{L}}(t)/dt \cdot Z^j_{\mathrm{L}}(t)\rangle = \frac{1}{2}\cdot A^{ij}_{\mathrm{NL}}$, where $\langle Z^i_{\mathrm{E}}\rangle = 0$ and $\langle Z^i_{\mathrm{L}}(t) \cdot Z^j_{\mathrm{L}}(t)\rangle = V^{ij}$ are applied. This relation reveals that correlations between states and state changes of different components are taken into account by the matrix A_{NL}.

As shown by Sawford, Borgas, and Guest,[28,29] different assumptions to rot $\mathbf{\Phi}$ produce significantly different results in the inertial subrange of neutral boundary layer flows. The derivation of conditions to estimate such open distribution parameters was studied by Borgas and Sawford considering two-particle dispersion.[28] They considered the reduction of open parameters under the constraint that one-particle statistics must follow from two-particle models. The parameters can be estimated by this reduction procedure up to one parameter, which remains open analogous to the appearance of λ here (the matrix A_{NL} is omitted by the assumed isotropic turbulence). Through the equations presented here, the effect of the choice of these open parameters on the modelling of non-neutral flows can be studied. The description of the interaction between the turbulent and the buoyant motion may be changed, e.g., by the choice of $A^{4k}_{\mathrm{NL}}(V^{-1})^{kj} \neq 0$ or $A^{4k}(V^{-1})^{kj}\neq 0$ appearing in (19b) and in (12), respectively. In this case, the change of the potential temperature is influenced by velocity fluctuations. The investigation of this effect can provide a better insight into the effect of the choice of these open parameters.

Consequently, taking reference to the Kolmogorov approximation (4) it is found that the consistency between the Lagrangian description and the infinite hierarchy of AHE is guaranteed, if the relations (16a) and (16b) are fulfilled. Adopting the approach of Du *et al.*[26] to construct the needed distribution density F, the non-uniqueness problem has to be solved, and in particular the total time derivative of F is required to solve (16b). For local Gaussian turbulence the latter problem is reduced according to (19a)–(19c) to the estimation of the gradients of mean values and variances. With DV^{ij}/Dt in the Rotta appproximation for the pressure redistribution (Sec. II), the calculation of the time scale τ is again needed (Sec. V).

IV. NONLINEAR NON-MARKOVIAN EQUATIONS

Up to now the suitability of different stochastic processes was considered to present solutions of the AHE, which means to have moments that obey the AHE. But these conditions tolerate a variety of different processes. Another approach to the description of motion and properties of particles consists in the derivation of equations from the microscopic dynamics. This is an important aim of statistical mechanics. It can be achieved, e.g., by the projection operator technique,[30–32] where the dynamics of observables is extracted from the coupled dynamics of all particles. A feature of this method is that the obtained equations are given as a superposition of systematic terms (which have in general a non-Markovian character) and of a term that shows properties of a stochastic force. The coefficients appearing in these equations are given as ensemble averages of microscopic quantities, which can be calculated using models for the Liouville operator. However, in most cases the calculation of these averages is very complicated, and fluctuation–dissipation theorems are more useful, which relate coefficients characterizing the intensity of stochastic forces with those characterizing the systematic motion. Whereas for equilibrium systems a well-established theory exists, there are many different attempts to extend this approach to the description of nonequilibrium processes. The estimation of coefficients that appear in these equations requires assumptions on the nonequilibrium probability density function. These approaches are limited to the consideration of isolated systems (or parts of isolated systems) reaching an equilibrium state, or assumptions on the kind of the nonequilibrium state are used. Instead, an approach is applied here,[33] where an identity replaces the Liouville equation of statistical mechanics and an initial distribution function plays the role of the nonequilibrium distribution function. Assumption on the initial state are not needed, which may be in a strong nonequilibrium. In this way, a nonlinear stochastic differential equation can be derived using formally the approach of nonequilibrium statistical mechanics. It is shown how correlations of stochastic forces lead to memory effects in the systematic terms, which is described by a fluctuation–dissipation theorem. Let us start from the abstract equation of motion ($i=1$–4, $I=1$–3)

$$\frac{d}{dt}\, x^I_{\mathrm{L}}(t) = Z^I_{\mathrm{L}}(t), \tag{21a}$$

$$\frac{d}{dt}\, Z^i_{\mathrm{L}}(t) = LZ^i_{\mathrm{L}}(t), \tag{21b}$$

corresponding with the Liouville equation in statistical mechanics ($-L$ being the Liouville operator), whereas Eq. (21b) appears here as an identity. Using the projection operator technique, the right-hand side of (21b) can be written such that this equation shows similar properties like a stochastic differential equation. Instead of this evolution equation for the process $\mathbf{Z}_{\mathrm{L}}(t)$, the dynamics of the generating function $\Psi(\mathbf{v},t) = \delta[\mathbf{v} - \mathbf{Z}_{\mathrm{L}}(t)]$ for all polynomials of $\mathbf{Z}_{\mathrm{L}}$ can be considered,

$$\frac{d}{dt}\, \Psi(\mathbf{v},t) = L\Psi(\mathbf{v},t), \tag{22}$$

from which (21b) is derived by multiplication with v^i and integration over $\mathbf{v}$. According to (22), the Taylor series of Ψ reads $\Psi(\mathbf{v},t) = \exp(Lt)\Psi_0(\mathbf{v})$, where $\Psi_0(\mathbf{v}) = \Psi(\mathbf{v}, t=0)$. The equation (21b) can be written with this series as $dZ^i_{\mathrm{L}}/dt = \int d\mathbf{v}v^i L\ \exp(Lt)\Psi_0(\mathbf{v})$. This expression is the starting point for the applied procedure. Let us consider an ensemble of particles with fixed initial positions $\mathbf{x}_{\mathrm{L}}(0) = \mathbf{x}_0$ for all particles and the space of products of functions $A_i[Z_{\mathrm{L}}(t_i)]$ with

$0 \leq t_i < \infty$, where any element $A(t_1, t_2, \ldots)$ of this space may be written as $A(t_1, t_2, \ldots) = A_1[\mathbf{Z}_{\mathrm{L}}(t_1)] \cdot A_2[\mathbf{Z}_{\mathrm{L}}(t_2)] \cdots$. The projection operator P is defined on this space by

$$PA(t_1, t_2, \ldots) = \int d\mathbf{v} g_0^{-1}(\mathbf{v}) \langle A(t_1, t_2, \ldots) \cdot \Psi_0(\mathbf{v}) \rangle \Psi_0(\mathbf{v}), \quad (23)$$

which projects any function $A(t_1, t_2, \ldots)$ onto the Ψ_0. The distribution function of fluctuating initial values $\mathbf{Z}_{\mathrm{L}}(0) = \mathbf{Z}_0$ (which is often used below) is abbreviated by $g_0(\mathbf{v}) = \langle \delta(\mathbf{v} - \mathbf{Z}_0) \rangle$. This function replaces the nonequilibrium distribution function of statistical mechanics. The definition (23) contains $\langle A(t_1, t_2, \ldots) \Psi_0(\mathbf{v}) \rangle$, which is an average over the considered ensemble, which means a conditional expectation value to be taken for $\mathbf{x}_{\mathrm{L}}(0) = \mathbf{x}_0$ (as all the other averages in this section).

The ensemble average $\langle A(t_1, t_2, \ldots) \Psi_0(\mathbf{v}) \rangle$ is determined by

$$\langle A(t_1, t_2, \ldots) \cdot \Psi_0(\mathbf{v}) \rangle = \int d\mathbf{v}' \int d\mathbf{v}'' \langle \delta(\mathbf{v} - \mathbf{Z}_0) \cdot \delta[\mathbf{v}' - \mathbf{Z}_{\mathrm{L}}(t_1)] \cdot \delta[\mathbf{v}'' - \mathbf{Z}_{\mathrm{L}}(t_2)] \cdots \rangle \cdot A_1[\mathbf{v}'] \cdot A_2[\mathbf{v}''] \cdots . \quad (24)$$

More frequent than P, the operator $Q = 1 - P$ is used, which is characterized by $QQ = Q$, $Q\Phi_0 = 0$, and $\langle QA \cdot B \rangle = \langle QB \cdot A \rangle$ for any elements A and B of the considered function space. Applying the usual identity of the projection operator technique,

$$\exp(Lt) = \exp(QLt) + \int_0^t dt' \exp(Lt') PL \exp[QL(t - t')], \quad (25)$$

which can be proved by differentiation, $dZ_{\mathrm{L}}^i/dt = \int d\mathbf{v} \mathrm{v}^i L \exp(Lt) \Psi_0(\mathbf{v})$ can be written as

$$\frac{d}{dt} Z_{\mathrm{L}}^i(t) = -\int d\mathbf{v}\, g_0^{-1}(\mathbf{v}) M^i(\mathbf{v},0) \Psi(\mathbf{v},t) - \int_0^t dt' \int d\mathbf{v}\, g_0^{-1}(\mathbf{v}) \frac{dM^i}{dt}(\mathbf{v}, t - t') \times \Psi(\mathbf{v}, t') + f^i(t), \quad (26)$$

where $M^i(\mathbf{v},t) = -\int d\mathbf{w}\, w^i \langle L \exp(QLt) \Psi_0(\mathbf{w}) \cdot \Psi_0(\mathbf{v}) \rangle$ and $f^i(t) = \int d\mathbf{w}\, w^i QL \exp(QLt) \Psi_0(\mathbf{w})$ are introduced. The integration of $\langle f^i(t) \cdot \Psi_0(\mathbf{v}) \rangle = \langle Qf^i(t) \cdot \Psi_0(\mathbf{v}) \rangle = \langle f^i(t) \cdot Q\Psi_0(\mathbf{v}) \rangle = 0$ over $\mathbf{v}$ leads to $\langle f^i(t) \rangle = 0$, so that this quantity can be interpreted as a stochastic force and the equation (26) as a nonlinear stochastic differential equation. The expressions for the coefficients appearing in (26) are given in contrast to (14b). Writing the time dependence of the state vector as $\mathbf{Z}_{\mathrm{L}}(t) = \mathbf{Z}_{\mathrm{L}}[\mathbf{x}_{\mathrm{L}}(t), t]$ to enable the comparisons considered below, the coefficient $M^i(\mathbf{v},0)$ appearing in the first term on the right-hand side of (26) is determined by

$$\frac{\partial}{\partial \mathrm{v}^i} M^i(\mathbf{v},0) = -\left\langle \frac{\partial \Psi_0(\mathbf{v})}{\partial \mathrm{v}^i} \cdot LZ_0^i \right\rangle = \langle L\Psi_0(\mathbf{v}) \rangle = \left[\left(\frac{\partial}{\partial t} + \frac{\partial}{\partial x^K} \mathrm{v}^k \right) \langle \Psi(\mathbf{v},t) \rangle \right] \quad (t = 0), \quad (27)$$

and $M^i(\mathbf{v},0) \to 0$ for $|\mathbf{v}| \to \infty$. The coefficient of the second term of (26) can be written as $dM^i(\mathbf{v},t)/dt = -\langle Lf^i(t) \cdot \Psi_0(\mathbf{v}) \rangle$. L becomes an anti-hermitian operator, if for arbitrary elements A and B of the considered function space $\langle LA \cdot B \rangle + \langle LB \cdot A \rangle = d\langle A \cdot B \rangle / dt = 0$, which means for homogeneous and stationary turbulence. In this case we find $\langle Lf^i(t) \cdot \Psi_0 \rangle = \langle LQf^i(t) \cdot \Psi_0 \rangle = -\langle f^i(t) \cdot QL\Psi_0 \rangle = \partial \langle f^i(t) \cdot f^k(0) \cdot \Psi_0(\mathbf{v}) \rangle / \partial \mathrm{v}^k$, and $dM^i(\mathbf{v},t)/dt$ can be converted into

$$\frac{d}{dt} M^i(\mathbf{v},t) = -\frac{\partial}{\partial \mathrm{v}^k} \langle f^i(t) \cdot f^k(0) \cdot \Psi_0(\mathbf{v}) \rangle. \quad (28)$$

This result represents a fluctuation–dissipation theorem, because the correlation of stochastic forces is related with the memory function $M^i(\mathbf{v},t)$ that characterizes the dissipation process. Let us consider the consequences of delta and exponential correlated forces. In the first case, the force is supposed to be proportional to a vectorial Wiener process, $f^i(t) = b^{ik}(\mathbf{Z}_{\mathrm{L}}, \mathbf{x}_{\mathrm{L}}, t) dW^k(t)/dt$, and the Markov limit of (26) for $t > 0$ is given by $[B_0 = B(\mathbf{Z}_0, \mathbf{x}_0, 0)]$

$$\frac{d}{dt} Z_{\mathrm{L}}^i(t) = -\int d\mathbf{v}\, \Psi(\mathbf{v},t) g_0^{-1}(\mathbf{v}) \left(M^i(\mathbf{v},0) - \frac{\partial}{\partial \mathrm{v}^k} \langle B_0^{ik} \Psi_0 \rangle \right) + b^{ik} \frac{dW^k}{dt}(t). \quad (29)$$

A comparison of this equation with the nonlinear Markov equation (14b) with the relations (16a) and (16b) shows, that for the considered homogeneous and stationary turbulence $F = g_0$, $\Phi^i = -M^i$ and $B^{ik} F = \langle B_0^{ik} \Psi_0 \rangle$. The equation (27) for M^i represents the relation (16b) for this case. As stated by Lindenberg and West (Chapter 1.2.2.),[32] a dependence of fluctuations $b^{ik}(\mathbf{Z}_{\mathrm{L}}, \mathbf{x}_{\mathrm{L}}, t) dW^k(t)/dt$ from the present and perhaps even the past state $\mathbf{Z}_{\mathrm{L}}$ is expected in general, but it is a question to be answered if such a state dependence can be included in a stochastic dynamic description. By the above results (29), only an averaged contribution of a state dependence of fluctuations can contribute to the considered systematic motion, since only $\langle B_0^{ik} \Psi_0 \rangle$ acts here. The first term on the right-hand side of (29) represents a mean drift. By averaging the equation (29) at $t = 0$, we find that $[d\langle Z_{\mathrm{L}}^i(t) \rangle / dt](t = 0) = -\int d\mathbf{v}\, M^i(\mathbf{v},0)$, since the other terms do not contribute. For the considered homogeneous and stationary turbulence we neglect $M^i(\mathbf{v},0)$, which is justified by (27). The quantity $\mathbf{Z}_{\mathrm{L}}$ is then produced by the white noise term and dissipated by the second term on the right-hand side of (29). The latter term becomes a linear function in $\mathbf{Z}_{\mathrm{L}}(t)$, if B_0^{ij} is supposed to be uncorrelated with Ψ_0 and the distribution function density of initial states is taken as Gaussian, $g_0(\mathbf{v}) \sim \exp[-\frac{1}{2}(d^{-1})^{kl} \mathrm{v}^k \mathrm{v}^l]$ with the dispersion matrix d. In this case we have

$$\int d\mathbf{v}\, g_0^{-1}(\mathbf{v})\Psi(\mathbf{v},t)\cdot \frac{\partial}{\partial \mathrm{v}^k}\langle B_0^{ik}\Psi_0\rangle$$

$$=-\langle B_0^{ik}\rangle(d^{-1})^{kl}\cdot Z_{\mathrm{L}}^1(t). \qquad (30)$$

Let us consider now the correlation function following from this nonlinear Markovian equation. Multiplying (29) for $M^i(\mathbf{v},0)=0$ with Z_0^i and averaging leads for $t>0$ to

$$\frac{d}{dt}\langle Z_{\mathrm{L}}^i(t)\cdot Z_0^j\rangle=\int d\mathbf{v}\langle\Psi(\mathbf{v},t)\cdot Z_0^i\rangle\cdot g_0^{-1}(\mathbf{v})\frac{\partial}{\partial \mathrm{v}^k}\langle B_0^{ik}\Psi_0\rangle, \qquad (31)$$

since the stochastic force $f^i(t)=b^{ik}\,dW^k(t)/dt$ is uncorrelated with Z_0^i, which follows from the above considered properties of f^i by $\int d\mathbf{v}\,\mathrm{v}^j\langle f^i(t)\cdot\Psi_0(\mathbf{v})\rangle=\langle f^i(t)\cdot Z_0^j\rangle=0$. Assuming B_0^{ij} to be uncorrelated with $\Psi_0(\mathbf{v})$, which means $\langle B_0^{ij}\cdot\Psi_0(\mathbf{v})\rangle=\langle B_0^{ij}\rangle\cdot\langle\Psi_0(\mathbf{v})\rangle$, and adopting a Gaussian $g_0(\mathbf{v})\sim\exp[-\frac{1}{2}(d^{-1})^{kl}\mathrm{v}^k\mathrm{v}^l]$ as before, this relation (31) becomes

$$\frac{d}{dt}\langle Z_{\mathrm{L}}^i(t)\cdot Z_0^j\rangle=-\langle B_0^{ik}\rangle\cdot(d^{-1})^{kl}\cdot\langle Z_{\mathrm{L}}^1(t)\cdot Z_0^j\rangle, \qquad (32)$$

providing with $d^{ij}=\langle Z_0^i\cdot Z_0^j\rangle$ the usual exponential correlation function for homogeneous and stationary turbulence (for $t\geqslant 0$),

$$\langle Z_{\mathrm{L}}^i(t)\cdot Z_0^j\rangle=\langle Z_0^i\cdot Z_0^k\rangle\cdot[\exp(-\langle B_0\rangle\cdot(d^{-1})\cdot t)]^{kj}. \qquad (33)$$

The limit $t\to 0$ of the derivative of $\langle Z_{\mathrm{L}}^i(t)\cdot Z_0^j\rangle$ is then determined by

$$\lim_{t\to 0}\frac{d}{dt}\langle Z_{\mathrm{L}}^i(t)\cdot Z_0^j\rangle=-\langle B_0\rangle^{ij}. \qquad (34)$$

It is important to note that this result is independent of the applied assumptions of uncorrelated B_0^{ij} and $\Psi_0(\mathbf{v})$ and a Gaussian $g_0(\mathbf{v})$, which can be seen from (31) for the correlation function of the nonlinear process at $t=0$ by applying partial integration. This result (34) is a shortcoming of the Markov theory, because the derivative of $\langle Z_{\mathrm{L}}^i(t)\cdot Z_0^j\rangle$ must vanish in the limit $t\to 0$.[34] This behavior is found if processes in the order of the Kolmogorov microscale are taken into account, which is considered now.

In order to do this let us consider as a second example an exponential function for the correlation of stochastic forces and as above a Gaussian initial distribution function $g_0\sim\exp[-\mathrm{v}^2/(2d)]$, where one component of the velocity is considered for simplicity with d as the dispersion parameter. The stochastic force f is driven by white noise,

$$\frac{df}{dt}=-\lambda f+\sqrt{2\lambda d_f}\cdot\frac{dW}{dt}, \qquad (35)$$

where d_f denotes the dispersion $d_f=\langle f^2\rangle$. The force vanishes in the ensemble average and its correlation is given by assumed stationarity by $\langle f(t)f(t+s)\rangle=d_f\exp\{-\lambda|s|\}$.[35] The correlation time of this colored noise f is λ^{-1}. In the limit $\lambda^{-1}\to 0$, the stochastic force becomes again delta correlated as assumed above, since $\langle f(t)f(t+s)\rangle\to 2d_f/\lambda\cdot\delta(s)$. The equation for the stochastic particle velocity reads with these assumptions for g_0 and $\langle f(t)f(t+s)\rangle$ as

$$\frac{d}{dt}Z_{\mathrm{L}}(t)=-\frac{d_f}{d}\int_0^t dt'\,\exp[-\lambda(t-t')]Z_{\mathrm{L}}(t')+f(t), \qquad (36)$$

where (28) is used and $M^i(\mathbf{v},0,\mathbf{x}_0)$ is neglected in the equilibrium, as discussed above. An equation for the acceleration is obtained, if (36) is differentiated by time and df/dt is substituted by (35),

$$\frac{d^2}{dt^2}Z_{\mathrm{L}}(t)=-\lambda\cdot\frac{dZ_{\mathrm{L}}}{dt}(t)-\frac{d_f}{d}\cdot Z_{\mathrm{L}}(t)+\sqrt{2\lambda d_f}\cdot\frac{dW}{dt}(t), \qquad (37)$$

which is again driven by white noise. This equation corresponds with Sawfords equation for the fluid particle acceleration,[5,34,36] if $\lambda=\beta_1+\beta_2$ and $d_f=\beta_1\beta_2 d$, with $\beta_1=1/T_{\mathrm{L}}^{(\infty)}$ and $\beta_2=(\mathrm{Re}^*)^{1/2}/T_{\mathrm{L}}^{(\infty)}$. Here Re* is a number that is proportional to the Reynolds number Re, $T_{\mathrm{L}}^{(\infty)}$ is the Lagrangian integral time scale in the limit $\mathrm{Re}\to\infty$, and β_2^{-1} is proportional to the Kolmogorov microscale.[34]

This model explains well the Reynolds number effects in models of the turbulent dispersion. For $\mathrm{Re}\to\infty$ we find $\lambda^{-1}\to 0$, so that the forces f become uncorrelated. But instead of (36), the velocity is modelled by Sawford by

$$\frac{d}{dt}Z_{\mathrm{L}}(t)=-\frac{1}{T_{\mathrm{L}}^{(\infty)}}\cdot Z_{\mathrm{L}}(t)+f(t), \qquad (38)$$

so that (37) has to be solved with another initial condition for the acceleration. Accordingly, we find the solution $Z_{\mathrm{L}}(t)$ in these two approaches by

$$Z_{\mathrm{L}}(t)=Z_{\mathrm{L}}(0)\cdot\begin{Bmatrix}\dfrac{\beta_2\exp(-\beta_1 t)-\beta_1\exp(-\beta_2 t)}{\beta_2-\beta_1} & \text{with (36)}\\ \exp(-\beta_1 t), & \text{with (38)}\end{Bmatrix}$$
$$-\frac{f(0)}{\beta_2-\beta_1}\cdot[\exp(-\beta_1 t)-\exp(-\beta_2 t)]$$
$$-\frac{\sqrt{2(\beta_1+\beta_2)\beta_1\beta_2 d}}{\beta_2-\beta_1}\cdot\int_0^t ds\,\frac{dW}{ds}(s)$$
$$\cdot\{\exp[-\beta_1(t-s)]-\exp[-\beta_2(t-s)]\}. \qquad (39)$$

In both models, for asymptotically large times the velocity autocorrelation function is obtained to be $\langle Z_{\mathrm{L}}(t)Z_{\mathrm{L}}(t+s)\rangle/d=[\beta_2\exp(-\beta_1|s|)-\beta_1\exp(-\beta_2|s|)]/(\beta_2-\beta_1)$, which arises from the third term on the right-hand side of (39). However, with $d=\langle[Z_{\mathrm{L}}(0)]^2\rangle$ the model (36) provides the corresponding expression for the decay of the initial correlation, which means $\langle Z_{\mathrm{L}}(0)Z_{\mathrm{L}}(t)\rangle/d=[\beta_2\exp(-\beta_1 t)-\beta_1\exp(-\beta_2 t)]/(\beta_2-\beta_1)$ in contrast to (38), which leads to $\langle Z_{\mathrm{L}}(0)Z_{\mathrm{L}}(t)\rangle/d=\exp(-\beta_1 t)$. This consistent description of the decay of correlations becomes important, if other processes have to be resolved over times of order β_2^{-1} (i.e., of the order of the Kolmogorov microscale). This may occur in cases where chemical transformations have to be considered near sources. By adopting the equation (36), we find for the correlation of initial values for small times

$\langle Z_L(0)Z_L(t)\rangle/d = 1-\beta_1\beta_2/2\cdot t^2$, such that the derivative of this function vanishes in the limit $t\to 0$ in contrast to (34).

V. THE PRODUCTION–DISSIPATION RELATION FOR THE TKE

As stated above, the Lagrangian equations can be chosen, so that the AHE are guaranteed up to second order for the moments of the stochastic processes, or for the whole infinite hierarchy of equations, respectively. But in these equations an unknown time scale τ may appear arising, e.g., from the applied approximations, as discussed in Sec. II. The estimation of this time scale τ for inhomogeneous turbulence is an important problem and needed, for instance, for the solution of the equation (1a) and (1b) with the relations (4), (10), and (13) for the coefficients and for the solution of the second-order equations (11).[37] This time scale determines the ratio of the total production $P^{ij}_{\mathrm{tot}} = P^{ij} - \beta g\ \delta_{i3}V^{3j} - \beta g\ \delta_{j3}V^{3i}$ in (11) to the dissipation given by the terms proportional to τ^{-1} on the right-hand side. For $\tau\to 0$, this input P^{ij}_{tot} is immediately dissipated and the terms on the left-hand side of (11) vanish, which means the fluxes are in balance with the gradients of the mean fields. For $\tau\to\infty$, the dissipation terms can be neglected and we have $DV^{ij}/Dt + R^{ij} = -P^{ij}_{\mathrm{tot}}$, which means the variances V^{ij} change as long as the spatial transport is in balance with the input P^{ij}_{tot}. Consequently, let us consider the relation between the production–dissipation ratio for TKE and τ, in order to calculate this time scale τ by assumptions on that scalar production–dissipation ratio. This ratio expresses under stationary conditions the amount of spatial transport of TKE, which plays, e.g., an essential role under convective conditions.[38,39] Hence, the deviation of the production–dissipation ratio for TKE from unity (caused by these transports) has to be assessed, which is simpler than finding τ directly for inhomogeneous conditions in correspondence with the gradients of the mean wind and potential temperature fields.

This production–dissipation relation will be derived now for a vertical stratified flow for simplicity. Let us suppose a mean horizontal wind U into the x^1 direction and a mean potential temperature Θ, which depend only on the vertical coordinate x^3. The mean vertical wind W is assumed as constant, so that $\langle \mathbf{Z}_E\rangle = [U(x^3), 0, W, \Theta(x^3)]$. The second-order equations (11) provide for the TKE budget,

$$\frac{Dq^2}{Dt} + R^{KK} = 2(P - \langle\epsilon\rangle), \tag{40}$$

where $P = -P^{KK}/2 + \beta g V^{34}$ is the production of TKE and the mean dissipation rate of kinetic energy is given by $\langle\epsilon\rangle = q^2/(2\tau)$, where the time scale τ is assumed as time independent. With this expression for P, the production–dissipation ratio $p = P/\langle\epsilon\rangle$ is then

$$p = -\frac{2}{q^2}\cdot(T\hat{V}^{13} - \hat{V}^{34}), \tag{41}$$

where the normalized time scale $T = \tau\ \partial U/\partial x^3$ is introduced. The variances $\hat{V}^{34}$ and $\hat{V}^{13}$ are elements of the matrix $\hat{V}$, which is introduced by

$$\hat{V} = \begin{pmatrix} \langle u^1u^1\rangle & \langle u^1u^2\rangle & \langle u^1u^3\rangle & \beta g\tau\langle u^1\theta\rangle \\ \langle u^2u^1\rangle & \langle u^2u^2\rangle & \langle u^2u^3\rangle & \beta g\tau\langle u^2\theta\rangle \\ \langle u^3u^1\rangle & \langle u^3u^2\rangle & \langle u^3u^3\rangle & \beta g\tau\langle u^3\theta\rangle \\ \beta g\tau\langle\theta u^1\rangle & \beta g\tau\langle\theta u^2\rangle & \beta g\tau\langle\theta u^3\rangle & (\beta g\tau)^2\langle\theta^2\rangle \end{pmatrix}, \tag{42}$$

where all elements have the dimension of the TKE. The relation (41) requires the calculation of $\hat{V}^{34}$ and $\hat{V}^{13}$, which can be obtained by the second-order equation system (11). In order to do this the consideration of modified gradients $\hat{R}$ of third moments is advantageous, which have the same dimension as $\hat{V}$ and are given by

$$\hat{R} = \tau\cdot\begin{pmatrix} R^{11} & R^{12} & R^{13} & \beta g\tau R^{14} \\ R^{21} & R^{22} & R^{23} & \beta g\tau R^{24} \\ R^{31} & R^{32} & R^{33} & \beta g\tau R^{34} \\ \beta g\tau R^{41} & \beta g\tau R^{42} & \beta g\tau R^{43} & (\beta g\tau)^2 R^{44} \end{pmatrix}. \tag{43}$$

If the time dependence is observed in $t' = t/\tau$, which is normalized to the time-independent τ, the operator $D/Dt'(\cdot) = [\partial/\partial t' + \partial/\partial x^3\ W\tau]\ (\cdot)$ is applied and the gradient Richardson number $\mathrm{Ri} = \beta g\ \partial\Theta/\partial x^3/(\partial U/\partial x^3)^2$ is used, we obtain from the second-order equations (11) the system of coupled equations,

$$\frac{D}{Dt'}\begin{pmatrix} \hat{V}^{13} \\ \hat{V}^{14} \\ \hat{V}^{34} \\ \hat{V}^{33} \\ \hat{V}^{44} \\ q^2 \end{pmatrix} + \begin{pmatrix} \hat{R}^{13} \\ \hat{R}^{14} \\ \hat{R}^{34} \\ \hat{R}^{33} \\ \hat{R}^{44} \\ \hat{R}^{KK} \end{pmatrix} = \begin{pmatrix} -k_1/2 & 1 & 0 & -T & 0 & 0 \\ -\mathrm{Ri}\ T^2 & -k_3/2 & -T & 0 & 0 & 0 \\ 0 & 0 & -k_3/2 & -\mathrm{Ri}\ T^2 & 1 & 0 \\ 0 & 0 & 2 & -k_1/2 & 0 & (k_1-2)/6 \\ 0 & 0 & -2\,\mathrm{Ri}\ T^2 & 0 & -k_4 & 0 \\ -2T & 0 & 2 & 0 & 0 & -1 \end{pmatrix}\begin{pmatrix} \hat{V}_{13} \\ \hat{V}^{14} \\ \hat{V}^{34} \\ \hat{V}^{33} \\ \hat{V}^{44} \\ q^2 \end{pmatrix}, \tag{44a}$$

which includes the variances $\hat{V}^{34}$ and $\hat{V}^{13}$. The other components of the matrix $\hat{V}$ obey the equations

$$\frac{D\hat{V}^{11}}{Dt'}+\hat{R}^{11}=-\frac{k_1}{2}\cdot\hat{V}^{11}-2T\cdot\hat{V}^{13}+\frac{k_1-2}{6}\cdot q^2, \quad (44b)$$

where q^2 and $\hat{V}^{13}$ are given as solutions of (44a), $\hat{V}^{22}$ is determined by $\hat{V}^{22}=q^2-\hat{V}^{33}-\hat{V}^{11}$, and the remaining components of $\hat{V}$ satisfy

$$\frac{D}{Dt'}\begin{pmatrix}\hat{V}^{12}\\ \hat{V}^{23}\\ \hat{V}^{24}\end{pmatrix}+\begin{pmatrix}\hat{R}^{12}\\ \hat{R}^{23}\\ \hat{R}^{24}\end{pmatrix}=\begin{pmatrix}-k_1/2 & -T & 0\\ 0 & -k_1/2 & 1\\ 0 & -\mathrm{Ri}\,T^2 & -k_3/2\end{pmatrix}\cdot\begin{pmatrix}\hat{V}^{12}\\ \hat{V}^{23}\\ \hat{V}^{24}\end{pmatrix}. \quad (44c)$$

For the considered vertical stratified flow, the horizontal components with $i=1,2$ and $j=1,2,3,4$ on the left-hand sides of the second-order equation system (44a)–(44c) will be neglected. The term $Dq^2/Dt'+\hat{R}^{KK}$ in (44a) can be replaced by $q^2(p-1)$ according to (40), and $D\hat{V}^{33}/Dt'+\hat{R}^{33}$ can be replaced in this equation system also by this expression, i.e. $D\hat{V}^{33}/Dt'+\hat{R}^{33}=q^2(p-1)$, because $\hat{V}^{11}$ and $\hat{V}^{22}$ are assumed to be not contributing to the budget of q^2 in this approximation. The equations system (44a) can be solved simply, if the time derivatives and gradients of triple correlations are considered as inhomogeneities. In this way $\hat{V}^{34}$ and $\hat{V}^{13}$ can be calculated, where a quantity p_θ appears, with $(p_\theta-1)\cdot q^2=(D\hat{V}^{44}/Dt'+\hat{R}^{44})/k_4+D\hat{V}^{34}/Dt'+\hat{R}^{34}$. The meaning of p_θ becomes clear, if the budgets for the variance V^{44} of heat fluctuations and the vertical heat flux V^{34} are considered in analogy to the budget equation (40). The second-order equations (11) give for these quantities

$$\frac{DV^{44}}{Dt}+R^{44}=2(P_{\theta\theta}-\langle\epsilon_{\theta\theta}\rangle), \quad (45a)$$

$$\frac{DV^{34}}{Dt}+R^{34}=2(P_{w\theta}-\langle\epsilon_{w\theta}\rangle), \quad (45b)$$

where $P_{\theta\theta}=-V^{34}\,\partial\Theta/\partial x^3$ and $P_{w\theta}=(-V^{33}\,\partial\Theta/\partial x^3-V^{34}\,\partial U/\partial x^3)/2$ are the production terms and $\langle\epsilon_{\theta\theta}\rangle=k_4V^{44}/(2\tau)$ as well as $\langle\epsilon_{w\theta}\rangle=k_3V^{34}/(4\tau)$ are the dissipation terms. The parameter p_θ is determined by these expressions by the relation

$$p_\theta-1=\frac{k_4}{4}\cdot\frac{(\beta gV^{44})^2}{\langle\epsilon\rangle\cdot\langle\epsilon_{\theta\theta}\rangle}\cdot\left(\frac{P_{\theta\theta}}{\langle\epsilon_{\theta\theta}\rangle}-1\right)+\frac{k_3}{4}\cdot\frac{\beta gV^{34}}{\langle\epsilon\rangle}\cdot\left(\frac{P_{w\theta}}{\langle\epsilon_{w\theta}\rangle}-1\right). \quad (46)$$

Hence, deviations of p_θ from unity are caused by deviations of $P_{\theta\theta}/\langle\epsilon_{\theta\theta}\rangle$ and $P_{w\theta}/\langle\epsilon_{w\theta}\rangle$ from unity, so that p_θ describes the production–dissipation ratio of heat in analogy to p describing this ratio for the TKE. Inserting the variances $\hat{V}^{34}$ and $\hat{V}^{13}$ calculated by (44a) in dependence on p, p_θ, T, and Ri into (41) leads then to the relation

$$p+\frac{4}{k_3}\cdot(p_\theta-1)=\frac{T^2}{T_0^2}\cdot\left(1-8\cdot\frac{T_0^2}{k_1^2}\cdot(p-1)\right)\cdot\frac{\mathrm{Pr}_{\mathrm{st}}-(p_\theta-1)\mathrm{Pr}_{ut}}{\mathrm{Pr}_0}-\frac{\mathrm{Ri}\;T^2}{\mathrm{Ri}_0\;T_0^2}\cdot\left(1+4\cdot\frac{T_0^2}{k_1^2}\cdot\frac{\mathrm{Ri}_c\;\mathrm{Pr}_0+\mathrm{Ri}_0}{\mathrm{Pr}_0^2}\cdot(p-1)\right), \quad (47)$$

for the production–dissipation ratio p. $\mathrm{Pr}_{\mathrm{st}}=k_3/k_1\cdot(k_1k_3+\mathrm{Ri}_c/\mathrm{Ri}_0\cdot 4\,\mathrm{Ri}\,T^2)/(k_1k_3+4\,\mathrm{Ri}\,T^2)$ is the turbulent Prandtl number $\mathrm{Pr}_t=\mathrm{Ri}\,T\hat{V}^{13}/\hat{V}^{34}$ for a production–dissipation ratio $p_\theta=1$ for heat, $\mathrm{Pr}_{ut}=2(k_1+2k_3)/[(k_1-2)/6-(p-1)]/(k_1k_3+4\,\mathrm{Ri}\,T^2)$ is a contribution related with an unbalanced ratio p_θ of heat, and $\mathrm{Pr}_0=k_3/k_1$ follows from $\mathrm{Pr}_{\mathrm{st}}$ for a neutral stratification, which means $\mathrm{Pr}_0=\mathrm{Pr}_{\mathrm{st}}(\mathrm{Ri}=0)$. Additionally, in the derived relation (47) the parameters $T_0^2=3k_1^2/[4(k_1-2)]$, $\mathrm{Ri}_0=3k_1k_3k_4/(4T_0^2[k_4(k_1+4)+3k_1])$, and $\mathrm{Ri}_c=(k_3-k_4)/\mathrm{k}_4\cdot\mathrm{Ri}_0/\mathrm{Pr}_0$ appear. This relation (47) quantifies the expectation that the production P of TKE (normalized to the dissipation) is determined by the (quadratic) wind shear $T^2=(\tau\;\partial U/\partial x^3)^2$ and the (unstable) temperature stratification $-\mathrm{Ri}\;T^2=-\tau^2\beta g\;\partial\Theta/\partial x^3$, which appear as factors on the right-hand side of (47). For balanced production–dissipation ratios $p=p_\theta=1$, this relation is given by the simple expression

$$p=1=\frac{T^2}{T_0^2}\cdot\left(\frac{\mathrm{Pr}_{\mathrm{st}}}{\mathrm{Pr}_0}-\frac{\mathrm{Ri}}{\mathrm{Ri}_0}\right). \quad (48)$$

This relation shows that T_0^2 is the value of T^2 for a neutral stratification Ri=0 for $p=p_\theta=1$. The relation (48) is characterized by the parameters Pr_0, Ri_0 as well as Ri_c, which limits the applicability of (48) as an equation for T by the condition $\mathrm{Ri}<\mathrm{Ri}_c$. These numbers can be interpreted as flow numbers.[37] As stated by Derbyshire,[40] in a free stratified shear layer of uniform shear and stratification at high Reynolds and Peclet numbers, turbulence is expected to grow or decay on a time scale of order $(\partial U/\partial x^3)^{-1}$, depending on the gradient Richardson number Ri. The crossover point at which turbulence neither grows nor decays defines a turbulent critical Richardson number. As shown below, the normalized time scale $T=\tau\;\partial U/\partial x^3$ (and for a finite wind shear τ also) becomes infinite for $\mathrm{Ri}\to\mathrm{Ri}_c$. Hence, for $\mathrm{Ri}\to\mathrm{Ri}_c$ the decay of turbulence by dissipation becomes weaker because of $\tau\to\infty$ and it diminishes at $\mathrm{Ri}=\mathrm{Ri}_c$, so that Ri_c defines the critical gradient Richardson number. Here $\mathrm{Pr}_0=\mathrm{Pr}_{\mathrm{st}}(\mathrm{Ri}=0)$ was found as turbulent Prandtl number $\mathrm{Pr}_{\mathrm{st}}$ for a neutral stratification and Ri_0 is a characteristic gradient Richardson number, as can be seen from (48). The first term on the right-hand side of (48), $\mathrm{Pr}_{\mathrm{st}}T^2/(\mathrm{Pr}_0\;T_0^2)$, is positive for a positive $\mathrm{Pr}_{\mathrm{st}}$, such that the condition $\tau^2\cdot(-\beta g\;\partial\Theta/\partial x^3)\cdot T_0^{-2}\leq\mathrm{Ri}_0$ arises from (48). This relation represents under unstable stratification a constraint for the dissipation that has to be large enough (the time scale τ has to be small enough) such that convective processes (spatial transports of TKE) are excluded and the relation (48) can be fulfilled. Consequently, Ri_0 characterizes the onset of convective processes under unstable stratification, in correspondence to Ri_c, which

TABLE I. The second-order closure parameters k_1, k_2, k_3, and k_4 estimated by different authors and the flow numbers Pr_0, Ri_c, and Ri_0 as well as C_0 and C_1, calculated by their relations with the closure parameters.

	k_1	k_2	k_3	k_4	Pr_0	Ri_c	Ri_0	C_0	C_1
Wichmann and Schaller[21]	5.0	0.0	3.4	1.48	0.68	0.20	0.11	1.00	−1.16
Mellor and Yamada[22]	6.0	0.08	7.5	1.66	1.25	0.68	0.24	1.33	5.68
Zeman and Lumley[41]	3.25	0.0	7.0	⋯	2.15	⋯	⋯	0.42	⋯
André *et al.*[42]	9.0	0.0	9.7	2.5	1.08	0.85	0.32	2.33	5.40
Wyngard *et al.*[43]	6.7	0.0	4.4	1.4	0.66	0.40	0.12	1.57	−0.70
Yamada[44]	5.0	0.05	11.8	2.0	2.36	0.89	0.43	1.00	14.60

characterizes the onset of turbulence under stable stratification.[37] These flow numbers Ri_c, Pr_0, and Ri_0 can be estimated by means of their relations with the second-order closure parameters k_1, k_3, and k_4. It is a common feature of the estimations of k_1, k_3, and k_4 that the fit of these parameters is related with the consideration of properties of neutral stratified flows,[22] or the closure parameters are fitted to the characteristics of different flows.[21] These estimations provide very different data for k_1, k_3, and k_4, as given in Table I. Additionally, the values of the flow numbers Pr_0, Ri_c, and Ri_0, and parameters $C_0=(k_1-2)/3$ and $C_1=2k_3-2k_4-k_1$ (Sec. II) are shown in dependence on k_1, k_3, and k_4. The obtained data for the flow numbers Pr_0, Ri_c, and Ri_0, as well as the advantage of considering these numbers, are discussed in relation to the solution of the second-order equations (11).[37] The values for C_0 in Table I are somewhat smaller than those estimated by Du *et al.*[45] for this quantity. By comparisons with water channel dispersion and wind tunnel measurements they derived $C_0=3.0\pm0.5$ and discussed the wide range of estimates of C_0 to be found in the literature (e.g., the relation to the much higher values derived for this quantity by Sawford[5,34] and Pope[6]). With the exception of the value obtained from the data of Zeman and Lumley, C_0 is found here in a range $1\leqslant C_0\leqslant 2.33$, which agrees qualitatively with the findings of Du *et al.* Measurements of C_1 seem to be unavailable. It is remarkable that negative values of C_1 are obtained with the data of Wichmann and Schaller[21] and Wyngaard *et al.*[43] This means, e.g., that in these cases more complicated coefficients G^{ij} of the linear Lagrangian model (1b) have to be chosen (in dependence on the variances), which cannot be described by the simple choice (13).

The equation (47) can be considered as a quadratic equation for τ^2 for given production–dissipation rates p and p_θ and gradients $\partial U/\partial x^3$ and $\partial\Theta/\partial x^3$. The normalized time scale $T=\tau\,\partial U/\partial x^3$ can be estimated by the equation

$$T^2=-\frac{1}{2A}\cdot\left(B+\sqrt{B^2-4AC}\right), \tag{49}$$

where the quantities A, B, and C are functions of the gradient Richardson number Ri and the production–dissipation ratios for TKE and heat, p and p_θ, respectively. They are found to be

$$A=4\,\mathrm{Ri}\cdot\Bigg(\frac{\mathrm{Ri}-\mathrm{Ri}_c}{\mathrm{Ri}_0}+(p-1)\cdot\frac{4T_0^2}{k_1^2\,\mathrm{Pr}_0^2\,\mathrm{Ri}_0}\cdot[\mathrm{Ri}(\mathrm{Ri}_c\,\mathrm{Pr}_0+\mathrm{Ri}_0)+2\,\mathrm{Ri}_c\,\mathrm{Pr}_0^2]\Bigg), \tag{50a}$$

$$B=4\,\mathrm{Ri}T_0^2\cdot p+k_1^2\,\mathrm{Pr}_0\cdot\frac{\mathrm{Ri}-\mathrm{Ri}_0}{\mathrm{Ri}_0}+(p-1)\cdot\frac{4T_0^2}{\mathrm{Pr}_0\,\mathrm{Ri}_0}\cdot[\mathrm{Ri}(\mathrm{Ri}_c\,\mathrm{Pr}_0+\mathrm{Ri}_0)+2\,\mathrm{Ri}_0\,\mathrm{Pr}_0^2]+(p_\theta-1)\cdot\frac{16T_0^2}{k_1\,\mathrm{Pr}_0}\cdot(\mathrm{Ri}+1+2\,\mathrm{Pr}_0), \tag{50b}$$

$$C=k_1^2\,\mathrm{Pr}_0\,T_0^2\cdot\left(p+(p_\theta-1)\cdot\frac{4}{k_1\,\mathrm{Pr}_0}\right). \tag{50c}$$

These quantities depend only on the flow numbers Pr_0, Ri_c, and Ri_0, since the closure parameter k_1 can be expressed by these numbers, $k_1=[2\,\mathrm{Pr}_0+3\,\mathrm{Ri}_c+4\,\mathrm{Ri}_0+3\,\mathrm{Ri}_0/Pr_0]/(Pr_0-Ri_0)$. Here $\mathrm{Ri}<\mathrm{Ri}_c$ appears as a condition for the solution of (49) for $p=p_\theta=1$. When the wind shear vanishes, we find from (47) a relation between $\tau^2\,\partial\Theta/\partial x^3$ and p and p_θ,

$$-\beta g\,\frac{\partial\Theta}{\partial x^3}\cdot\tau^2=\mathrm{Ri}_0\,T_0^2\cdot\frac{p+4/k_3\cdot(p_\theta-1)}{1+4\cdot T_0^2/k_1^2\cdot(\mathrm{Ri}_c\,\mathrm{Pr}_0+\mathrm{Ri}_0)/\mathrm{Pr}_0^2\cdot(p-1)}. \tag{51}$$

We note that real solutions of τ^2 only exist under unstable stratification. The values for the production–dissipation ratios for TKE and heat cannot be expected to be near unity, if considerable transports of these quantities occur, as for instance under convective conditions.[37] Since τ is only well defined by (47), if the dissipation is at least of the same order as the spatial transport, only ranges $0<(p,p_\theta)<2$ are considered in Figs. 1 and 2, where the Ri dependence of T is shown for $\mathrm{Ri}_c=0.3$, $\mathrm{Pr}_0=1$, and $\mathrm{Ri}_0=0.35$.[37] Here T becomes infinite at a critical number Ri that depends upon p and p_θ (and is equal to $\mathrm{Ri}_c=0.3$ for $p=p_\theta=1$). For increasing instability of stratification (Ri becomes smaller), $T=\tau\,\partial U/\partial x^3$ also becomes smaller. By assuming the wind shear $\partial U/\partial x^3$ as only a little influenced by the changing stratification, we find that the time scale $\tau=q^2/(2\langle\epsilon\rangle)$ of the TKE dissipation becomes

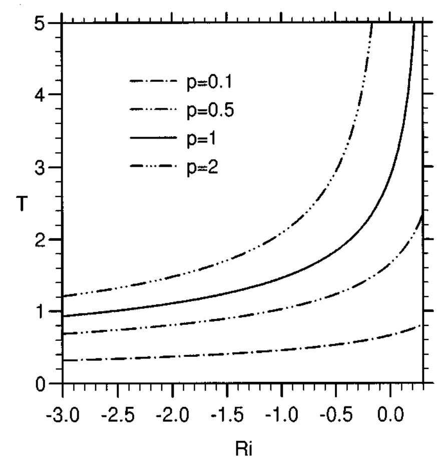

FIG. 1. The dependence of the normalized time scale $T=\tau\ \partial U/\partial x^3$ on the gradient Richardson number Ri for different production–dissipation ratios p for the TKE. The parameters are set to be $\mathrm{Ri}_c=0.3$, $\mathrm{Pr}_0=1$, and $\mathrm{Ri}_0=0.35$. The production–dissipation ratio for heat is assumed as balanced, which means $p_\theta=1$.

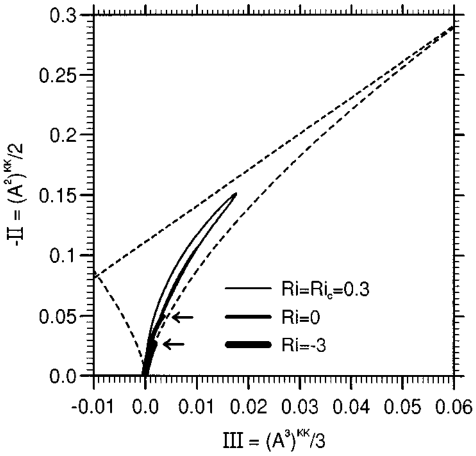

FIG. 3. The development of anisotropy calculated in the space of invariants II and III. Possible states of turbulence must lie within the Lumley triangle, which is depicted by the dashed lines. The solid lines give the evolution of the system for different gradient Richardson numbers Ri.

smaller by the enhanced intensity of turbulence. On the other hand, we find for growing values of the production–dissipation ratios for TKE and heat, p and p_θ, respectively, greater values of $T=\tau\ \partial U/\partial x^3$. Considering $\partial U/\partial x^3$ as constant, this is found to be plausible, because τ is expected to grow (the dissipation is diminished). Values greater than 1 for p and p_θ are found in particular near the surface, and values smaller than 1 are typical for the upper convective boundary layer for instance.[38,39] The estimation of τ explains well the basic features of buoyant plume rise in shear flows[14] and the development of anisotropy.[37]

For the further illustration of the equation (49) let us consider the described states of turbulence. This can be done by considering the turbulent motion in the space of invariants as often applied in the study of possible states of turbulence and conditions for the realizability of second-order closure models.[2,46] The anisotropy tensor $A^{ij}=V^{ij}/q^2-\frac{1}{3}\cdot\delta_{ij}$ provides two invariants, $\mathrm{II}=-(A^2)^{KK}/2$ and $\mathrm{III}=(A^3)^{KK}/3$.

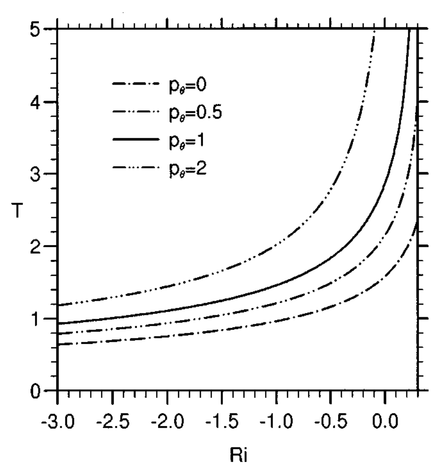

FIG. 2. The dependence of the normalized time scale $T=\tau\ \partial U/\partial x^3$ on the gradient Richardson number Ri for different production–dissipation ratios p_θ for heat. The parameters are as in Fig. 1 and the production–dissipation ratio for TKE is assumed as balanced, which means $p=1$.

These invariants have to be calculated by the second-order equation system (44a)–(44c), which depends upon the modified gradients of third moments $\hat{R}$, on the gradient Richardson number Ri and on p and p_θ, if (49) is applied for T. To close (44a)–(44c), the $\hat{R}$ will be neglected as justified for a homogeneous shear flow and T is calculated for balanced ratios for p and $p_\theta(p=p_\theta=1)$ in dependence on Ri. The equations (44a)–(44c) were solved numerically (by a Runge–Kutta procedure proved by the comparison with the analytical solution for Ri=0) under the assumption that the flow was initially isotropic, that means $\hat{V}^{ij}(t'=0)=1/3\ q_0^2\delta_{ij}$ were chosen for the initial values to solve (44a)–(44c). Here, q_0^2 is twice the TKE at the initial time and the chosen initial value of $\hat{V}^{44}$ leads only for $\mathrm{Ri}\neq 0$ to small variations in the initial stage of the calculated elements of A^{ij}, but does not influence the stationary values. This permits the calculation of the invariants as functions in time as shown in Fig. 3 for different gradient Richardson numbers Ri. The parameter values are $\mathrm{Pr}_0=1$, $\mathrm{Ri}_c=0.3$, and $\mathrm{Ri}_0=0.35$, as above. The dashed lines in Fig. 3 depict the Lumley triangle, where the limits describe two-dimensional turbulence (upper line) and axisymmetric states (the two other lines).[46] The motion starts in $(\mathrm{III},-\mathrm{II})=(0,0)$ and ends in stationary points $(\mathrm{III}_s,-\mathrm{II}_s)$, which appear as a consequence of the calculation of T. These stationary points are found for all gradient Richardson numbers Ri at the line

$$3\mathrm{III}_s+\frac{2}{k_1}\cdot\mathrm{II}_s+\frac{8}{9k_1^3}=0. \tag{52}$$

This is an important relation between these two invariants. It corresponds with the upper limit of the Lumley triangle for $k_1=2$. This result is only a consequence of the assumed balanced ratios $p=p_\theta=1$ for which T is calculated, since these conditions require that the stationary values of $\hat{R}$ are zero. This picture shows that the described states of turbulence are realizable. Whereas states for negative Ri reach directly its stationary point, it is shown in Fig. 4 for Ri very near Ri_c, that the state curve for positive values of Ri approaches

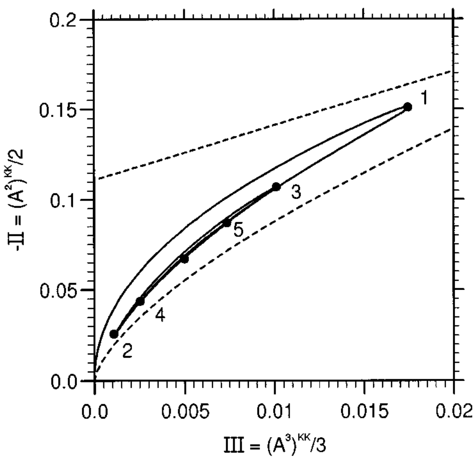

FIG. 4. The development of anisotropy in the space of invariants for $\mathrm{Ri}=\mathrm{Ri}_c$. The state curve approaches gradually to the stationary point. At first point 1 is reached, then the points 2,3,4,5 are passed, and the motion ends finally in the non-numbered stationary point between the points 4 and 5.

gradually to the stationary point. As the first point 1 is reached, the points 2, 3, 4, 5 are passed, and the motion ends finally in the non-numbered stationary point between the points 4 and 5. This qualitative different behavior seems to be caused by $\tau\rightarrow\infty$ for $\mathrm{Ri}\rightarrow\mathrm{Ri}_c$. A direction of turbulent motion is defined (aimed to the stationary state), if the time scale τ is finite. If τ becomes infinite, this directed motion diminishes and we find the oscillations as depicted in Fig. 4. This relation (52) can be used, for instance, for the calculation of k_1 by measurements of the invariants. Adopting the data $(A^{11},A^{22},A^{33},A^{13})=(0.137,-0.083,-0.053,-0.165)$ that were estimated by Champagne *et al.*[47] for $p=p_\theta=1$, one obtains $k_1=8.0$ in a good agreement with $k_1=8.3$, which follows from the applied values of the flow numbers and was used by Pope for simulations of developing anisotropy.[1]

VI. CONCLUDING REMARKS

Lagrangian models are very convenient tools for the investigation of the relations between production, dissipation, and turbulent transport of TKE as well as the description of fluid particle dispersion in complex flows. The derivation of such models is investigated here for non-neutral flows, i.e., the essential new aspect of the here presented equations is the incorporation of the potential temperature of particles. At first linear and nonlinear Markovian equations are derived in correspondence with the AHE (Secs. II and III). The AHE are taken in an approximation for the dissipation corresponding with Kolmogorov's theory and the simple Rotta model is used if pressure fluctuations have to be considered. By adopting these approximations, a time scale of the dissipation of TKE has to be determined. This is done in the fifth section by considering the production–dissipation ratio of TKE, where in particular nonlocal processes are taken into account in this time scale relation. The states of turbulence that are described in this way are found as realizable and as corresponding well with the Lumley theory,[46] where the simple relation (52) is found for the stationary values of the invariants. Nonlinearities of the velocity and potential temperature arise in the derived equations, if spatial gradients of the variances exist, as given by (19c). But the effects of these terms have to be further investigated, because the influence of these gradients can also be found in the drift terms of the equations that are linear in the velocity and potential temperature (and may be nonlinear in the particle position).

The nonuniqueness of these derived equations is discussed in the third section. It is shown that through the AHE only constraints appear for (symmetric) components of quantities that determine the particle properties (as, e.g., the matrix G, Sect. II). More explanation about these quantities can be expected, if the Lagrangian models are derived directly from the hydrodynamic equations. As a step into this direction, the derivation of (nonlinear and non-Markovian) stochastic equations according to concepts of the nonequilibrium statistical mechanics is investigated in Sec. IV. This permits the consideration of arbitrary colored noise and memory effects, which is important for the description of lower-Reynolds number flows. Sawford's equation[5,34] for the particle acceleration is derived and some consequences are discussed arising from the approach presented here. With respect to the nonuniqueness problem, it is worth emphasizing that the derived linear equations (1a)–(1b) with (4), (10), and (13) depend only on three flow numbers for balanced ratios of the production and dissipation of TKE and heat. This is formally analogous to the nonaveraged hydrodynamic equations, where the role of the molecular constants appearing in the latter equations is played now by the flow numbers in this larger scale. These equations are completely determined, because the mean values of wind and potential temperature can be calculated within the solution algorithm. This requires, first of all, the estimation of the mean pressure gradient, which can be calculated from Lagrangian quantities as observed by Pope.[17] Through their simplicity, these equations are very convenient to assess the effects of the other contributions appearing in (12). Further investigations in particular to the description of non-neutral stratified flows seem to be well suited to get further explanations to the performance of different Lagrangian models.

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