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Lagrangian particles with mixing. I. Simulating scalar transport

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The physical similarity and mathematical equivalence of continuous diffusion and particle random walk forms one of the cornerstones of modern physics and the theory of stochastic processes. The randomly walking particles do not need to posses any properties other than location in physical space. However, particles used in many models dealing with simulating turbulent transport and turbulent combustion do posses a set of scalar properties and mixing between particle properties is performed to reflect the dissipative nature of the diffusion processes. We show that the continuous scalar transport and diffusion can be accurately specified by means of localized mixing between randomly walking Lagrangian particles with scalar properties and assess errors associated with this scheme. Particles with scalar properties and localized mixing represent an alternative formulation for the process, which is selected to represent the continuous diffusion. Simulating diffusion by Lagrangian particles with mixing involves three main competing requirements: minimizing stochastic uncertainty, minimizing bias introduced by numerical diffusion, and preserving independence of particles. These requirements are analyzed for two limited cases of mixing between two particles and mixing between a large number of particles. The problem of possible dependences between particles is most complicated. This problem is analyzed using a coupled chain of equations that has similarities with Bogolubov–Born–Green–Kirkwood–Yvon chain in statistical physics. Dependences between particles can be significant in close proximity of the particles resulting in a reduced rate of mixing. This work develops further ideas introduced in the previously published letter [Phys. Fluids **19**, 031702 (2007)]. Paper I of this work is followed by Paper II [Phys. Fluids **19**, 065102 (2009)] where modeling of turbulent reacting flows by Lagrangian particles with localized mixing is specifically considered. © *2009 American Institute of Physics*.
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I. INTRODUCTION

Since the famous work of Einstein[1] on diffusion, the random walk of particles is conventionally considered as the stochastic process representing diffusion. The correspondence between random walk and diffusion equation was first established at a physical level in works of Einstein,[1] von Smoluchowski,[2] Fokker[3] and Planck,[4] and then was proven as a mathematical equivalence between Kolmogorov[5] diffusional equations and Ito[6] stochastic differential equations. In these approaches the particles are treated as elementary—particles have locations in physical space but they do not possess any internal scalar properties. The concentration of particles, which the diffusion equation is formulated for, is a group property possessed only by many particles.

The long-standing tradition of simulations of turbulent reacting flows[7,8] introduces particles of a different nature: these particles are treated as sufficiently macroscopic to possess some internal properties. The particles can move with turbulent flow (conventional Lagrangian particles or fluid particles) or, as introduced by Dreeben and Pope,[9] particle motion may involve some random walk generating fluctuations of particle positions around Lagrangian trajectories (diffusing Lagrangian particles). It is easy to see that diffusing Lagrangian particles represent a generalization of two fundamental concepts: elementary particles with random walk and Lagrangian (fluid) particles. In Lagrangian simulations, concentrations of reactive species are assigned to each individual particle. These concentrations can change due to chemical reactions or as the result of mixing when particles exchange fractions of their properties. The Lagrangian particles with mixing are commonly used as an effective numerical tool for approximate simulations of probability distributions in turbulent reacting flows. It seems, however, that the significance of Lagrangian particles with mixing extends beyond this role. Like the random walk of elementary particles, Lagrangian particles with mixing can also be considered as a process stochastically representing the diffusion equations. Lagrangian particles with mixing correspond to the physical understanding of turbulent reacting flows as a superposition of transport, reactions, and mixing while conventional random walk of elementary particles corresponds to treating reacting flows as a superposition of transport and reactions. In the latter case reactions result in the appearance and disappearance of elementary particles while mixing is not a fundamental process but one of the outcomes of the random walk. In turbulent flows, the mixing effect of molecular diffusion is always significant while spatial transport by molecular diffusion can often be neglected, and separate treatment of mixing and diffusional transport is a most logical step in this case (see Ref. 10 for further discussion of these issues).

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Major ideas, which are comprehensively examined in the present work, were introduced in the previously published letter.[11] The analysis presented here examines equations governing probability distributions and involves evaluations of the first, second, and the fourth moments of these equations. The multiparticle distribution equations form a chain which has similarities with the Bogolubov–Born–Green–Kirkwood–Yvon (BBGKY) chain well known in statistical physics.[12] Special attention is paid to the assumption of particle independence (chaos) which is commonly used in derivations related to interactions between particles and molecules (particle independence was simply assumed in Ref. 11). It is shown that this assumption is not always valid for Lagrangian particles with mixing and a quantitative affect of deviations from fully chaotic conditions is evaluated. The problem of preserving particle independence and at the same time minimizing numerical errors, which is of prime importance for numerical implementations, is considered and a direct comparison of efficiencies of different mixing schemes is carried out.

The following material is presented in this work. Section II introduces primary equations and mixing schemes. Section III evaluates mixing parameters and numerical diffusion on the basis of the hypothesis of particle independence when one-particle distribution is sufficient for derivations. One-particle distributions are always sufficient for mixing relaxing toward mean values.[13] Section IV introduces multiparticle distributions and a BBGKY-like chain for two-particle mixing.[14,15] Section V illustrates the effect of deviation from particle chaos in the case of spatial homogeneity. Section VI deals with nonchaotic effects in a general, nonhomogeneous case. Some of the most extensive considerations are moved from this section into Appendix. Using the developed theory, Sec. VII gives practical recommendations for numerical schemes based on Lagrangian particles. Major properties of particles with mixing are summarized in conclusions. This article represents Paper I of the work and is followed by Paper II,[16] which is dedicated to sparse-Lagrangian simulations of turbulent reacting flows where the number of particles is insufficient to resolve all scales in a turbulent flow.

II. PARTICLES WITH SCALAR PROPERTIES

A large number of particles, n, is moved stochastically in a d-dimensional physical space. This motion has a deterministic component determined by velocity of the flow and a random walk component. The random walk components are stochastically independent for different particles. The physical coordinates of pth particle are denoted as $\mathbf{x}_p = \{x_p^1, \ldots, x_p^d\}$. The flow velocity field is presumed to be fully specified. Each particle has n_s scalar properties $Z^{(1)}, \ldots, Z^{(n_s)}$. These properties change due to chemical reactions and due to mixing between the particles. Although particle positions in physical space are stochastically independent, values $Z_p^{(\alpha)}$ assigned to different particles may become dependent due to mixing. All particles are considered to be stochastically equivalent. This means that, at initial moment, particles are distributed in physical space stochastically and independently from each other. The properties $\mathbf{Z}$ are assigned to particle p according to its physical location $\mathbf{x}_p$ and irrespective of the number p. Hence the probability of observing one particle at a certain location and with certain properties is no different from the probability of this for another particle.

One of the advantages of particles with scalar properties and mixing is that many different scalar properties simulating many different scalars can be assigned to the same set of particles. This advantage is most significant when there is a need to simulate realistic reactions involving hundreds of species.

A. Main equations

The random walk of n particles with scalar properties and mixing is represented by the following equations:

$$dx_p^i = A^i(\mathbf{x}_p, t)dt + b^{ij}(\mathbf{x}_p, t)dw_p^j, \tag{1}$$

$$\frac{dZ_p^{(\alpha)}}{dt} = W_p^{(\alpha)} + \left[\frac{dZ_p^{(\alpha)}}{dt}\right]_{\text{mix}}, \tag{2}$$

where index p runs over all particles $p=1,\ldots,n$, index "α" runs over all scalars $\alpha=1,\ldots,n_s$, the vector $\mathbf{x}_p$ is pth particle position in d-dimensional physical space, w_p^j is the Wiener process with Eq. (1) being interpreted as Ito stochastic differential equation, $W_p^{(\alpha)}$ represents the effect of chemical reactions and the effect of mixing in Eq. (2) is symbolically denoted by the operator with subscript "mix." The mixing operator can create discontinuous trajectories—in this case generalized understanding of the time derivative in Eq. (2) is implied. The probability density function of a particle position in the physical space $P_x = P_x(\mathbf{x};t)$ satisfies the Fokker–Planck (direct Kolmogorov) equation

$$L[P_x] = 0, \quad L[\,\cdot\,] \equiv \frac{\partial(\,\cdot\,)}{\partial t} + \frac{\partial A^i(\,\cdot\,)}{\partial x^i} - \frac{\partial^2 B^{ij}(\,\cdot\,)}{\partial x^i\,\partial x^j}, \tag{3}$$

where $2B^{ml} = b^{mi}b^{li}$ is the diffusion coefficient, A^i is the drift coefficient and the differential operator $L[\cdot]$ is introduced to simplify notations. The summation convention over repeated indices applies here and further in the paper. The one-particle joint probability density function (PDF) $P_Z = P(\mathbf{Z},\mathbf{x};t)$ satisfies the equation

$$L[P_Z] + \frac{\partial W^{(\alpha)} P_Z}{\partial Z^{(\alpha)}} = \left[\frac{dP_Z}{dt}\right]_{\text{mix}}. \tag{4}$$

B. Interpretations in fluid mechanics

The first moment of Eq. (4) is of most interest in fluid mechanics since $\langle Z^{(\alpha)}\rangle$ is expected to be proportional to the αth scalar transported by the flow. This moment is obtained from Eq. (4) by multiplication by $Z^{(\alpha)}$ and integration over all $\mathbf{Z}$

$$L[\langle Z^{(\alpha)}\rangle P_x] - W^{(\alpha)} P_x = \left[\frac{d\langle Z^{(\alpha)}\rangle}{dt}\right]_{\text{mix}} P_x \approx 0. \tag{5}$$

As we expect that the mixing operator does not alter the mean values (at least to the leading order of the analysis), the right-hand side (rhs) of Eq. (5) is, essentially, zero. The "approximately equal" symbol used in this equation reflects nu-

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merical diffusion that exists as a side effect of mixing as discussed in Secs. III–VI. In fluid mechanics the diffusion coefficient is presumed isotropic $B^{kl}=D\delta^{kl}$. If all particles have the same weight, the properly normalized concentration of particles in physical space, which is proportional to P_x, is deemed to represent the density ρ. If density ρ and diffusivity D are constant, then the drift coefficient is the same as fluid velocity. Equations for a more general case of a compressible flow and particles with weights are given by[17,18]

$$\frac{\rho}{\rho_0} = \langle Z^{(0)}\rangle P_x, \qquad W^{(0)} = 0, \qquad Y^{(\alpha)} = \frac{\langle Z^{(\alpha)}\rangle}{\langle Z^{(0)}\rangle}, \tag{6}$$

$$A^i = U^i(\mathbf{x},t) + \frac{1}{\rho}\frac{\partial D\rho}{\partial x^i}, \qquad B^{ij} = \delta^{ij}D(\mathbf{x},t), \tag{7}$$

where ρ_0 is a normalization constant and the density ρ is linked to particle weight $Z^{(0)}$—the fraction of the total mass carried by a particle. From our perspective this value is just one of the scalars $Z^{(\alpha)}$. This scalar can be mixed in the same way as other scalars or remain unchanged for a given particle (note that this scalar has a distinctive condition of $W^{(0)}=0$, unless sources or sinks of mass are present in the flow). The value $Z_p^{(\alpha)}$ can be referred to as the amount scalar α in particle p and $Y^{(\alpha)}$ is treated as the mass concentration of scalar α. Substitution of definitions (6) and (7) into Eq. (5) with nullified rhs results in the conventional equations of continuity and scalar transport

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho U^i}{\partial x^i} = 0, \tag{8}$$

$$\frac{\partial \rho Y^{(\alpha)}}{\partial t} + \frac{\partial \rho Y^{(\alpha)} U^i}{\partial x^i} - \frac{\partial}{\partial x^i}\left(D \frac{\partial Y^{(\alpha)}}{\partial x^i} \right) = 0. \tag{9}$$

Although Lagrangian particle methods are specifically designed to handle multiple scalars $Z^{(\alpha)}$, we from this point consider only one selected scalar and omit the superscript index (α) for simplicity. The conclusions, of course are applicable to any of the scalars. The proof of convergence to scalar transport equations requires assessment of the mixing operator $[d\langle Z\rangle/dt]_{\mathrm{mix}}$ in rhs of Eq. (5) and estimation of the stochastic error $\langle z^2\rangle$, where $z \equiv Z - \langle Z\rangle$. In the first part of this work z is interpreted as a stochastic deviation from the desired mean value implying that the goal of simulations is full resolution of the scalar fields. The second part of this work[16] deals with sparse-Lagrangian simulations where z is interpreted as a model for subscale fluctuations of the scalars. It should be noted that, although the exact mean values of $\langle Z\rangle$ and $\langle z^2\rangle$ may remain unknown due to a limited number of particles in a small volume, these values do exist and the required statistics can be accumulated when needed. The stochastic errors in estimating means from Lagrangian particle data are not discussed here and are analyzed in Ref. 19, where it is shown that evaluation of averages over particle properties can have a stochastic error that is noticeably smaller than stochastic errors associated with evaluation of averages for concentration of particles.

C. Equations for higher moments

Among higher moments, the second and fourth moments are most important for our purposes. The second moment determines stochastic variations around means while the flatness factor $\Omega \equiv \langle z^4\rangle / \langle z^2\rangle^2$ is used as an indicator of deviations from Gaussian distributions. Some of the mixing operators are known to increase the flatness factor well above the Gaussian value of $\Omega = 3$. The equations for higher moments can be obtained by integrating Eq. (4):

$$L[\langle Z^m\rangle P_x] = mW\langle Z^{m-1}\rangle P_x + \left[\frac{d\langle Z^m\rangle}{dt}\right]_{\mathrm{mix}} P_x. \tag{10}$$

With the use of Eq. (3), these equations can be equivalently represented by

$$\tilde{L}[\langle Z^m\rangle] = mW\langle Z^{m-1}\rangle + \left[\frac{d\langle Z^m\rangle}{dt}\right]_{\mathrm{mix}}, \tag{11}$$

where a new differential operator $\tilde{L}[\,\cdot\,]$ is introduced by

$$\tilde{L}[\,\cdot\,] \equiv \frac{\partial(\,\cdot\,)}{\partial t} + \tilde{A}^i \frac{\partial(\,\cdot\,)}{\partial x^i} - B^{ij}\frac{\partial^2(\,\cdot\,)}{\partial x^i\,\partial x^j}, \qquad \tilde{A}^i \equiv A^i - \frac{2}{P_x}\frac{\partial B^{ij}P_x}{\partial x^j}. \tag{12}$$

Equations (4) and (11) can be expressed in terms of the variable z representing fluctuations of the scalar $z \equiv Z - \langle Z\rangle$ and the scalar dissipation χ defined below,

$$L[P_z]_z - \chi\frac{\partial^2 P_z}{\partial z^2} + \frac{\partial W P_z}{\partial z} = \left[\frac{dP_z}{dt}\right]_{\mathrm{mix}}, \qquad \chi \equiv B^{ij}\frac{\partial\langle Z\rangle}{\partial x^i}\frac{\partial\langle Z\rangle}{\partial x^j}, \tag{13}$$

$$\tilde{L}[\langle z^m\rangle] = m(m-1)\chi\langle z^{m-2}\rangle + mW\langle z^{m-1}\rangle + \left[\frac{d\langle z^m\rangle}{dt}\right]_{\mathrm{mix}}. \tag{14}$$

D. Mixing

If a particle has a nonzero probability of being mixed with another particle we refer to this as mixing interaction between the particles. These interactions need to be distinguished from particle collisions (particles move independently in physical space and do not collide) and from actual mixing events. For example, a particle may interact with several particles simultaneously but only allowed to be mixed with a single counterpart at a time. Each mixing event changes the values of n_m particles forming a mixing group according to the equation.

$$Z_p^{\mathrm{mix}} = Z_m + \mu(Z_p^{\circ} - Z_m), \qquad Z_m \equiv \frac{1}{n_m}\sum_{p}^{n_m} Z_p^{\circ}, \tag{15}$$

This equation describes n_m-particle mixing. The superscript "∘" is used to emphasize that this value is taken before the mixing event. The parameter μ specifies the extent of mixing with $\mu=0$ corresponding to complete mixing and $\mu=1$ corresponding to no mixing at all. This value can be stochastic, although in this case μ is stochastically independent of Z. It is easy to see that mixing specified by Eq. (15) is compliant with the conditions of "good mixing" listed in Refs. 7, 8, and

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11. In the present work, unlike in Ref. 11, we have chosen more specific mixing schemes to analyze more quantitative characteristics of particles with mixing. We consider only two limiting cases of Eq. (15): a very large n_m and the smallest possible n_m of 2. In the former case Z_m is essentially the same as $\langle Z\rangle$ due to averaging over a large number of particles,

$$Z_p^{[\langle Z\rangle]} = Z_m + \mu(\mathring{Z}_p - Z_m) = \eta Z_m + \mu \mathring{Z}_p, \quad Z_m = \langle Z\rangle, \tag{16}$$

where $\eta \equiv 1-\mu$ varies between 0 (no mixing) and 1 (complete mixing). The index $[\langle Z\rangle]$ indicates that the model involves evaluation of the mean and has to use a large number of particles in each mixing event. This mixing is referred to as mean-based mixing. For the case of $n_m=2$ and $Z_m=(Z_p+Z_q)/2$ the pth value after mixing particle p with particle q is given by

$$Z_{pq}^{[2]} = \frac{1+\mu}{2}\mathring{Z}_p + \frac{1-\mu}{2}\mathring{Z}_q = \mathring{Z}_p + \tilde{\eta}(\mathring{Z}_q - \mathring{Z}_p), \tag{17}$$

where mixing parameters are related to each other by

$$\tilde{\eta} \equiv \frac{1-\mu}{2} = \frac{\eta}{2}, \quad \eta + \mu = 1 \tag{18}$$

and the index [2] indicates that only two particles are allowed to participate in each nixing event. Two-particle mixing is known as Curl's mixing[14,15] while the model specified by Eq. (16) is very similar to IEM model (interactions by exchange with the mean[13]). Conventional Curl's and IEM models also differ by smoothness of their trajectories: IEM model is continuous while scalar values in Curl's model change in a series of jumps. In the present work, models (16) and (17) are distinguished only by the number of particles n_m participating in each mixing event while the continuity of generated trajectories is simply determined by the value of the model parameter η: the jumps are most significant when $\eta=1$ and trajectories become continuous as $\eta \to 0$. In general, parameter η can also be selected randomly (as in the modified Curl's model[14,15])—in this case its value is presumed to be stochastically independent of Z_p.

Equations (16) and (17) specify a single mixing event while the overall effect of mixing is proportional to parameter $\hat{\theta}$ that determines intensity of mixing events. The exact probabilistic meaning of parameter $\hat{\theta}$ is given in the section dealing with PDF formulation for the mixing operator. The intensity of mixing $\hat{\theta}=\hat{\theta}(\mathbf{x},t)$ may depend on location $\mathbf{x}$ and time t.

III. ANALYSIS OF MIXING UNDER CHAOTIC CONDITIONS

Analysis of mixing operators is conventionally performed under assumptions that we call here the chaos assumption. Although particles move independently from each other and particle locations $\mathbf{x}_p$ and $\mathbf{x}_q$ are stochastically independent when $p \neq q$, the scalar values Z_p and Z_q that belong to different particles may become stochastically dependent as the result of mixing. However, assuming that Z_p and Z_q can be treated as independent values when $p \neq q$ significantly simplifies the analysis. This means that correlations $\langle z_p z_q\rangle$, $\langle z_p^3 z_q\rangle$ and similar can be neglected when $p \neq q$. In Secs. III–VI we demonstrate that the chaos assumption is always valid for mean-based mixing but under certain conditions may lead to incorrect results for two-particle mixing. This section follows previous publications and derives major moment equations for the case when the chaos assumption is valid. The effect of numerical diffusion associated with mixing is also evaluated here.

A. Mixing effect on variance and flatness

Equations for the effect of the mixing on the second and fourth moments can generally be written in the form

$$\left[\frac{d\langle z^2\rangle}{dt}\right]_{\mathrm{mix}} = -2\frac{\langle z^2\rangle}{\tau}, \quad \left[\frac{d\langle z^4\rangle}{dt}\right]_{\mathrm{mix}} = -4\frac{\langle z^4\rangle}{\tau\omega}. \tag{19}$$

These equations effectively define the dissipation time τ and the dissipation time for the fourth moment $\tau\omega$. We note that quasistationary formulas for the second and fourth moments and the flatness factor Ω are expressed in terms of the parameters τ and ω

$$\langle z^2\rangle = \chi\tau, \quad \langle z^4\rangle = 3\chi\langle z^2\rangle\tau\omega, \quad \Omega \equiv \frac{\langle z^4\rangle}{\langle z^2\rangle^2} = 3\omega. \tag{20}$$

These formulas nullify rhs of Eq. (14) assuming that $W=0$. The flatness factor Ω is an indicator of deviation of probabilistic distributions from Gaussian.

The moment equations were previously given in several publications[20,21] for different mixing models. These equations can be easily obtained from Eqs. (17) and (16):

$$\left[\frac{d\langle z_1^m\rangle}{dt}\right]_{[\langle Z\rangle]} = -\hat{\theta}(1-\langle\mu^m\rangle)\langle z_1^m\rangle, \tag{21}$$

$$\left[\frac{\partial\langle z_1^m\rangle}{\partial t}\right]_{[2]} = -\hat{\theta}\langle z_1^m - (z_1(1-\tilde{\eta}) + \tilde{\eta} z_2)^m\rangle. \tag{22}$$

Here we retain particle indices to distinguish values related to different particles. Since all particles are stochastically equivalent, particles marked by indices 1 and 2 are selected as representative. If η is small the rhs of Eqs. (21) and (22) simplify into $-m\hat{\theta}\langle\eta\rangle\langle z^m\rangle$ and $-m\hat{\theta}\langle\tilde{\eta}\rangle\langle z^m\rangle$, respectively. This corresponds to continuous mixing which is used in the conventional IEM model. Under chaos assumptions the moment equations are simplified and yield the following values of the parameters τ and ω for

$$\tau = \frac{2}{\hat{\theta}\langle\gamma\rangle}, \quad \omega = \frac{2\langle\gamma\rangle}{\langle\gamma(2-\gamma)\rangle} \quad \text{for } n_m = \infty, \tag{23}$$

$$\tau = \frac{4}{\langle\gamma\rangle\hat{\theta}}, \quad \omega = \frac{8\langle\gamma\rangle}{\langle\gamma(8-\gamma)\rangle - 3\langle\gamma^2\rangle/\Omega} \quad \text{for } n_m = 2, \tag{24}$$

$$\gamma \equiv 1 - \mu^2 = 2\eta - \eta^2.$$

We consider quasisteady values of the moments that are determined by the balance of generation and dissipation terms in Eq. (14) assuming that $W=0$. Note that this case is inho-

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mogeneous and different from the asymptotic behavior of the moments in homogeneous turbulence previously considered by Pope.[20] Quasisteady values of the flatness factor are given by

$$\Omega_{[\langle Z\rangle]} = \frac{6\langle\gamma\rangle}{\langle\gamma(2-\gamma)\rangle}, \tag{25}$$

$$\Omega_{[2]} = 3\frac{\langle\gamma(8+\gamma)\rangle}{\langle\gamma(8-\gamma)\rangle}. \tag{26}$$

As γ varies from $\gamma\to 0$ to $\gamma=1$, the quasisteady value of the flatness factor Ω changes from $\Omega=3$ to $\Omega=6$ for $n_m\to\infty$ and from $\Omega=3$ to $\Omega=27/7$ for $n_m=2$.

B. Mixing and numerical diffusion

The mixing operator generally preserves the value of the first moment as indicated by Eq. (5). This is indeed the case, if the following values:

$$\left[\frac{d\langle Z_1\rangle}{dt}\right]_{[\langle Z\rangle]} = \hat{\theta}\langle\eta\rangle\langle Z_m - Z_1\rangle, \tag{27}$$

$$\left[\frac{d\langle Z_1\rangle}{dt}\right]_{[2]} = \hat{\theta}\langle\tilde{\eta}\rangle\langle Z_2 - Z_1\rangle_{V_1} \tag{28}$$

are zeros. If $Z_m=\langle Z\rangle$ then $\langle Z_1-Z_m\rangle=\langle z_1\rangle=0$ and ideal mean-based mixing does not alter $\langle Z\rangle$. However, the mean value $\langle Z\rangle$ is not known and has to be evaluated by averaging over sufficiently large number of particles and it is inevitable that these particles have some dispersion in physical space. In the same way, if two-particle mixing is allowed only when particles 1 and 2 are located at the same point then $\langle Z_2-Z_1\rangle=\langle Z_2\rangle-\langle Z_1\rangle=0$ and ideal two-particle mixing preserves $\langle Z\rangle$ but in multidimensional spaces, positions of different particles never coincide exactly. Due to a limited number of particles used in simulations, mixing or averaging has to be performed over a certain finite volume which is denoted here as V_1. This volume may be small but it cannot be shrunk into a point in practical simulations. The mixing terms in Eqs. (27) and (28) vanish at the leading order but we show that the higher-order terms result in numerical diffusion effects. These effects are neglected in the analysis of higher moments since mixing directly influences these moments but the prime effect of mixing on the first moment is that of numerical diffusion.

For mean-based mixing, the value Z_m has to be evaluated over a certain volume V_1

$$Z_m = \frac{\int_{V_1}\langle Z\rangle P_x d\mathbf{x}}{\int_{V_1} P_x d\mathbf{x}}.$$

When mixing (or averaging) volume is difficult to link to a specific particle p, we denote this volume by V_m. In order to minimize numerical diffusion, two-particle mixing should be permitted only when the particles are sufficiently close to each other. In Eq. (28) this is shown by the index V_1 which indicates that the difference is evaluated over a certain volume V_1 so that

$$\langle Z - Z_1\rangle_{V_1} = \frac{1}{V_1 P_{x1}}\int_{V_1}(\langle Z\rangle - \langle Z\rangle_1)P_x d\mathbf{x}.$$

Particle 1 is presumed to be located in the center of the volume V_1. Here, we use subscript "1" denoting values related to the location of particle 1 but omit indices of other particles. We obtain at the leading order

$$Z_m = \langle Z\rangle_1 + \frac{I^{ij}}{2P_{x1}}\left(\frac{\partial^2\langle Z\rangle P_x}{\partial x^i\,\partial x^j} - \langle Z\rangle\frac{\partial^2 P_x}{\partial x^i\,\partial x^j}\right)_1,$$

$$\langle Z - Z_1\rangle_{V_1} = \frac{I^{ij}}{2}\left(\frac{\partial^2\langle Z\rangle}{\partial x^i\,\partial x^j} + \frac{2}{P_x}\frac{\partial P_x}{\partial x^i}\frac{\partial\langle Z\rangle}{\partial x^j}\right)_1,$$

$$I^{ij} \equiv \frac{\int_{V_1} y^i y^j dV}{V_1}, \quad y^i = x^i - x_1^i.$$

In these equations integrals are evaluated by expanding $\langle Z\rangle$ and P_x into a Taylor series around the center of the volume, for example

$$\langle Z\rangle = \langle Z\rangle_1 + \left(\frac{\partial\langle Z\rangle}{\partial x^i}\right)_1 y^i + \frac{1}{2}\left(\frac{\partial^2\langle Z\rangle}{\partial x^i\,\partial x^j}\right)_1 y^i y^j + \cdots.$$

This expansion and estimation of deviations of Z_m from $\langle Z\rangle$ follows Pope's [see Ref. 22, Eq. (12.251)] assessment of errors associated with kernel estimates. The linear terms disappear from integrals and the largest correcting term is proportional to I^{ij}. Note that derivatives of the PDF P_x can be eliminated by assuming that particle 1 is located at the P_x-weighted center of the volume V_1. We now assume that volume V_1 represents a sphere of radius r_m with the center located at the particle 1 location $\mathbf{x}_1$ so that $|\mathbf{x}-\mathbf{x}_1|\leq r_m$. The moment of inertia of the volume V_1 can now be expressed as $I^{ij}=\hat{r}_1^2\delta^{ij}$ where

$$\hat{r}_1^2 = \frac{\hat{r}^2}{d} = \frac{1}{d}\frac{\int_0^{r_m} r^2 r^{d-1}dr}{\int_0^{r_m} r^{d-1}dr} = \frac{r_m^2}{d+2},$$

$\hat{r}^2$ is average value of the square of a radius within the volume V_1, and $\hat{r}_1^2=\hat{r}^2/d$ is the average of the squared value of the radius projection on a given direction. The dimension of the space is denoted by d. With correcting terms, the mixing operator takes the same form for both two-particle and mean-based mixing

$$\left[\frac{d\langle Z\rangle}{dt}\right]_{\text{mix}} = D_m\left(\frac{\partial^2\langle Z\rangle}{\partial x^i\,\partial x^i} + \frac{2}{P_x}\frac{\partial P_x}{\partial x^i}\frac{\partial\langle Z\rangle}{\partial x^i}\right), \tag{29}$$

$$D_m \equiv \frac{\hat{r}_1^2}{\tau\beta} = \frac{r_m^2}{(d+2)\tau\beta}, \quad \beta = \frac{\langle\gamma\rangle}{\langle\eta\rangle} = \frac{2\langle\gamma\rangle}{\langle\tilde{\eta}\rangle}. \tag{30}$$

The correcting terms result in additional diffusion which can be referred to as numerical diffusion. Numerical diffusion is characterized by the coefficient D_m. Another term represents a drift correction that effectively results in replacing D by effective diffusion $D_{\text{eff}}=D+D_m$ in formula for $\tilde{A}^i$. Numerical diffusion decreases with improved localization and decrease in r_m. Intensification of mixing and reduction in τ increases numerical diffusion. In general, numerical diffusion can be used to simulate diffusion (differential diffusion effects, for

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example). In this case it is more appropriate to label this diffusion as diffusion induced by mixing since the term numerical diffusion indicates that it is preferable to reduce this type of diffusion as much as possible. It should be remembered, however, that the coefficient of mixing-induced diffusion may differ for different moments $\langle Z^m \rangle$.

Equation (30) for the coefficient of numerical diffusion D_m coincides for both models and with equation for D_m obtained in Ref. 11 from more general but more intuitive considerations. The mean-based mixing is probably more complicated to implement compared to two-particle mixing but, surprisingly, mean-based mixing does not offer any reduction in numerical diffusion for the same r_m and τ. Note that particle 1 is assumed to be at the center of volume V_1 when Z_m is evaluated for mean-based mixing in Eq. (27) (this is, typically, the case in the IEM model). A fully conservative mean-based mixing requires Z_m to be the same for all particles of a selected mixing group located within a certain volume V_m and, in this case, a particle member of the group can be located anywhere in the volume V_m. This results in twofold increase in the effective values of $\hat{r}_1^2$ and D_m while the radius r_m of the volume remains fixed. Mixing also introduces some drift terms in Eq. (29) related to gradients of the PDF P_x. These terms can be eliminated by placing particle 1 at the P_x-weighted center of the mixing volume—the details of numerical diffusion depend on details of mixing but numerical diffusion with the diffusion coefficient given by Eq. (30) is ubiquitously present as long as particles are spaced apart by distance $\hat{r}_1$ during mixing events.

IV. PDF FORMULATION OF THE MIXING OPERATOR

Mixing between particles can generally be considered to be a Poisson stream of mixing events (see Ref. 7). This is more convenient for theoretical considerations, although in numerical implementations mixing events between different particles are usually synchronized with the time step. In this section we give a formulation of the problem in terms of PDF (probability density functions). The equations governing the combined Markov process (continuous diffusional process with respect to variables x^i and discontinuous Poisson process with respect to Z) were introduced by Pope.[7]

A. Mean-based mixing

The mean-based mixing scheme (16) allows for closed formulation using one-particle PDF $P_Z = P_Z(Z, \mathbf{x}; t)$ which satisfies the equation

$$L[P_Z] + \frac{\partial W P_Z}{\partial Z} = \hat{\theta} \int P_t(Z|Z^\circ) P_Z(Z^\circ) dZ^\circ . \tag{31}$$

Intensity of the mixing process is determined by parameter $\tilde{\theta}$. This equation corresponds to a Markov process that combines two parts: continuous diffusion in physical space specified by lhs of this equation and Poisson jumps of values Z reflected by integral on rhs. The PDF $P_t(Z|Z^\circ)$ is the transitional probability of mixing process given by

$$P_t(Z|Z^\circ) = \int \delta(Z - Z^{[\langle Z \rangle]}) P_\mu(\mu) d\mu - \delta(Z - Z^\circ), \tag{32}$$

where δ is Dirac's delta function, $P_\mu(\mu)$ is the PDF of μ and $Z^{[\langle Z \rangle]}$ is the value after mean-based mixing given by Eq. (16). The subscript p is omitted since only a single particle is considered here. Evaluation of moments of the PDF Eq. (31) results in the formulas considered in Sec. III.

The closed equation for the one-particle PDF can be rigorously obtained for the case when $Z_m = \langle Z \rangle$ in Eq. (16). Practically, Z_m is evaluated over $n_m < \infty$ particles resulting in some fluctuations of Z_m over $\langle Z \rangle$ so that $z_m = Z_m - \langle Z \rangle \neq 0$. If n_m is large then $\langle z_m^2 \rangle \sim \langle z^2 \rangle / n_m$ is small and the practical effect of these fluctuations is a small increase in the mixing time (23).

B. Two-particle mixing and BBGKY-like chain

In the rest of this article we mainly deal with the case of two-particle mixing[14,15] when $n_m = 2$ and the mixing event is specified by Eq. (17). Although a group of three or more particles is not allowed to be mixed with each other at the same moment, many particles can be simultaneously engaged in mixing interactions (i.e., many particles may have a nonzero probability of being mixed with a given particle). We use the term "mixing interactions" to characterize a nonzero probability of mixing between two particles and the term "mixing event" to characterize the actual act of mixing between these particles. A particle can have mixing interactions with many particles (even with all particles if mixing is not localized) but the probability of simultaneous mixing with more than one particle is zero and mixing events occur only between couples. Even if the initial conditions can be specified independently for each particle, stochastic dependence between Z_p and Z_q appears after mixing of particles p and q. Thus the scalar values at different particles are stochastically dependent and can be rigorously characterized only by the all-particle PDF $P_n(Z_1, \ldots, Z_n, \mathbf{x}_1, \ldots, \mathbf{x}_n)$. We assume that n particles are present in the flow. These particles are treated as stochastically equivalent, hence, P_n is symmetric and does not change when two particles are swapped. For example, $P_n(Z_1, \ldots, Z_n, \mathbf{x}_1, \ldots, \mathbf{x}_n) = P_n(Z_n, \ldots, Z_1, \mathbf{x}_n, \ldots, \mathbf{x}_1)$.

$$L_n^+[P_n] = \sum_{(p,q)=1}^{n} \theta_{pq} \int P_t(Z_p, Z_q | Z_p^\circ, Z_q^\circ) \times P_n(Z_1, \ldots, Z_p^\circ, \ldots, Z_q^\circ, \ldots, Z_n, \mathbf{x}_1, \ldots, \mathbf{x}_n) dZ_p^\circ dZ_q^\circ, \tag{33}$$

$$P_t(Z_p, Z_q | Z_p^\circ, Z_q^\circ) = \int \delta(Z_p - Z_{pq}^{[2]}) \delta(Z_q - Z_{qp}^{[2]}) \times P_{\tilde{\eta}}(\tilde{\eta}) d\tilde{\eta} - \delta(Z_p - Z_p^\circ) \delta(Z_q - Z_q^\circ),$$

where sum is taken over $n(n-1)$ couples that can be formed by n particles, the intensity of mixing θ_{pq} is defined by $\theta_{pq} = \theta(\mathbf{x}_p, \mathbf{x}_q)$, the mixing operation $Z_{pq}^{[2]}$ is specified previously in Eq. (17), the following mean-based diffusion operators are used here and further in the paper:

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$$
\begin{aligned}
L_k^{+}[\,\cdot\,] &= L_k[\,\cdot\,] + \sum_{p=1}^{k} \frac{\partial(\,\cdot\,)W_p}{\partial Z_p},\\
L_k[\,\cdot\,] &\equiv \sum_{p=1}^{k}\left(\frac{\partial(\,\cdot\,)}{\partial t} + \frac{\partial A_p^i(\,\cdot\,)}{\partial x_p^i} - \frac{\partial^2 B_p^{ij}(\,\cdot\,)}{\partial x_p^i\,\partial x_p^j}\right),
\end{aligned}
\tag{34}
$$

$$
\widetilde{L}_k[\,\cdot\,] \equiv \sum_{p=1}^{k}\left(\frac{\partial(\,\cdot\,)}{\partial t} + \widetilde{A}_p^i\frac{\partial(\,\cdot\,)}{\partial x^i} - B_p^{ij}\frac{\partial^2(\,\cdot\,)}{\partial x_p^i\,\partial x_p^j}\right). \tag{35}
$$

The intensity of mixing is presumed to be localized in physical space, that is,

$$
\begin{aligned}
\theta_{pq} &= \theta(\mathbf{x}_p - \mathbf{x}_q) = \left\{\begin{matrix} \theta_0, & |\mathbf{x}_p - \mathbf{x}_q| \le r_m \\ 0, & |\mathbf{x}_p - \mathbf{x}_q| > r_m \end{matrix}\right\},\\
\theta_V &= \int \theta_{12} P_x(\mathbf{x}_2)d\mathbf{x}_2 = \theta_0 P_x(\mathbf{x}_1)V_1,\\
\hat{\theta} &= \theta_V(n-1) = \theta_0 n_a, \quad n_a \equiv (n-1)V_1 P_x(\mathbf{x}_1).
\end{aligned}
\tag{36}
$$

This means that particle p has a positive probability of being mixed (has mixing interactions) only with the average of n_a other particles located in the mixing volume V_p represented by r_m-vicinity of $\mathbf{x}_p$. The equation for one-particle distribution $P_1(Z_1,\mathbf{x}_1)$ can be easily obtained from Eq. (33) by integrating over the variables representing particles $2,\ldots,n$,

$$
\begin{aligned}
L_1^{+}[P_1] &= (n-1)\int \theta_{12} P_t(Z_1|\mathring{Z}_1,\mathring{Z}_2)\\
&\quad \times P_2(\mathring{Z}_1,\mathring{Z}_2,\mathbf{x}_1,\mathbf{x}_2)d\mathring{Z}_1 d\mathring{Z}_2 d\mathbf{x}_2,\\
P_t(Z_p|\mathring{Z}_p,\mathring{Z}_q) &= \int P_t(Z_p,Z_q|\mathring{Z}_p,\mathring{Z}_q)dZ_q,
\end{aligned}
\tag{37}
$$

but this equation involves the two-particle PDF P_2 and thus is not closed. The PDF P_1 is essentially the same as P_Z but we use P_1 to emphasize that the equation for this one-particle PDF is not closed. Attempts to obtain equations for higher-order distributions $k=2,3,\ldots$ results in the following chain of equations:

$$
\begin{aligned}
L_k^{+}[P_k] = \sum_{(p,q)=1}^{k} \theta_{pq} &\int P_t(Z_p,Z_q|\mathring{Z}_p,\mathring{Z}_q)\\
&\times P_k(Z_1,\ldots,\mathring{Z}_p,\ldots,\mathring{Z}_q,\ldots,Z_k,\mathbf{x}_1,\ldots,\mathbf{x}_k)\\
&\times d\mathring{Z}_p d\mathring{Z}_q + (n-k)\sum_{p=1}^{k}\int \theta_{p,k+1} P_t(Z_p|\mathring{Z}_p,\mathring{Z}_{k+1})\\
&\times P_{k+1}(Z_1,\ldots,\mathring{Z}_p,\ldots,\mathring{Z}_{k+1},\mathbf{x}_1,\ldots,\mathbf{x}_{k+1})\\
&\times d\mathring{Z}_p d\mathring{Z}_{k+1} d\mathbf{x}_{k+1},
\end{aligned}
\tag{38}
$$

where every equation for $k=1,\ldots,n-1$ is not closed since equation for P_k depends on P_{k+1} in the second integral. Only at $k=n$ the fully closed Eq. (33) is obtained but this equation is difficult to use in practice due to a very large number of independent variables. This problem is similar to the well-known BBGKY chain of equations that plays an important role in statistical physics. The BBGKY chain appears in derivation of Boltzmann equation for one-molecule distributions from Liouville equation for the joint distribution of all molecules.[12] The hypothesis of molecular chaos is the fundamental physical assumption that is used in the derivation of the Boltzmann equation. According to this hypothesis, characteristics of different molecules can be effectively treated as stochastically independent from each other. In application to our particles with mixing, this hypothesis can be written as

$$
P_k(Z_1,\ldots,Z_k,\mathbf{x}_1,\ldots,\mathbf{x}_k) = P_1(Z_1,\mathbf{x}_1),\ldots,P_1(Z_k,\mathbf{x}_k) \tag{39}
$$

for any k from the range $1 \le k \le n$. Note that the PDF of particle locations $P_x(\mathbf{x}_1,\ldots,\mathbf{x}_k)$ can always be written as $P_x(\mathbf{x}_1)\ldots P_x(\mathbf{x}_k)$ since particle motions in physical space are stochastically independent. The conventional treatment of two-particle mixing is based using one-particle PDF and assuming stochastic independence (particle chaos assumption) is generally reasonable but may result in significant errors under certain conditions discussed in Secs. V–VII. Although we use some of the terminology that is conventionally used in statistical physics, it is important to remember that we consider objects (Lagrangian particles) and equations that are different from these considered in statistical physics.

C. Divergent form of the mixing operator

The normalization conditions imposed on PDFs and conservative properties of mixing indicate that

$$
\int \left[\frac{dP_Z}{dt}\right]_{\text{mix}} dZ = 0, \tag{40}
$$

$$
\int Z\left[\frac{dP_Z}{dt}\right]_{\text{mix}} dZ = \left[\frac{d\langle Z\rangle}{dt}\right]_{\text{mix}} \approx 0. \tag{41}
$$

The approximate equality is used in the second equation to indicate that numerical diffusion is neglected. The mixing operator can be written in the divergent form

$$
\left[\frac{dP_Z}{dt}\right]_{\text{mix}} = \frac{\partial J}{\partial Z} \approx \frac{\partial^2 N}{\partial Z^2} \tag{42}
$$

and it is easy to see that this representation is compliant with restrictions (40) and (41). In the case of a single variable Z, the equations for the functions J and N can be explicitly represented in the same form as the rhs of Eqs. (31) and (37) but the transitional PDF P_t replaced by its integrals over Z.

The situation is more complicated in the case of $n_s > 1$ scalar variables $Z^{(1)},\ldots,Z^{(n_s)}$. The mixing operator can still be represented by a divergent expression

$$
\left[\frac{dP_Z}{dt}\right]_{\text{mix}} = \frac{\partial J^{(\alpha)}}{\partial Z^{(\alpha)}} \approx \frac{\partial^2 N^{(\alpha\beta)}}{\partial Z^{(\alpha)}\,\partial Z^{(\beta)}} \tag{43}
$$

but this representation is not unique since a solenoidal vector field in Z-space can always be added to $J^{(\alpha)}$ without changing the mixing operator. A unique divergent representation of the mixing operator can however be obtained in the limiting case of $\eta \to 0$. The following continuous mixing operators:

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$$\left[\frac{dP_Z}{dt}\right]_{[\langle Z\rangle]} = \frac{\partial}{\partial Z^{(\alpha)}}\left(\frac{\langle Z^{(\alpha)}\rangle - Z^{(\alpha)}}{\tau} P_Z\right), \tag{44}$$

$$\left[\frac{dP_1}{dt}\right]_{[2]} = \frac{\partial}{\partial Z_1^{(\alpha)}}\int \theta_{12}\frac{Z_2^{(\alpha)} - Z_1^{(\alpha)}}{\tau} P_2(\mathbf{Z}_1,\mathbf{Z}_2,\mathbf{x}_1,\mathbf{x}_2)\,d\mathbf{Z}_2\,d\mathbf{x}_2, \tag{45}$$

have the same expressions for all moments as Eqs. (21) and (22) at the limit of $\eta,\tilde{\eta}\rightarrow 0$. This means that Eqs. (44) and (45) represent the divergent forms of the mixing operators for the case of small η. The mixing time τ is defined by $\tau^{-1}=\hat{\theta}\langle\eta\rangle$ for mean-based mixing and by $\tau^{-1}=\hat{\theta}\langle\tilde{\eta}\rangle=\hat{\theta}\langle\eta\rangle/2$ for the two-particle mixing—these equations represent the limits of Eqs. (23) and (24) as $\eta,\tilde{\eta}\rightarrow 0$. Note that the PDF representations of the mixing operator is somewhat different for mixing performed independently for different scalars and for the same simultaneous mixing of all scalars during a mixing event (the latter approach is commonly used in practical implementations) but the limit of $\eta,\tilde{\eta}\rightarrow 0$ is given by Eqs. (44) and (45) for both of these representations.

The divergent form of the mixing operator is closely linked to interpretation of this operator as conditional diffusion term in Lagrangian PDF modeling of turbulent flows. The evolutionary effect of turbulent diffusion is continuous and the conditional diffusion term has a divergent form. The nonuniqueness of the divergent interpretation of mixing operator in multidimensional spaces is linked to the fact that the mixing trajectories are discontinuous for finite η. The value before mixing $\mathbf{Z}^{\circ}$ changes to the value after mixing $\mathbf{Z}$ in a jump. In multidimensional spaces, the values $\mathbf{Z}^{\circ}$ and $\mathbf{Z}$ can be connected by different continuous trajectories corresponding to different continuous interpretations of mixing. This nonuniqueness is more philosophical than practical issue since different interpretations correspond to the same equation and the same solution for the PDF. As η becomes small the value $\mathbf{Z}$ approaches $\mathbf{Z}^{\circ}$ and the trajectory becomes close to continuous. This case corresponds to a unique divergent form of the mixing operator.

V. TWO-PARTICLE MIXING: MOMENTS IN THE SPATIALLY HOMOGENEOUS CASE

In this section, mixing between particles is not affected by particle positions in physical space; in fact, these particles can be considered as having only scalar property Z and there is no need to assign $\mathbf{x}_p$ to each particle. The particles with scalar properties and without physical coordinates are conventionally used in simulation of mixing in homogeneous turbulence. Each of the particles can have mixing interactions with all other particles (that is $n_a=n-1$ and a particle can, in principle, be mixed with any of the remaining particles) but, according to the two-particle mixing scheme, a selected particle can be mixed only with a single particle at a given moment.

Even if the initial values of Z_p are set independently for every particle, these values become dependent after mixing. Indeed, Eq. (17) indicates that variance and correlation between two selected particles numbered here by "1" and "2" are changed in a two-particle mixing event according to

$$\begin{bmatrix}\langle z_1^2\rangle\\ \langle z_1 z_2\rangle\end{bmatrix} = \begin{bmatrix}1-\gamma/2 & \gamma/2\\ \gamma/2 & 1-\gamma/2\end{bmatrix}\cdot\begin{bmatrix}\langle (z_1^{\circ})^2\rangle\\ \langle z_1^{\circ} z_2^{\circ}\rangle\end{bmatrix}. \tag{46}$$

Unless $\gamma=0$ (the trivial case of no mixing) or $\langle (z_1^{\circ})^2\rangle=0$ (the trivial case of absence of fluctuations), $\langle z_1 z_2\rangle$ becomes nonzero even when the correlation before mixing is zero $\langle z_1^{\circ} z_2^{\circ}\rangle$. This point of existence of correlations between particles after mixing can also be illustrated by the following consideration. If value of $\mu=\mu_f$ is fixed so that $0<\mu_f<1$, it is easy to see that mixing events become reversible: mixing with the new value of $\mu=\mu_r=\mu_f^{-1}$ would return the particles to their original states. Thus, after a sequence of mixing events, this sequence can be replayed back in reverse order with $\mu_r=\mu_f^{-1}$ and the initial state of all particles is then restored. It should be noted that the absolute values of $\tau_f^{-1}\sim 1-\mu_f^2$ and $\tau_r^{-1}\sim 1-\mu_r^2=1-\mu_f^{-2}$ are not consistent with each other [the dissipation time τ is given by Eq. (24)]. Indeed since the variance increases in the reverse process at the same rate as it decreases in forward process, one would expect that $\tau_r^{-1}=-\tau_f^{-1}$ where the negative sign of τ_r^{-1} indicates increase in the variance. However, Eq. (24) predicts a much faster rate of variance growth given by $\tau_r^{-1}\sim 1-\mu_f^{-2}$. The apparent paradox is explained by the fact that Eq. (24) is derived for the states without significant initial correlations between the particles while the reverse mixing process begins at the state with significant dependences between the particles created by the preceding forward mixing process (any small disturbance of this state results in rapid relaxation of τ from $\tau_r^{-1}=-\tau_f^{-1}$ to $\tau_r^{-1}\sim 1-\mu_f^{-2}$). This consideration indicates that the chaos hypothesis should be used with care in case of two-particle mixing.

The consideration of forward and reverse mixing effectively involves conditioning on a selected sequence of mixing events. However, correlations between particles may appear in two-particle mixing even if the final state is averaged over all possible mixing sequences. The homogeneous case has some advantages, which are exploited in this section: as demonstrated below, this case allows for exact closed formulation for the moment equations even when the chaos assumption is not valid and the equation for one-particle PDF is not closed.

In homogeneous case, operators L and $\tilde{L}$ retain only a single term—the time derivative and that $\theta_{pq}=\theta_0$ does not depend on $\mathbf{x}_p-\mathbf{x}_q$. Since two-particle mixing strictly preserves the first moment $\langle Z\rangle$, we can put $\langle Z\rangle=0$ and replace Z by $z=Z-\langle Z\rangle$. This makes our consideration more transparent and does not involve any loss of generality. The equations for the second moments, which can be obtained from PDF Eq. (38) at $k=2$, become

$$\frac{d\langle z_1^2\rangle}{dt} = \theta_0(n-1)\langle (z_1(1-\tilde{\eta})+\tilde{\eta}z_2)^2 - z_1^2\rangle, \tag{47}$$

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$$\frac{d\langle z_1 z_2\rangle}{dt} = \theta_0\langle (z_1(1-\tilde{\eta}) + \tilde{\eta} z_2)(z_2(1-\tilde{\eta}) + \tilde{\eta} z_1) - z_1 z_2\rangle + 2\theta_0(n-2)\langle (z_1(1-\tilde{\eta}) + \tilde{\eta} z_3)z_2 - z_1 z_2\rangle. \tag{48}$$

At this point we use the fact that all particles are stochastically equivalent and $\langle z_1^2\rangle = \langle z_2^2\rangle$ and $\langle z_1 z_2\rangle = \langle z_1 z_3\rangle = \langle z_3 z_2\rangle$. With the use of these relations, the equations for the second moments can now be written in the form

$$\frac{d\langle z_1^2\rangle}{dt} = -2\theta(n-1)\langle\tilde{\gamma}\rangle(\langle z_1^2\rangle - \langle z_1 z_2\rangle), \tag{49}$$

$$\frac{d\langle z_1 z_2\rangle}{dt} = 2\theta\langle\tilde{\gamma}\rangle(\langle z_1^2\rangle - \langle z_1 z_2\rangle), \tag{50}$$

where

$$\tilde{\gamma} \equiv \tilde{\eta} - \tilde{\eta}^2 = \gamma/4.$$

Equations for the moments are derived from the BBGKY chain of PDF equations in Eq. (38). Nonzero correlations between particles $\langle z_1 z_2\rangle$ are generated by the first term on rhs of Eq. (38) at $k=2$ which is $n-k$ times smaller than the following term. The first term in Eq. (38) at $k=2$ is closed but the correlations that are generated by this term make the following term not closed since the following term involves P_3. However, the system of the second moment Eqs. (49) and (50) appears to be closed in this case due to equivalence of the correlations noted above. For example, if $P_2(z_1,z_2)$ is known while $P_2(z_1,z_2,z_3)$ is not, then $\langle z_1 z_3\rangle$ is known since $\langle z_1 z_3\rangle$ is the same as $\langle z_1 z_2\rangle$ which is determined by $P_2(z_1,z_2)$. In general, for each m, all equations for the moments of order that does not exceed m form a closed system. Derivations of these equations require m equations $k=1,\ldots,m$ from the BBGKY chain (38).

With the use of new variables G and K, The second moment equations can now be rewritten in the form

$$\frac{dG}{dt} = 0, \quad G = \langle z_1^2\rangle + (n-1)\langle z_1 z_2\rangle, \tag{51}$$

$$\frac{dK}{dt} = -2\theta n\langle\tilde{\gamma}\rangle K, \quad K = \langle z_1^2\rangle - \langle z_1 z_2\rangle, \tag{52}$$

where mixing does not change G but reduces K. Assuming that $\langle z_1^2\rangle = \langle z_1^2\rangle_0$ and $\langle z_1 z_2\rangle = 0$ at the initial moment of $t=0$, we obtain that $G = \langle z_1^2\rangle_0$. The physical meaning of G is now considered. Let $S_n = z_1 + \cdots + z_n$ be the sum of all-particle values. It is obvious that $\langle S_n\rangle = 0$ since $\langle z_p\rangle = 0$ but $\langle S_n^2\rangle = n\langle z_1^2\rangle + n(n-1)\langle z_1 z_2\rangle = nG = n\langle z_1^2\rangle_0$ is nonzero. Values z_p are generated independently at the initial moment and $\langle S_n^2\rangle$ must be greater than zero. Since the value of S_n is preserved by mixing, which is strictly conservative, the variance $\langle S_n^2\rangle$ does not change with time while the correlation $\langle z_1 z_2\rangle$ increases during the mixing process. Mixing is effectively terminated when $K \to 0$ and $\langle z_1 z_2\rangle \to \langle z_1^2\rangle$. Finally, we take into account that $G = \langle z_1^2\rangle_0$ and obtain $\langle z_1 z_2\rangle = \langle z_1^2\rangle = \langle z_1^2\rangle_0 / n$ at the limit of $t = \infty$.

We can now conclude that $\langle z_1 z_2\rangle$ becomes positive during mixing process and this indicates violation of the chaotic conditions. However, if the number of particles n is reasonably large (and this is the case in practical applications), then the correlation $\langle z_1 z_2\rangle$ is small $(\sim 1/n)$ and can be neglected in the homogeneous case. Dependences between particles are spread over all particles and appear to be small when n is large. Hence, in two-particle mixing the chaos assumptions are not exact but represent a good approximation provided a sufficiently large number of particles present in the simulations. We need to note that smallness of the interparticle correlations is determined by the smallness of the first term compared to the second term on rhs of the BBGKY chain (38). Higher moments require considerations of equations with higher k in this chain and this results in a reduced relative value of the second term on rhs of Eq. (38). Hence statistical dependence is more pronounced for higher moments than for lower moments.

VI. LOCALIZED TWO-PARTICLE MIXING: DEVIATIONS FROM CHAOTIC CONDITIONS

In this section, we investigate deviations from fully chaotic conditions for the case of two-particle mixing which is localized in physical space. Localization in physical space means that particles are engaged in mixing interactions only if they are sufficiently close to each other in physical space. The intensity of mixing can be expressed for this case by Eq. (36)—the intensity of mixing between particle p and another particle is positive only if the other particle is located within V_p—the volume of mixing interactions for particle p. In the previous section, we determined that the effects of dependence between particles do exist in two-particle mixing but appear to be small when these effects are spread over a large number of particles. Nevertheless, when mixing is local, these effects can be locally amplified with dramatic consequences for the intensity of mixing even if the overall number of particles n is very large. This section introduces analysis of nonchaotic behavior on localized mixing.

The radius r_m of the spherical volume V_m (or V_p when this volume is associated with particle p) is presumed to be small compared to r_0—the characteristic scale of the problem under consideration. In this section, for the sake of simplicity, we neglect the influence of the source terms (i.e., we put $W=0$) since in consideration of effects associated with small scales, the diffusion terms are dominant over the source terms. Strict asymptotic expressions can be obtained for two-particle mixing for two limiting cases of $n_a \ll 1$ and $n_a \gg 1$ while a general case is subject to approximate treatment that is compliant with the asymptotic expressions and with diffusional interpretation of interactions with third molecules. Here, n_a denotes the average number of particles in the mixing volume V_p not counting the particle p (i.e., in the terms used here the average number of interacting particles); n_a is defined after Eq. (36).

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A. Mixing rate

The equation for the scalar variance $\langle Z_1^2\rangle_1$

$$\widetilde{L}_1[\langle Z_1^2\rangle_1] = (n-1)\langle \theta_{12}((Z_1(1-\widetilde{\eta}) + \widetilde{\eta}Z_2)^2 - Z_1^2)\rangle_1$$
$$= (n-1)\,\theta_0 2\langle\widetilde{\gamma}\rangle\int_{V_1}(((\langle Z_1Z_2\rangle_{12} - \langle Z_1^2\rangle_{12}))P_x(\mathbf{x}_2)d\mathbf{x}_2, \tag{53}$$

which can be obtained from Eq. (37) is not closed since this equation involves correlations $\langle Z_1Z_2\rangle_{12}$. Here, subscript indices are used to indicate conditions for averaging. For example, $\langle Z_1^2\rangle_1$ is the average of Z_1^2 conditioned on a given location $\mathbf{x}_1$ of particle 1 while $\langle Z_1^2\rangle_{12}$ is the average of the same value, Z_1^2, conditioned on positions $\mathbf{x}_1$ and $\mathbf{x}_2$ of particles 1 and 2. Thus, $\langle Z_1^2\rangle_1$ depends on $\mathbf{x}_1$, while $\langle Z_1Z_2\rangle_{12}$ and $\langle Z_1^2\rangle_{12}$ depend on both $\mathbf{x}_1$ and $\mathbf{x}_2$.

Equations for $\langle Z_1^2\rangle_{12}$ and $\langle Z_1Z_2\rangle_{12}$ are obtained form (38) with $k=2$:

$$\widetilde{L}_2[\langle Z_1^2\rangle_{12}] = \theta_{12}\langle (Z_1(1-\widetilde{\eta}) + \widetilde{\eta}Z_2)^2 - Z_1^2\rangle_{12}$$
$$+ (n-2)\langle \theta_{13}((Z_1(1-\widetilde{\eta}) + \widetilde{\eta}Z_3)^2 - Z_1^2)\rangle_{12}, \tag{54}$$

$$\widetilde{L}_2[\langle Z_1Z_2\rangle_{12}]$$
$$= \theta_{12}(\langle (Z_1(1-\widetilde{\eta}) + \widetilde{\eta}Z_2)(Z_2(1-\widetilde{\eta}) + \widetilde{\eta}Z_1) - Z_1Z_2\rangle_{12})$$
$$+ (n-2)\langle \theta_{13}((Z_1(1-\widetilde{\eta}) + \widetilde{\eta}Z_3)Z_2 - Z_1Z_2)\rangle_{12}$$
$$+ (n-2)\langle \theta_{23}((Z_2(1-\widetilde{\eta}) + \widetilde{\eta}Z_3)Z_1 - Z_1Z_2)\rangle_{12}. \tag{55}$$

The rhs of these equations involves two terms. The first term is closed but this term represents direct interactions of particles 1 and 2 and this term is responsible for deviations from fully chaotic conditions. The second term represents interactions with other particles (other than 1 and 2). This is closed only under chaotic conditions since, unlike in the homogeneous case, $\langle Z_1Z_2\rangle_{12}$ and $\langle Z_1Z_3\rangle_{12}$ are not the same due to spatial conditioning. The overall influence of the second term averaged over all values of $\mathbf{x}_2$ is always dominant over the first term since the second term is multiplied by a large number, $n-2\sim n$. The first term, however, can be the largest in the mixing interaction volume V_1. Indeed, in the mixing interaction volume, the first term is proportional to θ_0 while the second term is proportional to $\theta_V(n-2)\approx\theta_V(n-1)=\theta_0 n_a$. Hence if $n_a \gg 1$, the second term is always dominant and chaotic conditions are not violated at the leading order of the analysis. If $n_a \ll 1$, the first term asymptotically determines the structure of the solution near the mixing volume and we can write the following leading order expressions:

$$\widetilde{L}_2[\langle Z_1^2\rangle_{12}] = -2\theta_{12}\langle\widetilde{\gamma}\rangle(\langle Z_1^2\rangle_{12} - \langle Z_1Z_2\rangle_{12}), \tag{56}$$

$$\widetilde{L}_2[\langle Z_1Z_2\rangle_{12}] = 2\theta_{12}\langle\widetilde{\gamma}\rangle(\langle Z_1^2\rangle_{12} - \langle Z_1Z_2\rangle_{12}). \tag{57}$$

With the use of the variables G and K, these equations can be transformed into

$$\widetilde{L}_2[G] = 0, \quad G = \langle Z_1^2\rangle_{12} + \langle Z_1Z_2\rangle_{12}, \tag{58}$$

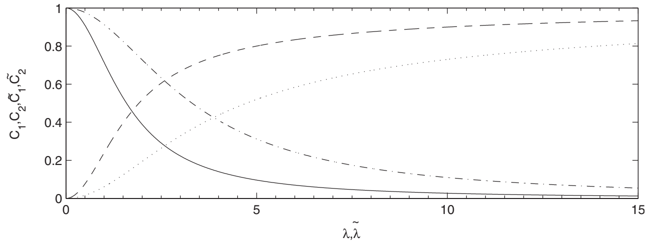

FIG. 1. Effects of dependences between particles on mixing in three-dimensional case $d=3$; (—-) $C_1(\lambda)$, (—) $C_2(\lambda)$, (-·-·-·-) $\widetilde{C}_1(\widetilde{\lambda})$, and (·····) $\widetilde{C}_2(\widetilde{\lambda})$.

$$\widetilde{L}_2[K] = -4\theta_{12}\langle\widetilde{\gamma}\rangle K, \quad K = \langle Z_1^2\rangle_{12} - \langle Z_1Z_2\rangle_{12}. \tag{59}$$

If mixing is localized and r_m is much smaller than the characteristic scale of the problem r_0, diffusion appears to be the dominant term in the operator $\widetilde{L}_2$. The solution is sought in form of

$$K = \langle z^2\rangle\Phi\left(\frac{r}{r_m},\lambda,d\right), \quad y^i = x_2^i - x_1^i, \quad r^2 = y^iy^i, \tag{60}$$

$$\lambda^2 \equiv \frac{2\theta_0\langle\widetilde{\gamma}\rangle}{D}r_m^2 \tag{61}$$

and given in the Appendix. This solution indicates that the mixing volume is surrounded by a variance depression where the particles have strong correlation

$$\langle z_1^2\rangle_{V_1} = \frac{1+C_2}{2}\langle z_1^2\rangle_1, \quad \langle z_1z_2\rangle_{V_1} = \frac{1-C_2}{2}\langle z_1^2\rangle_1. \tag{62}$$

The main global effect of this depression is attenuation of the mixing rate which can be determined by substituting the solutions for $K=\langle Z_1^2\rangle_{12} - \langle Z_1Z_2\rangle_{12}$ into Eq. (53) yielding the following equations:

$$\widetilde{L}_1[\langle Z_1^2\rangle_1] = -2(n-1)\langle\widetilde{\gamma}\rangle\theta_V C_2\langle z_1^2\rangle_1, \tag{63}$$

$$\tau = \frac{1}{n_a\langle\widetilde{\gamma}\rangle\theta_0 C_2} = \frac{\tau_0}{C_2} = \frac{\tau_D}{n_a C_1}, \tag{64}$$

$$\tau_0 = \frac{4}{\langle\gamma\rangle\hat{\theta}} = \frac{1}{n_a\langle\widetilde{\gamma}\rangle\theta_0}, \quad \tau_D \equiv \frac{r_m^2}{Dd(d-2)}, \tag{65}$$

$$C_2(\lambda,d) = d(d-2)\frac{C_1}{\lambda^2}, \tag{66}$$

$$C_1(\lambda,d) = \left(1 + \frac{I_{d/2-1}(\lambda)}{\lambda I_{d/2}(\lambda)}(d-2)\right)^{-1}. \tag{67}$$

The parameter λ determines the degree of deviation from chaotic conditions—these deviations are large when λ is large and small when λ is small. The function $C_2(\lambda,d)$ determines the increase in effective mixing time τ and are shown in Fig. 1 jointly with $C_1(\lambda,d)$ for $d=3$. It should be noted that this increase is not related to any reductions in the frequency of mixing—the programmer may think that it is the old value of the mixing time denoted here by τ_0 that

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determines the mixing rate. In fact, mixing can be much slower when λ becomes large. At the limit of very large λ, the mixing rate does not depend on θ_0 but is linked to the diffusion time τ_D which determines the diffusion flux of variance toward the mixing volume V_1. The effect of correlation between particles, which may seem to be small when examined on large scales, can play a significant or even a dominant role in a mixing locality.

The reduction in intensity of mixing due to correlations between particles can be interpreted as effective "premixing" between particles prior to a mixing event. The effective extent of mixing for this premixing is $\gamma_{\text{prem}}=1-C_2$ so that the reduction of variance and generation of correlations specified by Eq. (46) with $\langle \overset{\circ}{z}_1 \overset{\circ}{z}_2\rangle=0$ is the same as given by Eq. (62). This imaginary premixing is followed by the actual mixing event whose impact is reduced due to correlations between z_1 and z_2. The resulting effective extent of mixing is given by $\gamma_{\text{eff}}=(1-C_2)+C_2\gamma$. Here we use that the overall extent of mixing of two sequential mixing events with γ_{prem} and γ is given by $\gamma_{\text{eff}}=\gamma_{\text{prem}}+(1-\gamma_{\text{prem}})\gamma$ which corresponds to $\mu_{\text{eff}}=\mu_{\text{prem}}\mu$. From a practical point of view, evaluation of correlations between particles may be complicated and stochastically inaccurate when the number of particles is small. It seems that the simplest indicator of the dependences between particles could be repeated mixing between the same particles. The fraction of couples experiencing multiple mixing events should be small under proper chaotic conditions.[11] We note, however, that two-particle mixing is complicated and it is possible to have mixing between different couples of particles selected from a fixed group of three or four resulting in stochastic dependences appearing within this group.

We note that steady localized solutions of the problem can be obtained only for dimensions of $d\geq 3$. In the one-dimensional case, the zones of variance depression will gradually expand creating growing areas of particles with correlated values. The details of this solution are problem dependent. In the two-dimensional case, the solution is reasonably close to localized steady solutions observed in higher dimensions. However, the variance depression has logarithmic dependence on r which indicates that this depression tends to slowly propagate outward and this propagation terminates only when the outer scale of the problem, r_0, is reached. This is used in the appendix to obtain an approximate solution for the two-dimensional case.

B. Numerical diffusion

It can be expected that existence of correlations can have an attenuating effect not only on the rate of mixing but also on numerical diffusion. Indeed, due to correlations between Z_2 and Z_1, the value $\langle Z_2\rangle_{12}-\langle Z_1\rangle_{12}$ in the integral

$$\begin{aligned}\tilde{L}_1[\langle Z_1\rangle_1] &= (n-1)\langle \theta_{12}(Z_1+\tilde{\eta}(Z_2-Z_1)-Z_1)\rangle_1\\ &= (n-1)\theta_0\langle\tilde{\eta}\rangle\int_{V_1}(\langle Z_2\rangle_{12}-\langle Z_1\rangle_{12})P_x(\mathbf{x}_2)d\mathbf{x}_2 \end{aligned} \tag{68}$$

would appear to be smaller than under fully chaotic conditions. Equation (68) is obtained by evaluating first moment of Eq. (37). This equation is not closed and involves first moments $\langle Z_2\rangle_{12}$ and $\langle Z_1\rangle_{12}$ which are conditioned on locations of both particles. The equations for this doubly conditioned moments can be obtained from Eq. (38) with $k=2$ so that we obtain for $\langle Z_1\rangle_{12}$

$$\begin{aligned}\tilde{L}_2[\langle Z_1\rangle_{12}] &= \theta_{12}\langle Z_1+\tilde{\eta}(Z_2-Z_1)-Z_1\rangle_{12}\\ &\quad +(n-2)\langle \theta_{13}(Z_1+\tilde{\eta}(Z_3-Z_1)-Z_1)\rangle_{12}. \end{aligned} \tag{69}$$

This equation is also not closed due to the presence of $\langle Z_3\rangle_{12}$ terms on rhs. However, as in Sec. VI A, two limiting cases of $n_a \gg 1$ and $n_a \ll 1$ allow for asymptotic solutions. In the case of $n_a\gg 1$, the second term on rhs of Eq. (69) is dominant and this corresponds to chaotic conditions since deviations from chaos induced by the first term are small uniformly in the domain. In the opposite case of $n_a\ll 1$, the first term on rhs of Eq. (69) appears to be dominant in the mixing volume. The leading order equations in the vicinity of the mixing volume take the form

$$\tilde{L}_2[\langle Z_1\rangle_{12}]=\theta_{12}\langle\tilde{\eta}\rangle(\langle Z_2\rangle_{12}-\langle Z_1\rangle_{12}), \tag{70}$$

$$\tilde{L}_2[\langle Z_2\rangle_{12}]=\theta_{12}\langle\tilde{\eta}\rangle(\langle Z_1\rangle_{12}-\langle Z_2\rangle_{12}). \tag{71}$$

Here, we use the fact that the equations for $\langle Z_1\rangle_{12}$ and $\langle Z_2\rangle_{12}$ must be equivalent due to stochastic equivalence of all particles. With introductions of new variables $\tilde{G}$ and $\tilde{K}$, this system of equations takes the form

$$\tilde{L}_2[\tilde{G}]=0, \quad \tilde{G}=\langle Z_1\rangle_{12}+\langle Z_2\rangle_{12}, \tag{72}$$

$$\tilde{L}_2[\tilde{K}]=-2\theta_{12}\langle\tilde{\eta}\rangle\tilde{K}, \quad \tilde{K}=\langle Z_2\rangle_{12}-\langle Z_1\rangle_{12}. \tag{73}$$

The function $\tilde{K}(\mathbf{x}_1,\mathbf{x}_2)$ is obviously an antisymmetric function of its arguments. This and other properties are used in the appendix to seek the solution in the following form:

$$\tilde{K}=\left(\left(\frac{\partial\langle Z\rangle}{\partial x^i}\right)_1+\frac{1}{2}\left(\frac{\partial^2\langle Z\rangle}{\partial x^i\,\partial x^j}\right)_1 y^j\right)\frac{y^i}{r}\tilde{\Phi}_r\left(\frac{r}{r_m},\tilde{\lambda},d\right), \tag{74}$$

where a new parameter $\tilde{\lambda}$ is introduced by

$$\tilde{\lambda}^2\equiv\frac{\theta_0\langle\tilde{\eta}\rangle}{D}r_m^2=\frac{\lambda^2}{\beta}, \quad \beta=\frac{2\langle\tilde{\gamma}\rangle}{\langle\tilde{\eta}\rangle}=\frac{\langle\gamma\rangle}{\langle\eta\rangle}. \tag{75}$$

The asymptotic solution found in Appendix determines that the value of numerical diffusion is effectively reduced when $\tilde{\lambda}$ is sufficiently large according to the following equations:

$$D_m=\frac{n_a\theta_0\langle\tilde{\eta}\rangle\hat{r}_1^2}{2}\tilde{C}_2=D_{m0}\tilde{C}_2=\frac{\hat{r}_1^2}{\beta\tau}\frac{\tilde{C}_2(\tilde{\lambda})}{C_2(\lambda)}=D\frac{n_a}{2d}\tilde{C}_1. \tag{76}$$

Here, D_{m0} represents the old value of the numerical diffusion coefficient obtained under assumptions of particle chaos:

$$D_{m0}\equiv\frac{\hat{r}_1^2 n_a\langle\tilde{\eta}\rangle\theta_0}{2}=\frac{\hat{r}_1^2}{\tau_0\beta}. \tag{77}$$

The solution exists for all dimensions $d\geq 1$ and the functions $\tilde{C}_2(\tilde{\lambda},d)$ and $\tilde{C}_1(\tilde{\lambda},d)$ given by

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$$\tilde{C}_2(\tilde{\lambda},d) = d(d+2)\frac{\tilde{C}_1(\tilde{\lambda},d)}{(\tilde{\lambda})^2} = C_2(\tilde{\lambda},d+2), \tag{78}$$

$$\tilde{C}_1(\tilde{\lambda},d) = C_1(\tilde{\lambda},d+2). \tag{79}$$

These functions are shown in Fig. 1 for $d=3$. As $\tilde{\lambda}$ increases, the numerical diffusion coefficient D_m becomes smaller than its evaluations D_{m0} based on the assumptions of particle chaos. Note that in one-dimensional case ($d=1$), particle trajectories do intersect and it is possible (at least in principle) to introduce mixing without numerical diffusion associated with positive distances between mixed particles.

C. Estimations for other regimes

As discussed in Secs. VI A and VI B, asymptotic solutions can be obtained in two limiting cases $n_a \gg 1$ and $n_a \ll 1$ since in this case one of the terms on rhs of Eq. (69) is dominant over the other term. The practical use of Lagrangian particles with mixing requires consideration of a general case of $n_a \sim 1$. This case is most difficult due to complex interactions between the two terms on rhs of Eq. (69). This problem, of course, can be treated by considering a large number of higher moments but this approach seems to use extensive calculations at the expense of the physics of the problem. The alternative way of treating this problem is the diffusional interpretation of mixing interactions with a randomly selected particle. In the same way as we obtain the diffusional term for approximating rhs of Eq. (68) we can interpret influence of mixing of particles 1 and 2 with randomly selected particle 3 on the first moment as diffusion which is, to a certain extent, equivalent to random alterations of the particle positions. The qualitative physical interpretation of this effect is given in Ref. 11. With this interpretation, we can write Eq. (69) in the form

$$\tilde{L}_2[\langle Z_1\rangle_{12}] = \theta_{12}\langle Z_1 + \tilde{\eta}(Z_2 - Z_1) - Z_1\rangle_{12} + D_m\left(\frac{\partial\langle Z_1\rangle_{12}}{\partial x_1^i\,\partial x_1^i} + \frac{\partial\langle Z_1\rangle_{12}}{\partial x_2^i\,\partial x_2^i}\right) \tag{80}$$

that approximates the last term in Eq. (69) by numerical diffusion. We have to stress the approximate nature of this equation. The difference between Eqs. (68) and (69) in multipliers ($n-1$ in one case and $n-2$ in the other) is insignificant since n is very large. In spite of the high degree of similarity between last terms in Eqs. (68) and (69), there is a difference between these terms since one of the terms is single conditioned while the second term is double conditioned (on presence of two particles 1 and 2). Presence of particles other than 1 and 2 in the mixing volume results in enhancement of diffusion due to likely exchanges between all of these particles. The previously developed theory can now be generalized for arbitrary values of n_a by replacing D by D_{eff},

$$\tilde{\lambda}^2 = \frac{\theta_0\langle\tilde{\eta}\rangle}{D_{\text{eff}}} r_m^2, \quad D_{\text{eff}} = D + D_m. \tag{81}$$

The old value of this parameter is denoted by

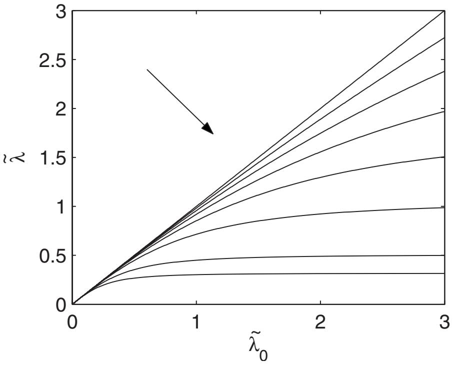

FIG. 2. Dependence of the effective value of $\tilde{\lambda}$ on the parameter $\tilde{\lambda}_0$ shown for $d=3$ and $n_a=[0, 0.4, 1, 2, 4, 10, 40, 100]$. The arrow shows the direction of n_a increase.

$$\tilde{\lambda}_0^2 = \frac{\theta_0\langle\tilde{\eta}\rangle}{D} r_m^2 = \frac{2r_m^2}{\beta D \tau_0 n_a}, \tag{82}$$

while the ratio D_m/D_{eff} [rather than the ratio D_m/D as in Eq. (76)] is determined by $\tilde{C}_1$:

$$\frac{D_m}{D_{\text{eff}}} = d\frac{n_a}{2}\tilde{C}_1(\tilde{\lambda},d), \tag{83}$$

where

$$\tau = \frac{\tau_0}{C_2(\lambda,d)}, \quad \tau_0 = \frac{1}{n_a\langle\tilde{\gamma}\rangle\theta_0}, \quad \frac{\lambda_0^2}{\tilde{\lambda}_0^2} = \frac{\lambda^2}{\tilde{\lambda}^2} = \beta. \tag{84}$$

Using Eqs. (81) and (82), we consider the ratio $\tilde{\lambda}^2/\tilde{\lambda}_0^2$ and obtain after simple transformations

$$\tilde{\lambda}^{-2} = \tilde{\lambda}_0^{-2}\frac{D_{\text{eff}}}{D} = \tilde{\lambda}_0^{-2}\left(1 + \frac{D_m}{D}\right). \tag{85}$$

The ratio D_m/D can be found by multiplying Eq. (83) by $D_{\text{eff}}/D = \tilde{\lambda}_0^2/\tilde{\lambda}^2$ and taking into account that $\tilde{C}_2/(d+2) = \tilde{C}_1 d/(\tilde{\lambda})^2$ according to Eq. (78)

$$\frac{D_m}{D} = \frac{n_a}{2(d+2)}\tilde{\lambda}_0^2\tilde{C}_2(\tilde{\lambda},d). \tag{86}$$

Substitution of this expression for D_m/D into Eq. (85) results in

$$\tilde{\lambda}_0^2 = \frac{\tilde{\lambda}^2}{1 - d\frac{n_a}{2}\tilde{C}_1(\tilde{\lambda})}. \tag{87}$$

This equation relates the effective value of $\tilde{\lambda}$ to the value of $\tilde{\lambda}_0$ which is known and defined by Eq. (82). Dependence of $\tilde{\lambda}$ on $\tilde{\lambda}_0$ for $d=3$ and different values of n_a is shown in Fig. 2. As expected for the limiting case of one-particle interactions $n_a \ll 1$, we obtain $\tilde{\lambda}_0^2 = \tilde{\lambda}^2$ while for mean-based interactions $n_a \gg 1$, Eq. (87) determines that $\tilde{C}_1(\tilde{\lambda}) = 2d/n_a$, $\tilde{\lambda}^2 = 2(d+2)/n_a \to 0$ and assumptions of particle chaos are valid. Here we use that $\tilde{C}_1(\tilde{\lambda}) \to d(d+2)\tilde{\lambda}^2$ as $\tilde{\lambda} \to 0$. Considering that $\tilde{C}_2(0) = 1$, this asymptotics can be easily obtained from Eq. (78). Equation (87) can be used to estimate deviations from

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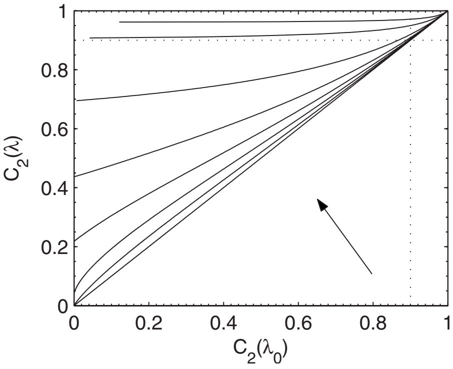

FIG. 3. Dependence of $C_2(\lambda)$ and as function of $C_2(\lambda_0)$ for $d=3$, $\beta=1$, and $n_a=[0,0.4,1,2,4,10,40,100]$ (solid lines). The dotted lines mark conditions $C_2(\lambda)=0.9$ and $C_2(\lambda_0)=0.9$ discussed in the text. The arrow shows the direction of n_a increase.

fully chaotic conditions for any value of n_a. Figure 3 shows dependence of $C_2(\lambda)$ on $C_2(\lambda_0)$ for $\beta=1$. Values of C_2 approaching 0 indicate significant deviations from chaotic conditions and substantially decreased rate of mixing. One can see that probable presence of several particles in a mixing volume reduces dependences between particles.

The effect of dependences between particles is controlled by parameter λ. This results in increase in the mixing time according to $\tau=\tau_0/C_2(\lambda)\geq\tau_0$ where τ_0 is the mixing time that corresponds to fully chaotic conditions. The parameters λ and $\tilde{\lambda}=\lambda\beta^{-1/2}$ depend on the parameter $\tilde{\lambda}_0$ as discussed above. In the case of two-particle interactions ($n_a \ll 1$) parameters $\tilde{\lambda}$ and $\tilde{\lambda}_0$ coincide. Multiparticle interactions ($n_a>1$) result in decrease in $\tilde{\lambda}$ compared to $\tilde{\lambda}_0$ and this corresponds to weaker dependences between particles.

VII. IMPLICATIONS FOR NUMERICAL METHODS

The requirements of minimizing stochastic variations $\langle z^2\rangle$, minimizing bias induced by numerical diffusion and preserving independence of particles are competing requirements. Existence of competing requirements for minimizing errors is common in stochastic calculations.[11,19,22] The variance $\langle z^2\rangle$ can be reduced by increasing intensity of mixing and reducing τ but this amplifies the numerical diffusion since $D_m \sim r_m^2/\tau$. Decreasing τ also increases $\tilde{\lambda}$ [see Eq. (82)] so that dependences between particles become more significant. The numerical diffusion can be reduced by localizing mixing and reducing r_m. If $d>2$, then n_a decreases faster than r_m^2 so that $\tilde{\lambda}$ increases. Under chaotic conditions when values Z_p assigned to different particles are stochastically independent, a single particle is sufficient to characterize all other particles. Deviations from chaotic conditions are always small on a large scale provided the number of particles is sufficiently large. This, however, does not ensure that the particle independence is preserved locally, in the vicinity of mixing region. The effect of particle dependence is localized for high dimensions $d\geq 3$ resulting in reduction in the mixing rate and attenuation of numerical diffusion. The local effect of particle dependence is small provided diffusion coefficient D is sufficiently large or provided a sufficiently large number of particles n_a interact with a given particle. In one-dimensional case, the effects particle dependence does not stay localized and expands slowly to surrounding domain. We now examine the issues of reducing numerical errors in greater detail.

A. Convergence with localization and convergence with intensification

Among various possibilities for selecting mixing parameters under conditions of increasing number of particles n, it is worthwhile to distinguish two cases:

- convergence with intensification $n\to\infty$, $r_m \sim \text{const}$, $\tau \sim \text{const}$, $D_m \sim r_m^2/\tau \sim \text{const}$, and $n_a \to \infty$
- convergence with localization: $n\to\infty$, $r_m\to 0$, $\tau\to 0$, $D_m \sim r_m^2/\tau \to 0$, and $n_a \sim \text{const}$.

The first limit corresponds to fixed localization and fixed dissipation rate. The increasing number of particles results only in progressive enforcement of chaotic conditions while scattering of Z around its mean $\langle Z\rangle$ remains. Once particles become effectively independent, any further increase in n does not change anything. Although the whole set of particles can be characterized by stochastic properties of a single selected particle, having more independent particles in the flow allows us to evaluate averages (stochastic errors associated averaging over particles are assessed in Ref. 19). The exact solution of the scalar transport equations is not achieved in this case due to persisting scattering and smoothing effect of the numerical diffusion. In context of large eddy simulations (LESs) of scalar transport in turbulent flows, we can refer to this convergence as convergence to a model. In this case, the scale r_m is linked to the LES filter (grid) scale and large $n_a\to\infty$ allows for accurate evaluation of the averages within cells. According to the long-standing tradition of PDF and FDF (filtered density function) simulations,[7,17,18] the valiance $\langle z^2\rangle$ is treated in this framework as a model for subgrid scalar variations. The issues related to modeling of turbulent reacting flows are specifically considered in Paper II of this work.[16] Note that it is possible to use a combination of the limits so that $n\to\infty$, $r_m\to 0$, $\tau\sim\text{const}$, $D_m\sim r_m^2/\tau \to 0$, and $n_a\to\infty$. In this case, slow reduction in r_m is compensated by increase of n so that n_a also increases enforcing particle independence. Due to vanishing smoothing effect of numerical diffusion, this limit requires an explicit LES filtering procedure to suppress subgrid fluctuations of the simulated scalar fields.

In the second limit the increasing number of particles is used to localize $r_m\to 0$ and intensify $\tau\to 0$ mixing. Intensification of mixing tightens the stochastic scattering $\langle z^2\rangle \sim \chi\tau$ [see Eq. (20)], although parameters must be selected in a way that ensures attenuation of numerical diffusion. We also note that $\tilde{\lambda}_0^2 \sim D_m/(Dn_a)\to 0$ and chaotic conditions are also enforced at this limit. Thus, the value of Z converges to the exact solution of the scalar transport equation since the stochastic scattering and the bias due to numerical diffusion disappear at this limit. We can treat these particles with scalar properties and mixing as alternative stochastic process representing the diffusion equation. In case of a turbulent

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flow this limit can be referred to as convergence to direct numerical simulations fully resolving all details of the scalar field. The problem of best selection of mixing parameters which minimizes the associated numerical errors is considered below.

B. Minimizing numerical errors for two-particle mixing

In the rest of this paper, we consider the three-dimensional case of $d=3$. We also note that, as shown in Ref. 11, the parameter β is restricted by $1 \le \beta \le 2$ with $\beta=1$ corresponding to conventional Curl's model with $\eta=1$. In order to provide numerically accurate simulation we wish to ensure minimal scattering of $\langle z^2\rangle$ under condition that numerical diffusion D_m and dependence between different particles remain small:

$$\frac{D_m}{D} \le \varepsilon_D \quad \text{and} \quad \frac{\langle z_1 z_2\rangle_{V_1}}{\langle z_1^2\rangle_1} \le \varepsilon_Z, \tag{88}$$

where ε_D and ε_Z are some small values representing accuracy that we deem satisfactory. The second of these conditions implies that $1-C_2 \le 2\varepsilon_Z$ in Eq. (62). Expansion of Eq. (A9) into Taylor series $C_2(\lambda,3)=1-\frac{2}{5}\lambda^2+\cdots$, we obtain the following restriction $\lambda^2 \le 5\varepsilon_Z$ or $\tilde{\lambda}^2 \le 5\varepsilon_Z/\beta$. This restriction can be satisfied by $\lambda_0^2 \le 5\varepsilon_Z$ and $\tilde{\lambda}_0^2 \le 5\varepsilon_Z/\beta$ or, if we take into account asymptotics $\tilde{\lambda}^2 \to 10/n_a$ as $n_a \to \infty$, which is derived in the paragraph after Eq. (87), by $n_a \ge 2\beta/\varepsilon_Z$. This point is illustrated in Fig. 3, where $C_2(\lambda)$ is plotted versus $C_2(\lambda_0)$ for $\beta=1$ (in this case we do not need to distinguish λ from $\tilde{\lambda}$ and λ_0 from $\tilde{\lambda}_0$). We take $\varepsilon_Z=0.5$ which corresponds to $C_2(\lambda) \ge 0.9$ (and to $\lambda^2 \lesssim 0.5$). This condition, which is shown by the horizontal dotted line in Fig. 3, can be satisfied by $C_2(\lambda_0) \ge 0.9$ (and by $\lambda_0^2 \lesssim 0.5$), which is shown in Fig. 3 by the vertical dotted line, or by the condition $n_a \ge 2/\varepsilon_Z = 40$ (this condition is represented by the area above the second curve from the top in Fig. 3).

Assuming that ε_Z is sufficiently small so that $\tau \approx \tau_0$ and $D_m \approx D_{m0}$ at the leading order, we can write Eq. (82) as $\tilde{\lambda}_0^2 = 10 D_m/(D n_a \beta)$. Hence, $2n_a^{-1} D_m/D \le \varepsilon_Z$ ensures that $\tilde{\lambda}_0^2 \le 5\varepsilon_Z/\beta$. The restrictions imposed on the parameters are now given by

$$\frac{D_m}{D} \le \varepsilon_D \tag{89}$$

and

$$\text{(a)} \quad n_a \ge \frac{2\beta}{\varepsilon_Z} \quad \text{or} \quad \text{(b)} \quad n_a \ge \frac{2}{\varepsilon_Z}\frac{D_m}{D}. \tag{90}$$

Equation (89) ensures that numerical diffusion is small compared to physical diffusion while either of the conditions specified in Eq. (90) is sufficient to guarantee the required level of particle independence: condition (90) by offering choice of many partners for mixing and condition (90) by stirring particles by random walk. If $D_m/D \le \beta$ then satisfying condition (90) is easier than satisfying condition (90). One can see that $n_a \ge 2\varepsilon_D/\varepsilon_Z$ ensures particle independence in this case. Note that if we impose very tight restrictions on numerical diffusion but not on particle independence then relatively small n_a can be sufficient to satisfy these requirements. The conditions of intensive particle random walk (large D/D_m) and very small n_a would require special measures of detecting the events of particles randomly approaching each other or a correct assessment of probability of these events within a time step.

For constraint (90), the minimal allowable value of r_m is given by $r_m^3 = 2\varepsilon_D/(\varepsilon_Z n_r)$ where $n_r \equiv (4/3)\pi(n-1)P_x(\mathbf{x}_1)$ is proportional to the particle density and $V_1=(4/3)\pi r_m^3$ is implied. Condition (89) with $D_m = r_m^2/(5\beta\tau)$ determines the corresponding minimal values for τ and quasisteady variance $\langle z^2\rangle = \chi\tau$ by

$$\langle z^2\rangle_{\min} = \frac{1}{5\beta\varepsilon_D}\left(\frac{2}{n_r}\frac{\varepsilon_D}{\varepsilon_Z}\right)^{2/3}\frac{\chi}{D}. \tag{91}$$

One can see from this equation that enforcing stricter accuracy conditions by reducing ε_D and/or reducing ε_Z increases stochastic scattering and vice versa.

In some cases the value of D can be very small, for example, $D=0$ for conventional Lagrangian particles. In this case, we may have to allow some numerical diffusion D_m which is small with respect to other parameters of the problem but is significantly greater than D. In this case of large or infinite D_m/D, it could be advantageous to satisfy the first (but not the second) condition in Eq. (90). This means that if particles are not engaged in a random walk of noticeable intensity, preserving the independence of particles requires sufficiently large values of n_a as determined by $n_a \ge 2\beta/\varepsilon_Z$. Note that the estimate $\langle z_1 z_2\rangle_{V_1} \sim \langle z_1^2\rangle_1/n_a$ is similar to the estimate $\langle z_1 z_2\rangle \sim \langle z_1^2\rangle_0/n$ obtained in Sec. V for nonlocalized mixing where in both cases n_a and n represent the average number of potential mixing partners.

C. Choosing between mixing schemes

As shown in Sec. III B, numerical diffusion is expressed by the same (or, depending on exact implementations of the models, similar) equations for both two-particle and mean-based mixing. Hence, both mixing schemes will have the same bias induced by numerical diffusion for the same values of r_m and τ. However, we can show that, under certain conditions, two-particle mixing model can afford much smaller values of r_m than mean-based mixing and, because of this, the former model becomes more computationally efficient than the latter.

First we note that Z_m does not exactly coincide with $\langle Z\rangle$; indeed, the level of fluctuations of $z_m = Z_m - \langle Z\rangle$ is given by $\langle z_m^2\rangle = \langle z^2\rangle/n_m$. Here we use Eqs. (15) and (16) while assuming that values Z_p, which belong to n_m particles participating in mixing event, are independent. If we expect that a mean-based mixing model is implemented with a given precision and that stochastic errors in evaluation of $\langle Z\rangle$ do not exceed $\langle z_m^2\rangle/\langle z^2\rangle = n_m^{-1} \le \varepsilon_Z$—the same precision that we demanded for the implementation of the two-particle mixing model in Sec. VII B. While the conventional interpretation of two-particle models requires independence of particles, our interpretation of the mean-based mixing models requires accurate

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evaluation of $\langle Z \rangle$. This requirement distinguishes our definitions of two-particle and mean-based models (without this requirement we can make n_m as small as we like and the smallest possible $n_m=2$ converts the mean-based mixing into two-particle mixing). In order to have n_m particles participating in each mixing event, we must ensure a sufficient average number of particles $n_a \geq c_a n_m$ is located in the mixing volume. Since c_a is, obviously, restricted by $c_a \geq 1$, we assume that c_a is a constant of order of unity. The accuracy restriction imposed on the mean-based mixing model takes the form

$$n_a \geq c_a n_m \geq \frac{c_a}{\varepsilon_Z}. \tag{92}$$

This restriction determines the minimal possible level of stochastic scattering by equation

$$\langle z^2 \rangle_{\min} = \frac{1}{5\beta\varepsilon_D}\left(\frac{c_a}{n_r \varepsilon_Z}\right)^{2/3}\frac{\chi}{D}, \tag{93}$$

which is to be compared to Eq. (91). Note that condition (92) of having a sufficient number of particles in the mixing volume is essentially the same as Eq. (90). However, in the case of two-particle mixing it is sufficient to satisfy condition (90) to ensure the same level of accuracy. We obtain from Eqs. (91) and (93) the following estimates for the minimal stochastic scattering of the models

$$(\langle z^2 \rangle_{\min})_{[2]} \sim \frac{\chi/D}{\varepsilon_D^{1/3}\varepsilon_Z^{2/3} n^{2/3}}, \tag{94}$$

$$(\langle z^2 \rangle_{\min})_{[\langle Z \rangle]} \sim \frac{\chi/D}{\varepsilon_D \varepsilon_Z^{2/3} n^{2/3}}. \tag{95}$$

Two-particle mixing models generally have a better performance compared to mean-based models since the former allow for better localization of mixing. These equations also indicate that, in order to ensure the same numerical accuracy and scattering of the simulations, the mean-based mixing needs a significantly larger number of particles than two-particle mixing,

$$\frac{(n)_{[\langle Z \rangle]}}{(n)_{[2]}} \sim \frac{1}{\varepsilon_D}. \tag{96}$$

We need to stress that this difference in performance of the models is related only to n_m—the number of particles participating in each mixing event. Smoothness of the trajectories generated by mixing models is another factor that can affect the model performance, although to a smaller extent. As shown in Ref. 11, the value of parameter β varies between 1 for discontinuous mixing $\eta=1$ and 2 for continuous mixing $\eta \to 0$. Equations (91) and (93) indicate that a continuous mixing model would perform better and allow for twice smaller scattering $\langle z^2 \rangle_{\min}$ compared to a highly discontinuous model.

VIII. CONCLUSIONS

The main conclusions of this work are now summarized:

- Diffusion can be accurately modeled by Lagrangian particles with random walk and localized mixing which can be seen as an alternative representation of the diffusion.
- Unless the number of particles tends to infinity and localization of mixing is progressively enforced, numerical diffusion is present in the simulations.
- The mean-based models (mixing models involving evaluation of means) are easier to analyze since particles (i.e., scalar values assigned to the particles) are fully independent in these models while two-particle models are probably easier to implement than the mean-based models. Two-particle mixing is shown to be more efficient than mean-based mixing in reducing numerical diffusion for conditions when a limited number of particles is used in simulations.
- In two-particle models, dependences between particles are, typically, small and can be neglected at a global scale. However, the effect of these dependences can be dominant in mixing localities resulting in significant reduction of the mixing rate. The problem of dependences between particles has similarities with the problem of deviations from molecular chaos—the hypothesis of molecular chaos is widely used in statistical physics. We offer a comprehensive analysis of this problem which is based on using a chain of PDF equations that has some similarities with the BBGKY chain used in statistical physics.

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APPENDIX: ASYMPTOTIC EVALUATION OF THE DAMPENING FUNCTIONS

In Appendix we obtain asymptotic solutions for the first two moments in vicinity of mixing volume. Mixing considered here is localized which means that the radius of mixing volume r_m is much smaller than the scale of the problem under consideration r_0. Under these conditions, the diffusion term is dominant so that we can write

$$\tilde{L}_2 \approx -D\nabla_2^2 = -D\frac{\partial^2}{\partial x_1^i \,\partial x_1^i} - D\frac{\partial^2}{\partial x_2^i \,\partial x_2^i} \approx 2D\frac{\partial^2}{\partial y^i \,\partial y^i}, \tag{A1}$$

where the difference between particle positions is denoted as $y^i = x_2^i - x_1^i$ and we use that dependence of the moments on distance between particles which is used in the definition of the mixing volume $r^2 \equiv y^i y^i \leq r_m^2$. In spherical coordinates with the use of $r^2 = y^i y^i$ and $\cos(\varphi) = y^1/r$

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$$\frac{\partial^2}{\partial y^i\,\partial y^i} = \frac{1}{r^{d-1}}\frac{\partial}{\partial r}\left(r^{d-1}\frac{\partial}{\partial r}\right) + \frac{1}{r^2\,\sin(\varphi)^{d-2}}\frac{\partial}{\partial \varphi}\left(\sin(\varphi)^{d-2}\frac{\partial \tilde{K}}{\partial \varphi}\right) + \cdots, \tag{A2}$$

$$r^2 = y^i y^i, \quad \cos(\varphi) = \frac{y^1}{r}.$$

Here, d denotes the dimensionality of the space and we retain only these terms in the Laplace operator that are needed.

1. Variance depression

With $K=K(r)$, Eq. (59) takes the form

$$K'' + \frac{(d-1)}{R}K' = \Lambda^2 K. \tag{A3}$$

Here we denote the derivatives by $K'=dK/dR$ and use Λ defined by $\Lambda=\lambda$ for $R\leq 1$ and $\Lambda=0$ for $R>1$. The boundary conditions are given by

$$K' \to 0 \ \text{as} \ R \to 0 \quad \text{and} \quad K \to \langle z_1^2\rangle_1 \ \text{as} \ R \to \infty. \tag{A4}$$

The solution is represented by the following functions:

$$K = \left\{\begin{array}{ll} \langle z_1^2\rangle_1 C_0 F(\lambda R), & R<1 \\ \langle z_1^2\rangle_1\left(1-\dfrac{C_1}{R^{d-2}}\right), & R>1 \end{array}\right\}, \tag{A5}$$

$$F(\lambda) = \frac{I_{d/2-1}(\lambda)}{\lambda^{d/2-1}}, \quad F'(\lambda) = \frac{I_{d/2}(\lambda)}{\lambda^{d/2-1}}, \quad C_0 = \frac{(1-C_1)}{F(\lambda)}, \tag{A6}$$

$$C_1 = \left(1 + \frac{F(\lambda)}{\lambda F'(\lambda)}(d-2)\right)^{-1}, \quad C_2 = d(d-2)\frac{C_1}{\lambda^2}, \tag{A7}$$

where I_ν denotes the Bessel function of order ν. The integral of K over the mixing volume

$$\int_0^1 \lambda^2 K R^{d-1} dR = \left(R^{d-1}\frac{\partial K}{\partial R}\right)_{R=1} = (d-2)C_1 K_0,$$

$$\int_0^1 R^{d-1} dR = \frac{1}{d}, \tag{A8}$$

$$\langle Z_1^2\rangle_{V_1} - \langle Z_1 Z_2\rangle_{V_1} = \frac{1}{V_1}\int_{V_1} K dV = \frac{d(d-2)C_1}{\lambda^2}\langle z_1^2\rangle_1 = C_2\langle z_1^2\rangle_1$$

determines the intensity of mixing. The limiting asymptotes for the functions C_1 and C_2 are given by $C_1(\lambda)\to 0$, $C_2(\lambda)\to 1$ as $\lambda\to 0$ and $C_1(\lambda)\to 1$, $C_2(\lambda)\to 0$ as $\lambda\to\infty$. In three-dimensional space the equations for C_1 and C_2 can be simplified

$$C_1 = 1 - \frac{\tanh(\lambda)}{\lambda}, \quad C_2 = 3\frac{\lambda - \tanh(\lambda)}{\lambda^3}. \tag{A9}$$

In two-dimensional case obtaining fully steady solution satisfying the boundary conditions is impossible due to logarithmic behavior of the solution. The depression becomes dependent on the outer scale of the problem r_0. For this case we can still approximately write

$$C_1 = \left(\varkappa + \frac{F(\lambda)}{\lambda F'(\lambda)}\right)^{-1}, \quad C_2 = d\frac{C_1}{\lambda^2}, \quad \varkappa = \ln(R_0) = \ln\left(\frac{r_0}{r_m}\right). \tag{A10}$$

Note that one of the asymptotes is different for two-dimensional case: $C_1(\lambda)\to\varkappa^{-1}$ as $\lambda\to\infty$.

2. Attenuation of numerical diffusion

We first expand the mean $\langle Z_p\rangle$ for particles $p=1$ and $p=2$ into the Taylor series up to the second order terms inclusive

$$\langle Z_p\rangle = \langle Z\rangle_0 + \left(\frac{\partial\langle Z\rangle}{\partial x^i}\right)_0 (x_p^i - x_0^i) + \frac{1}{2}\left(\frac{\partial^2\langle Z\rangle}{\partial x^i\,\partial x^j}\right)_0 (x_p^i - x_0^i)(x_p^j - x_0^j) + \cdots.$$

The difference of the single-conditioned first moments is given by

$$\langle Z_2\rangle_2 - \langle Z_1\rangle_1 = Q_0^i(\mathbf{X})y^i, \tag{A11}$$

where Q_0^i denotes the following expression:

$$Q_0^i = \left(\frac{\partial\langle Z\rangle}{\partial x^i}\right)_0 + \frac{1}{2}\left(\frac{\partial^2\langle Z\rangle}{\partial x^i\,\partial x^j}\right)_0 (X^j - 2x_0^j). \tag{A12}$$

The solution of Eq. (73) for $\tilde{K}$ is sought in the form

$$\tilde{K} = \langle Z_2\rangle_{12} - \langle Z_1\rangle_{12} = Q_0^i(\mathbf{X})\tilde{\Phi}(r,y^i), \tag{A13}$$

where equation for $\tilde{\Phi}$ takes the form

$$\frac{\partial^2\tilde{\Phi}}{\partial y^i\,\partial y^i} = \frac{\tilde{\Lambda}^2}{r_m^2}\tilde{\Phi} \tag{A14}$$

and $\tilde{\Lambda}$ is defined as $\tilde{\Lambda}=\tilde{\lambda}$ for $r\leq r_m$ and $\tilde{\Lambda}=0$ for $r>r_m$. The boundary conditions for function $\tilde{\Phi}$ are given by

$$\tilde{\Phi}(r,0) = 0 \quad \text{and} \quad \tilde{\Phi}(r,y^i) \to y^i \ \text{as} \ r\to\infty. \tag{A15}$$

We also note that the functions $\tilde{K}$ and $\tilde{\Phi}$ are antisymmetric with respect to the variable $\mathbf{y}$ that is $\tilde{\Phi}(r,y^1)=-\tilde{\Phi}(r,-y^1)$. With the use of spherical coordinates $r=|\mathbf{y}|$, $y^1=r\cos(\varphi)$, $R=r/r_m$ we represent the solution in the form $\tilde{\Phi}=\cos(\varphi)\tilde{\Phi}_r(R)$ so that

$$\frac{1}{R^{d-1}}\frac{\partial}{\partial R}\left(R^{d-1}\frac{\partial\tilde{\Phi}_r}{\partial R}\right) - \frac{d-1}{R^2}\tilde{\Phi}_r = \tilde{\Lambda}^2\tilde{\Phi}_r, \tag{A16}$$

$$\tilde{\Phi}_r = \left\{\begin{array}{ll} \tilde{C}_0\tilde{F}(\tilde{\lambda}R), & R<1 \\ r - \dfrac{\tilde{C}_1}{R^{d-1}}, & R>1 \end{array}\right\}, \tag{A17}$$

$$\tilde{F}(R) = \frac{I_{d/2}(R)}{R^{d/2-1}}, \quad \tilde{C}_0 = \frac{(1-\tilde{C}_1)}{\tilde{F}(\tilde{\lambda})}, \tag{A18}$$

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$$\tilde{C}_1 = \left(1+\frac{d}{\tilde{\lambda}}\frac{I_{d/2}(\tilde{\lambda})}{I_{d/2+1}(\tilde{\lambda})}\right)^{-1}, \quad \tilde{C}_2 = d(d+2)\frac{\tilde{C}_1}{\tilde{\lambda}^2}. \tag{A19}$$

As in the previous subsection, I_ν denotes the Bessel function of order ν. Since evaluation of the integral

$$\left[\frac{d\langle Z_1\rangle_1}{dt}\right]_{[2]} = (n-1)\theta_0\langle\tilde{\eta}\rangle\int_{V_1}(\langle Z_2\rangle_{12}-\langle Z_1\rangle_{12})P_x(\mathbf{x}_2)d\mathbf{x}_2$$

is performed for constant $\mathbf{x}_1$, the point of expansion $\mathbf{x}_0$ can now be selected at the location of particle 1 so that $x_0^i=x_1^i$ and $X^j-2x_0^j=x_2^i-x_1^i=y^i$. The integral gives zero values for terms linear in y^i, hence only the quadratic terms have to be considered,

$$(\langle Z_2\rangle_{12}-\langle Z_1\rangle_{12})P_x(\mathbf{x}_2) = \cdots + \left(\frac{1}{2}\frac{\partial^2\langle Z\rangle}{\partial x^i\,\partial x^i}P_x + \frac{\partial\langle Z\rangle}{\partial x^i}\frac{\partial P_x}{\partial x^i}\right)_1 y^i\tilde{\Phi}(r,y^i). \tag{A20}$$

With assistance of the integrals

$$\frac{\displaystyle\int_0^1 R^d\frac{I_{d/2}(\tilde{\lambda}R)}{(\tilde{\lambda}R)^{d/2-1}}dR}{\displaystyle\int_0^1 R^{d-1}dR} = d\frac{I_{d/2+1}(\tilde{\lambda})}{(\tilde{\lambda})^{d/2}},$$

$$\frac{\int_0^{\pi/2}\cos(\varphi)^2\sin(\varphi)^{d-2}d\varphi}{\int_0^{\pi/2}\sin(\varphi)^{d-2}d\varphi} = \frac{1}{d},$$

evaluation of the quadratic terms

$$\frac{1}{V_1}\int_{V_1} y^1\tilde{\Phi}(r,y^1)d\mathbf{y} = \frac{r_m^2}{d+2}\tilde{C}_2$$

results in the following expression for the numerical diffusion coefficient:

$$D_m = \frac{n_a\theta_0\langle\tilde{\eta}\rangle\hat{r}_1^2}{2}\tilde{C}_2 = \frac{\hat{r}_1^2}{\beta\tau}\frac{\tilde{C}_2(\tilde{\lambda})}{C_2(\lambda)}. \tag{A21}$$

In the three-dimensional case the equations for $\tilde{C}_1$ and $\tilde{C}_2$ can be simplified:

$$\tilde{C}_1 = \frac{\tilde{\lambda}^2+3-3\tilde{\lambda}\coth(\tilde{\lambda})}{\tilde{\lambda}^2}, \quad \tilde{C}_2 = 15\frac{\tilde{\lambda}^2+3-3\tilde{\lambda}\coth(\tilde{\lambda})}{\tilde{\lambda}^4}. \tag{A22}$$

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