Kinetic and dynamic probability-density-function descriptions of disperse turbulent two-phase flows

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This article analyzes the status of two classical one-particle probability density function (PDF) descriptions of the dynamics of discrete particles dispersed in turbulent flows. The first PDF formulation considers only the process made up by particle position and velocity $\mathbf{Z}_p = (\mathbf{x}_p, \mathbf{U}_p)$ and is represented by its PDF $p(t; \mathbf{y}_p, \mathbf{V}_p)$ which is the solution of a kinetic PDF equation obtained through a flux closure based on the Furutsu-Novikov theorem. The second PDF formulation includes fluid variables into the particle state vector, for example, the fluid velocity seen by particles $\mathbf{Z}_p = (\mathbf{x}_p, \mathbf{U}_p, \mathbf{U}_s)$, and, consequently, handles an extended PDF $p(t; \mathbf{y}_p, \mathbf{V}_p, \mathbf{V}_s)$ which is the solution of a dynamic PDF equation. For high-Reynolds-number fluid flows, a typical formulation of the latter category relies on a Langevin model for the trajectories of the fluid seen or, conversely, on a Fokker-Planck equation for the extended PDF. In the present work, a new derivation of the kinetic PDF equation is worked out and new physical expressions of the dispersion tensors entering the kinetic PDF equation are obtained by starting from the extended PDF and integrating over the fluid seen. This demonstrates that, under the same assumption of a Gaussian colored noise and irrespective of the specific stochastic model chosen for the fluid seen, the kinetic PDF description is the marginal of a dynamic PDF one. However, a detailed analysis reveals that kinetic PDF models of particle dynamics in turbulent flows described by statistical correlations constitute incomplete stand-alone PDF descriptions and, moreover, that present kinetic-PDF equations are mathematically ill posed. This is shown to be the consequence of the non-Markovian characteristic of the stochastic process retained to describe the system and the use of an external colored noise. Furthermore, developments bring out that well-posed PDF descriptions are essentially due to a proper choice of the variables selected to describe physical systems and guidelines are formulated to emphasize the key role played by the notion of slow and fast variables.

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I. INTRODUCTION

As indicated by their name, polydisperse two-phase turbulent flows refer to a two-phase flow regime characterized by the presence of one phase as a set of discrete elements or "particles" having a range of diameters (or typical sizes) and embedded in a flow that is usually turbulent. This regime is always met when the discrete phase is made up by solid particles but is also desirable for droplets and bubbles since it allows to increase the surface to volume ratio and enhance transfer processes between the two phases. Disperse two-phase flows are ubiquitous in industrial and environmental flows, which explains that there is considerable impetus to develop modeling approaches for simulating these complex flows.

In most laboratory, industrial or environmental flows, the Kolmogorov scale (the scale of the smallest eddies in a turbulent flow) is in the range of [50 μm, 1 mm]. This means that small particles or droplets having a diameter of less than, say, 30 μm are usually well below Kolmogorov scales and that a pointwise approximation, whereby these discrete elements are treated as points moving in the fluid flow, is reasonable. This is less the case for bubbly flows as bubbles tend to have larger sizes. In the present work, we consider statistical descriptions in which this pointwise approximation is made. From a physical point of view, this indicates that we are essentially dealing with small particles or droplets dispersed by turbulent flows while the instantaneous fluid flow fields are still regarded as existing at the particle positions. Moreover, we leave out any possible change in the particle or droplet volumes (inert particles) and, therefore, phenomena such as coalescence and breakup (for droplets) or agglomeration and fragmentation (for solid particles) are not considered. Actually, PDF methods are precisely attractive when complex physics, such as chemical reactions, polydisperse particles with evolving diameters, etc., is to be accounted for (see Refs. [1–3] for single-phase flows and Refs. [4–6] for disperse two-phase flows). Yet, though they are not necessary for the developments to follow, the above assumptions are made for the sake of simplicity and in order to concentrate on the treatment of particle dynamics in PDF approaches.

Not surprisingly, disperse two-phase flow modeling can be addressed from different standpoints corresponding to different levels of description that can be classified as microscopic, mesoscopic, and macroscopic. This is a classical terminology of statistical physics where it is usually associated to spatial coarse-graining procedures when one goes from atomistic to hydrodynamical descriptions. It was carried out to two-phase flow statistical modeling in Ref. [4] with the caveat that the coarse-graining procedure has to be understood as a reduction of the available information through a reduction of the degrees of freedom that are explicitly simulated in each formulation. Since then, this classification has proved

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an adequate reference system to bring out the articulation of various statistical models [6–8]. At one end of the spectrum of possible descriptions, microscopic approaches correspond to formulations where all the degrees of freedom of the carrier turbulent fluid flow $N_f^{\rm df}$ plus all the degrees of freedom associated to the chosen description of the set of particles $N_p^{\rm df}$ are explicitly computed. In that case, the total number of degrees of freedom is $N_f^{\rm df}+N_p^{\rm df}$, where $N_f^{\rm df}$ scales as $\mathrm{Re}^{9/4}$ with Re the characteristic Reynolds number of the fluid flow and $N_p^{\rm df}$ is, for example, equal to $6N_p$ with N_p the total number of particles if a description in terms of particle positions $\mathbf{x}_p$ and velocity $\mathbf{U}_p$ is retained for each particle. This represents a full direct numerical simulation (DNS) of the fluid and particle system. At the other end, macroscopic approaches consist in describing two-phase flows by a few statistical moments (such as the particle mean velocity or concentration fields). These macroscopic descriptions are usually referred to as "two-fluid models" and appear as the counterparts of traditional descriptions of single-phase turbulent flows derived by direct application of Reynolds averaging [2,4,9,10]. A direct formulation of the closed set of moments that make up a two-fluid model, following the classical approach to single-phase turbulent flows that leads to models such as the well-known k-ϵ and Reynolds-stress models (RSM), has proved difficult especially for polydisperse particles for which characteristic response times vary over a wide range of values [4,6]. In between these two limits, mesoscopic approaches are typically made up by probabilistic descriptions where the exact dynamics of two-phase flows is replaced by a stochastic model (or its probabilistic formulation in the corresponding sample space). These formulations are called PDF models since they attempt to simulate the probability density function of the variables regarded as relevant to describe two-phase flows. PDF approaches have a twofold interest. First, they provide a direct possibility to simulate two-phase flows (this will be referred to as a "stand-alone PDF description" in the rest of the paper). Second, they represent a consistent way to derive realizable macroscopic models by integration over the sample space variables from which closed two-fluid models can be safely obtained [2,4,9,10]. Though a few models have been proposed for a PDF description of both the fluid and particle phases [4,10], a more practical approach is to follow an hybrid approach where the turbulent fluid flow is described by a macroscopic formulation (a moment approach) while the probabilistic approach is limited to the particle phase. In the present work, we retain this hybrid method and only consider probabilistic descriptions of the disperse phase.

However, what probabilistic formulation should we choose to describe the dynamics of small particles in turbulent flows when only limited information (which consists in a few statistical moments) is available for the fluid flow? Quite interestingly, the choice of such a PDF model involves not one but two hierarchies (see a detailed discussion in Ref. [4]). The first one corresponds to the choice between one-particle, two-particle, etc., or N_p-particle PDF descriptions. This is the famous Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy of statistical physics [11,12], where single-particle dynamics is described by the same set of variables [typically with a particle state vector $\mathbf{Z}_p$ made up by particle position and velocity $\mathbf{Z}_p=(\mathbf{x}_p,\mathbf{U}_p)$]. For wall-bounded inhomogeneous turbulent flows, one-particle PDF remains a relevant choice as two-particle (or N-particle) PDF models have not yet reached a mature level for this general case. For this reason, we adopt a one-particle PDF description which corresponds to a one-point statistical description for the resulting (Eulerian) moments of the disperse phase, in line with what is currently achieved for the fluid phase. This first hierarchy is compounded by a second one related to the choice of the variables used to describe particle dynamics: should we retain only particle positions in the particle state vector (i.e., $\mathbf{Z}_p=[\mathbf{x}_p(t)]$) or include particle position and velocity [i.e., $\mathbf{Z}_p=(\mathbf{x}_p(t),\mathbf{U}_p(t))$] or particle position, velocity, and acceleration, etc.? Note that, since only limited statistical information is available on the fluid flow, this latter choice amounts to include fluid-related variables "seen" by particles in the particle state vector (this point will be exemplified with the governing equations through the typical drag force expression). The issue of selecting the particle state vector in PDF descriptions of particle in turbulent flows is at the core of present considerations.

At the moment, two such one-particle PDF descriptions have been proposed in the literature. The first one retains a particle state vector made up by particle location and velocity, that is, $\mathbf{Z}_p=(\mathbf{x}_p,\mathbf{U}_p)$. This model is developed in sample space through the derivation of an evolution equation for the corresponding PDF $p(t;\mathbf{y}_p,\mathbf{V}_p)$ [where $\mathbf{y}_p$ and $\mathbf{V}_p$ designate the sample space variables corresponding to the random variables $\mathbf{x}_p(t)$ and $\mathbf{U}_p(t)$ respectively]. In this approach, the fluid velocity is treated as an external field "noise" and a closed PDF equation in sample space is proposed by resorting to the Furutsu-Novikov theorem. Given the particle state vector, this model is referred to as a "kinetic PDF model." In the second PDF description, the particle state vector is extended to include the fluid velocity "seen" by particles along their trajectories, whereby we have now $\mathbf{Z}_p=(\mathbf{x}_p,\mathbf{U}_p,\mathbf{U}_s)$ for the particle state vector. This model is essentially developed in physical space through the formulation of a stochastic diffusion process for $\mathbf{Z}_p$ and is often called a Langevin approach since Langevin models are typically used to describe the evolution of the fluid velocity seen $\mathbf{U}_s$ [4–6,13]. As we are dealing with weak approaches where only the law of stochastic processes is approximated [4,8,14,15], the formulation in terms of stochastic differential equations is equivalent to a Fokker-Planck equation for the corresponding density $p(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)$. Given this state vector and that particle accelerations are explicitly treated, this second model is referred to as a "dynamic PDF model."

The question of the choice of the variables entering the particle state vector has been discussed in some works [4,6,13], mostly in connection with the selection of the fluid velocity seen as a relevant particle variable in high-Reynolds-number turbulent fluid flows and in relation with the formulation of a Langevin model for this variable. To the best of the authors' knowledge, no such explicit discussion has ever been proposed in the works devoted to the development of kinetic PDF models [16–19]. This is perhaps due to the fact that $\mathbf{Z}_p=(\mathbf{x}_p,\mathbf{U}_p)$ appears as the direct extension of the "natural choice" made in molecular dynamics studies. However, it is worth emphasizing that the selection of the particle state vector is indeed a choice when we do not consider particles (or molecules) in void but discrete elements embedded in random continuous media, in our case particles carried by turbulent fluid flows.

Thus, a comparative evaluation of the different choices of the particle state vector is needed. Furthermore, these two PDF descriptions are proposed in different formulations and it is of great interest to assess whether they correspond to intrinsically different models or are not two sides of the same coin. For example, since a kinetic PDF $p(t;\mathbf{y}_p,\mathbf{V}_p)$ is always retrieved as the marginal of a dynamic one $p(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)$ by integration over the fluid seen variable $\mathbf{V}_s$, is the kinetic-based PDF model obtained with Furutsu-Novikov-type closure obtained as the marginal of a dynamic-based PDF model closed with Langevin-type models?

The purpose of this article is to analyze the differences and relations between kinetic-PDF and dynamic-PDF models. More precisely, drawing on the issues brought out in the previous paragraph, the aims of the present work are to provide answers to the following questions:

(i) When can we consider that a PDF model constitutes a complete probabilistic description?

(ii) When can we regard a given PDF model as being well posed and an acceptable description?

(iii) Are present kinetic PDF models retrieved as the marginal of current dynamic PDF ones?

(iv) What are the specific assumptions made in each formulation and how can their range of validity be assessed?

(v) Is the selection of the particle state vector a free choice or are there constraints to respect?

In spite of the mathematical flavor of some of the above points, these questions are directly connected to physical issues. In particular, it will be shown that they lead to a new, and arguably simpler, derivation of the kinetic PDF equation while revealing the physical content of the dispersion tensors. Furthermore, well-based PDF approaches are essential for future developments and answers to point (v) are of central interest for physically oriented concerns. Indeed, as mentioned above, proper formulations are needed both to ensure that stand-alone simulations can be run and also to assess the validity of the closed two-fluid models derived from such PDF descriptions.

To that effect, the article is organized as follows. The governing fluid and particle equations are first recalled in Sec. II. The salient aspects of probabilistic descriptions needed for present considerations are recalled in Sec. III, with the definition of the criteria used to assess well-based PDF models stated in Sec. III E. The kinetic and dynamic PDF models are presented in Sec. IV. In Sec. V, a new derivation of the kinetic PDF is obtained which reveals that kinetic-based formulations are retrieved as marginals of dynamic PDF models and a new interpretation of the dispersion tensors is worked out. An in-depth analysis of the kinetic-based PDF equation is detailed in Sec. VII that brings out shortcomings as a stand-alone PDF description. General guidelines are developed in Sec. VIII where it is demonstrated that well-posedness is essentially related to the choice of the particle state vector rather than to a specific closure method. Finally, conclusions and propositions for future works are given in Sec. X.

II. GOVERNING EQUATIONS

In this work, we consider incompressible fluid flows and the fluid phase is described by the continuity and Navier-Stokes equations which, for constant-property flows, have the following form:

$$\frac{\partial U_{f,k}}{\partial x_k} = 0, \tag{1a}$$

$$\frac{\partial U_{f,i}}{\partial t} + U_{f,k}\frac{\partial U_{f,i}}{\partial x_k} = -\frac{1}{\rho_f}\frac{\partial P_f}{\partial x_i} + \nu_f \frac{\partial^2 U_{f,i}}{\partial x_k \partial x_k}, \tag{1b}$$

where $\mathbf{U}_f(t,\mathbf{x})$ is the fluid velocity field, ρ_f its density, ν_f its dynamical viscosity, and $P_f(t,\mathbf{x})$ the fluid pressure. Source terms can be added to account for momentum exchange between the fluid and the discrete particles when two-way coupling (whereby particles influence the fluid phase) is deemed important. However, for dilute flows these source terms are not significant and we do not consider them in the rest of the present work. This is done for the sake of simplicity and to concentrate on the probabilistic treatment of the particle momentum equation.

For particle diameters of the same order of magnitude as the Kolmogorov length scale, the particle momentum equation involves the well-known pressure-gradient, drag, added-mass, and Basset forces [20,21]. In the case of particles smaller than the Kolmogorov scale and heavier than the fluid (droplets in a gas, solid particles in a gas or liquid), the particle momentum equation can be simplified to the following form which retains only the drag and external forces (other forces can be added but this is sufficient for the present discussion):

$$\frac{d\mathbf{x}_p}{dt} = \mathbf{U}_p, \tag{2a}$$

$$\frac{d\mathbf{U}_p}{dt} = \frac{1}{\tau_p}(\mathbf{U}_s - \mathbf{U}_p) + \mathbf{F}_{\text{ext}}. \tag{2b}$$

In Eq. (2b), $\mathbf{F}_{\text{ext}}$ is the acceleration due to some external field forces (typically, $\mathbf{F}_{\text{ext}} = \mathbf{g}$, where $\mathbf{g}$ is the gravity acceleration) and it is seen that the drag force is written with the particle relaxation time τ_p defined as

$$\tau_p = \frac{\rho_p}{\rho_f}\frac{4d_p}{3C_D|\mathbf{U_r}|}, \tag{3}$$

where ρ_p is the particle density and d_p its diameter and where the local instantaneous relative velocity between the fluid and the particle velocity is $\mathbf{U_r} = \mathbf{U_s} - \mathbf{U_p}$. The drag coefficient C_D is usually expressed as a nonlinear function of the particle-based Reynolds number, $\text{Re}_p = d_p|\mathbf{U_r}|/\nu_f$, based on empirical formulas [22]. For example, an often-retained formula is [22,23]

$$C_D = \begin{cases} \frac{24}{\text{Re}_p}\left[1 + 0.15\text{Re}_p^{0.687}\right] & \text{if } \text{Re}_p \leqslant 1000, \\ 0.44 & \text{if } \text{Re}_p \geqslant 1000. \end{cases} \tag{4}$$

In the Stokes regime, that is, when the particle-based Reynolds number Re_p is small (say, $\text{Re}_p \leqslant 1$), the drag coefficient C_D can be safely approximated by $C_D \simeq 24/\text{Re}_p$. In that case, the particle relaxation time τ_p tends towards the Stokes expression τ_p^{st} and we retrieve the well-known formula that

$$\tau_p^{st} = \frac{\rho_p}{\rho_f}\frac{d_p^2}{18\nu_f}. \tag{5}$$

In Eq. (2b) and in the expression of the particle relaxation time scale in Eq. (3), the central variable for the present concerns is $\mathbf{U}_s = (\mathbf{U}_f(t,\mathbf{x}_p(t)), t \geqslant 0)$ which is the "fluid velocity seen," i.e., the fluid velocity sampled along the particle trajectory $\mathbf{x}_p$ as it moves across a turbulent flow [4,13].

At this point, two remarks can be made. First, though the velocity of the fluid is defined from the fluid velocity field (or, in other words, from a Eulerian field), it is a Lagrangian property attached to each particle [a time series of $\mathbf{U}_s(t)$ is only meaningful along a certain particle trajectory]. Second, it is seen that, apart from the Stokes regime when τ_p is a constant, the particle relaxation time is actually a function of both the instantaneous particle and fluid velocities which we can express by writing $\tau_p = \mathcal{F}(\mathbf{U}_p,\mathbf{U}_s)$, where $\mathcal{F}$ stands for the function on the right-hand side of Eq. (3) and with the specific expressions given in Eq. (4).

III. THE PDF FRAMEWORK

The theory of stochastic processes is by now well established and can be found in mathematical reference works [24–27]. Over the past few decades, it has also been summarized in physics textbooks [14,15,28,29] for physically oriented readers with a view towards applications, as well as in comprehensive review articles for PDF approaches to single-phase and two-phase flow modeling [1,4]. For this reason, we limit ourselves to recalling only the key aspects that are of direct interest in the developments to follow. At the core of present considerations are the question of the complete law of a stochastic process, the physically important issue of Markovian and non-Markovian processes, and the relation between Markovian character and so-called "colored-noise" or "white-noise" in dynamical systems. The latter point was the subject of constant interrogation in physics [28,30] but clarifications have emerged in recent years. On this debate, we refer in particular to an interesting and pedagogical paper by Van Kampen [31] that addresses these questions for physicists. These reminders are helpful to bring out specific criteria that are used to assess whether a given probabilistic description is complete and well posed. This is done in Sec. III E.

A. The law of a stochastic process

A stochastic process, noted as $\mathbf{Z}$ or $[\mathbf{Z}(t), t \geqslant 0]$, is a family of random variables indexed by a parameter which is usually time (with $t \in T$ where T is a time interval). We consider vector-valued stochastic processes whose values (in the corresponding sample space which is typically $\Re^d$ where d is the dimension) are noted $\mathbf{z}$. Simply speaking, knowing the law of a stochastic process is equivalent to the knowledge of the joint PDF $p(t_1,\mathbf{z}_1;t_2,\mathbf{z}_2,\ldots;t_N,\mathbf{z}_N)$ for any set of N times and for any values of the chosen times $(t_1;t_2,\ldots;t_N)$. It is thus clear that the amount of information required is huge and, in particular, much larger than the sole access to the one-time PDF $p(t,\mathbf{z})$.

B. Markovian processes

Loosely speaking, a Markov process is a stochastic process for which "knowledge of the whole past" or "knowledge of the present" amounts to the same for predicting the future (in a probabilistic sense). This notion is actually the direct extension of classical deterministic mechanics where the knowledge of an initial condition and of the rate of change is enough to predict the future of a dynamical system. A rigorous definition requires adapted-σ algebras [15,25] but for our purpose it is sufficient to illustrate the Markov property of a stochastic process in a discrete setting by writing that

$$p(t_{n+1},\mathbf{z}_{n+1}|(t_n,\mathbf{z}_n;t_{n-1},\mathbf{z}_{n-1};\ldots;t_1,\mathbf{z}_1)) = p(t_{n+1},\mathbf{z}_{n+1}|(t_n,\mathbf{z}_n)), \tag{6}$$

where t_{n+1} stands for the future, t_n for the present, and $(t_{n-1},\ldots,t_1)$ for the past while $\mathbf{z}_i$ represents the value of the process at $t = t_i$ [i.e., $\mathbf{Z}(t_i) = \mathbf{z}_i$].

It is important to note that the above condition, which is simply written as $(t_n,\mathbf{z}_n)$ in the right-hand side of Eq. (6), represents in fact the *whole information known* at the present time $t = t_n$. For example, if we consider a system upon which an external action is exerted, then this action must be fully determined by the knowledge of $(t_n,\mathbf{z}_n)$ for the Markov property to be valid. This point is essential in the choice of the treatment of "external noises" and will resurface in Sec. VII A.

The fundamental property of Markov processes is that the law of the stochastic process can be completely determined from the knowledge of only two functions: $p(t_0,\mathbf{z}_0)$, which represents an initial PDF condition, and $p(t,\mathbf{z}|(t_0,\mathbf{z}_0))$, which is the conditional or transition PDF from a state $\mathbf{z}_0$ at $t = t_0$ to a state $\mathbf{z}$ at a later time t. This transition PDF plays the role of the time rate of change in classical mechanics and is the central quantity. Then, all the N-time PDFs are easily reconstructed by application of the Chapman-Kolmogorov equation (or chain-rule) [14,15,29] which indicates that the complete law of the process is indeed simulated. This means that any evolution equation for the transition PDF lives up to its name of a master equation (ME) [28,31]. If we introduce the operator $\mathcal{L}$ as the generator of the Markov stochastic process $\mathbf{Z}$, the master equation is the forward-Kolmogorov equation that can be written as

$$\frac{\partial p(t,\mathbf{z}|(t_0,\mathbf{z}_0))}{\partial t} = \mathcal{L}^{\perp}[p(t,\mathbf{z}|(t_0,\mathbf{z}_0))], \tag{7}$$

where $\mathcal{L}^{\perp}$ is the adjoint of the operator $\mathcal{L}$. For a stochastic diffusion process, the forward Kolmogorov equation takes the form of the well-known Fokker-Planck equation

$$\frac{\partial p(t,\mathbf{z}|(t_0,\mathbf{z}_0))}{\partial t} = -\frac{\partial [A_i\, p(t,\mathbf{z}|(t_0,\mathbf{z}_0))]}{\partial z_i} + \frac{1}{2}\frac{\partial^2 [D_{ij}\, p(t,\mathbf{z}|(t_0,\mathbf{z}_0))]}{\partial z_i \partial z_j}, \tag{8}$$

where $\mathbf{A} = (A_i)_{i=1,d}$ is a drift vector and $\mathbf{D} = (D_{ij})_{i,j=1,d}$ is a symmetric definite-positive diffusion matrix.

C. Non-Markovian processes

When the Markov property is not satisfied, not much can be said about a stochastic process. It is of course always possible to consider the one-time PDF $p(t,\mathbf{z})$ of the random variable $\mathbf{Z}(t)$, or the transition PDF $p(t,\mathbf{z}|(t_0,\mathbf{z}_0))$, from which one-time statistical moments can be derived (through the definition of the related distribution function). However, it is not possible to

reconstruct further information on the stochastic process from this single PDF and, therefore, it is not possible to simulate the complete law of the stochastic process. Similarly, one can construct an evolution equation for the one-time PDF (or the transition one) $p(t,\mathbf{z})$, for example, of the form

$$\frac{\partial p(t,\mathbf{z})}{\partial t} = \mathcal{G}[p(t,\mathbf{z})], \tag{9}$$

where $\mathcal{G}$ is an operator, but this equation cannot be considered as the equivalent of a ME [31].

D. To be Markovian or not

It is worth emphasizing that the Markovian property is not related to a physical system but to how we choose to describe it. In other words, this is not an intrinsic characteristic of a physical system but a reflection of the modeling standpoint that we adopt. A classical example illustrates that point and helps to anticipate on the discussions to follow in Sec. VII (see detailed accounts in Refs. [28,30,31], for example, about the historical Kramers equation). We consider a 1D stochastic process Z whose trajectories evolve according to

$$\frac{dZ}{dt} = A(t,Z) + B(t,Z)\xi(t), \tag{10}$$

where ξ is an external noise acting on Z (with A and B given functions), for instance, a stationary Gaussian process. If ξ takes independent values, then ξ corresponds to a "Gaussian white-noise" whose integration over a small time interval can be written as the increment of the Wiener process W, leading to the usual (and mathematically correct) formulation of a stochastic differential equation (SDE) in the Itô sense [15,25],

$$dZ = A(t,Z)\,dt + B(t,Z)\,dW(t). \tag{11}$$

In that case, the increments $dW(t)$ of the "external noise" are independent: The future can be predicted based on present values and, thus, Z is a Markov process. On the other hand, if ξ has a nonzero correlation time τ (a so-called Gaussian colored noise), then it is clear that Z is no longer a Markov process. Yet such a colored Gaussian noise is easily simulated as a stationary Ornstein-Uhlenbeck (OU) process [14] of the form

$$d\xi(t) = -\frac{\xi(t)}{\tau}\,dt + \sqrt{\frac{2\langle\xi^2\rangle}{\tau}}\,dW(t). \tag{12}$$

It is then immediate to see that, while Z is not Markovian, the joint process (Z,ξ) does make up a Markovian process [14,28,29].

E. Criteria for complete and well-posed PDF descriptions

From the above reminders on stochastic processes, we can put forward two criteria. The first one (C1) is that a PDF description is complete if it allows the complete law of the stochastic process to be simulated. With the results of Sec. III B, it is seen that the derivation of the transition PDF of a Markov process, for example, as the solution of an evolution equation of the form Eq. (7), plus an initial PDF is enough to guarantee that this PDF description is complete. For a non-Markovian process, the same amount of information is not enough and, in that case, the PDF description is said to be incomplete. Note that for a non-Markovian process a PDF description can still be complete but this requires additional sources of information on the process. This first criteria is only meaningful if the evolution equations, such as Eq. (7) or Eq. (9), are well-posed stand-alone equations. We raise this as the second criterion (C2) to express that a given PDF model corresponds to a well-based PDF description. These criteria will be at play in Sec. VII.

IV. KINETIC AND DYNAMIC PDF DESCRIPTIONS FOR DISPERSE TWO-PHASE FLOWS

In this section, we present the main characteristics of the kinetic and dynamic PDF models as found in published works. This is useful to introduce the rationale behind each approach and to outline the physical assumptions made in each derivation. Only the relevant aspects and key formula that are of direct interest for the developments in Sec. V and for the discussions in Sec. VII are recalled. Note that notations sometimes differ from those in the original works but this is done to make the connection between the two PDF descriptions easier to read (connections with the original notations are made whenever necessary).

A. The kinetic-based PDF approach

Over the past 20 years, several derivations of a kinetic-based PDF equation have been proposed. Yet, in the following, we refer essentially to a recent study [18] that discusses these attempts and proposes an updated derivation based on the Furutsu-Novikov theorem (therefore, historical references can be found in Ref. [18]).

As mentioned in the Introduction, the kinetic PDF model retains only particle position and velocity in the particle state vector, which means that we are dealing with $\mathbf{Z}_p = (\mathbf{x}_p,\mathbf{U}_p)$ and its corresponding PDF $p(t;\mathbf{y}_p,\mathbf{V}_p)$. Note that we explicitly use different notations to distinguish between the random variables at a given time t and their possible values in sample space: for that reason, we use $\mathbf{y}_p$ for the sample space value of the random variable $\mathbf{x}_p(t)$ (regrettably, the same notation $\mathbf{x}$ is often used in PDF derivations which can lead to confusion, especially when Lagrangian PDFs where $\mathbf{x}$ is a variable and Eulerian PDFs where $\mathbf{x}$ is a parameter are to be manipulated) and $\mathbf{V}_p$ for the random variable $\mathbf{U}_p(t)$ at time t.

The PDF equation is usually derived from the governing equations Eqs. (2) by manipulation of the fine-grained PDF $\mathcal{P}(t;\mathbf{y}_p,\mathbf{V}_p) = \delta(\mathbf{x}_p(t)-\mathbf{y}_p)\delta(\mathbf{U}_p(t)-\mathbf{V}_p)$ since we have that $p(t;\mathbf{y}_p,\mathbf{V}_p) = \langle\mathcal{P}(t;\mathbf{y}_p,\mathbf{V}_p)\rangle$. In this description, the velocity of the fluid seen $\mathbf{U}_s(t)$ is an external variable. From the particle momentum equation Eq. (2a), it is then clear that the drag force term induces unclosed terms in the corresponding PDF equation due to the velocity of the fluid seen but also to the particle relaxation time [since $\tau_p = \mathcal{F}(\mathbf{U}_p,\mathbf{U}_s)$ as discussed at the end of Sec. II]. The exact unclosed PDF equation has the following general form [4]:

$$\frac{\partial p}{\partial t} + \frac{\partial[V_{p,i}\,p]}{\partial y_{p,i}} = -\frac{\partial[F_{\mathrm{ext},i}\,p]}{\partial V_{p,i}} + \frac{\partial}{\partial V_{p,i}}\left[\left\langle\frac{V_{p,i}}{\tau_p}|(\mathbf{y}_p,\mathbf{V}_p)\right\rangle p\right] - \frac{\partial}{\partial V_{p,i}}\left[\left\langle\frac{U_{s,i}}{\tau_p}|(\mathbf{y}_p,\mathbf{V}_p)\right\rangle p\right]. \tag{13}$$

It is worth noting that the complete expression of the particle relaxation time formulated in Eqs. (4) cannot be exactly accounted for in the kinetic PDF description. In practice, one has to approximate τ_p by a constant, that is, assume that particles remain in the Stokes regime so $\tau_p \simeq \tau_p^{\rm st}$ with the Stokes time scale expressed in Eq. (5).

With this approximation, the unclosed kinetic PDF equation is written as

$$\frac{\partial p}{\partial t} + \frac{\partial [V_{p,i}\ p]}{\partial y_{p,i}} = -\frac{\partial [F_{{\rm ext},i}\, p]}{\partial V_{p,i}} + \frac{\partial}{\partial V_{p,i}}\left[\frac{V_{p,i}}{\tau_p^{\rm st}} p\right] - \frac{\partial}{\partial V_{p,i}}\left[\frac{1}{\tau_p^{\rm st}}\langle U_{f,i}\rangle p\right] - \frac{\partial}{\partial V_{p,i}}\left[\frac{1}{\tau_p^{\rm st}}\langle u_{s,i}^{'}|(\mathbf{y}_p,\mathbf{V}_p)\rangle p\right], \quad (14)$$

where the velocity of the fluid seen has been decomposed as $\mathbf{U}_s = \langle \mathbf{U}_f\rangle(t,\mathbf{x}_p(t)) + \mathbf{u}_s^{'}$. This is the form given in Ref. [18] (cf. Eq. (4) in Ref. [18]) where the notation f_i corresponds to $f_i(t,\mathbf{x}_p(t)) = u_{f,i}(t,\mathbf{x}_p(t))/\tau_p^{\rm st} = u_{s,i}^{'}/\tau_p^{\rm st}$ [where $\mathbf{u}_f(t,\mathbf{x}) = \mathbf{U}_f(t,\mathbf{x}) - \langle \mathbf{U}_f(t,\mathbf{x})\rangle$ is the fluctuating velocity field of the fluid] and since we have that

$$\langle \mathcal{P} f_i\rangle = \frac{1}{\tau_p^{\rm st}}\langle u_{s,i}^{'}|(\mathbf{y}_p,\mathbf{V}_p)\rangle\, p(t;\mathbf{y}_p,\mathbf{V}_p). \quad (15)$$

From the above equations, we can note that the original derivation handles both a Lagrangian PDF $p(t;\mathbf{y}_p,\mathbf{V}_p)$ and an external (Eulerian) field since the velocity of the fluid seen is treated as the value of the field $\mathbf{f}(t,\mathbf{x})$ at the particle location. As a consequence, it is essential to remark that in Eq. (14) the velocity of the fluid seen has been decomposed with respect to the mean value of *the fluid velocity field* at the particle position, namely $\langle \mathbf{U}_f\rangle(t,\mathbf{x}_p(t))$. However, if we can say that at a given point $\mathbf{x}$ we have of course that $\langle f_i(t,\mathbf{x})\rangle = 0$ which follows from the very definition of its (Eulerian) mean value since $f_i(t,\mathbf{x}) = 1/\tau_p^{\rm st}(U_{f,i}(t,\mathbf{x}) - \langle U_{f,i}(t,\mathbf{x})\rangle)$, it cannot be said that $\langle f_i(t,\mathbf{x}_p(t))\rangle = 0$ even for particles located at a given point. The distinction between the parameter $\mathbf{x}$ and the variable $\mathbf{x}_p(t)$ is here at play. The usually nonzero value of $\langle u'_{s,i}\rangle$ is called the drift velocity and will be shown to play an important role in Sec. V.

Closure of the flux in the kinetic sample space due to the velocity of the fluid seen is obtained by resorting to the Furustsu-Novikov theorem [which is also called the Furutsu-Novikov-Donsker (FND) relation [32–35], a terminology that we retain in the following]. The explicit derivation is detailed in Ref. [18] (see in particular the appendix in Ref. [18]) and is not repeated here. It is, however, useful to state the form of the FND relation which was applied as well as an equivalent form for particle-based functionals. Following the above remarks, the FND relation is written to express the correlation between a Gaussian field with zero mean and an arbitrary functional of that field. It is applied here by assuming that the fluctuating velocity field ($\mathbf{u}_f(t,\mathbf{x})$) is indeed a random Gaussian field and writing for an adapted functional $F[t;\mathbf{u}_f]$ of that Gaussian field

$$\langle u_{f,i}(t,\mathbf{x}) F[t;\mathbf{u}_f]\rangle = \int_0^t \int_{\mathbf{x}'} R_{ik}(t,\mathbf{x};t',\mathbf{x}')\left\langle \frac{\delta F[t;\mathbf{u}_f]}{\delta u_{f,k}(t',\mathbf{x}')}\right\rangle d\mathbf{x}' dt', \quad (16)$$

where $R_{ik}(t,\mathbf{x};t',\mathbf{x}') = \langle u_{f,i}(t,\mathbf{x})\, u_{f,k}(t',\mathbf{x}')\rangle$ is the fluid two-point two-time correlation (this corresponds to Eq. (A1) in Ref. [18] with $u_{f,i} = \tau_p^{\rm st} f_i$). Yet, for particle variables which are expressed as a functional F^p of the field $\mathbf{u}_f(t,\mathbf{x})$ (for example, $F^p = \mathcal{P}$), only functional derivatives at points $\mathbf{x}'$ corresponding to previous particle locations contribute to the integral and we have

$$\frac{\delta F^p}{\delta u_{f,k}(t',\mathbf{x}')} = \frac{\delta F^p}{\delta u_{f,k}(t',\mathbf{x}_p(t'))}\delta(\mathbf{x}_p(t') - \mathbf{x}'), \quad (17)$$

which yields the practical form of the FND relation for such functionals F^p

$$\langle u_{f,i}(t,\mathbf{x})\, F^p\rangle = \int_0^t \left\langle R_{ik}(t,\mathbf{x};t',\mathbf{x}_p(t'))\frac{\delta F^p}{\delta u_{f,k}(t',\mathbf{x}_p(t'))}\right\rangle dt'. \quad (18)$$

This form is identical to Eqs. (A3) and (A5) in the appendix of Ref. [18]. Then, by applying these formulas to the fine-grained PDF $\mathcal{P}$, the following closure for the flux in sample space $\langle \mathcal{P} f_i\rangle$ is obtained [18]:

$$\langle \mathcal{P} f_i\rangle = \kappa_i\ p - \frac{\partial [\lambda_{ij}\ p]}{\partial y_{p,j}} - \frac{\partial [\mu_{ij}\ p]}{\partial V_{p,j}}. \quad (19)$$

In this equation, $\lambda_{ij}(t;\mathbf{y}_p,\mathbf{V}_p)$, $\mu_{ij}(t;\mathbf{y}_p,\mathbf{V}_p)$, and $\kappa_i(t;\mathbf{y}_p,\mathbf{V}_p)$ are referred to as the "dispersion tensors" and are derived by further manipulation of the above relations [see Eqs. (6)–(8) in Ref. [18] where, for a fixed i, the notation λ_{ki} is used]. As the expressions for λ_{ij} and μ_{ij} play an important role in the new approach developed in Sec. V, they are repeated here

$$\lambda_{ij} = \frac{1}{\tau_p^{\rm st}}\int_0^t \langle \Gamma_{jk}(t,t') R_{ik}(t,\mathbf{y}_p;t',\mathbf{x}_p(t'))\rangle_{(\mathbf{y}_p,\mathbf{V}_p)}\, dt', \quad (20a)$$

$$\mu_{ij} = \frac{1}{\tau_p^{\rm st}}\int_0^t \langle \dot{\Gamma}_{jk}(t,t')\, R_{ik}(t,\mathbf{y}_p;t',\mathbf{x}_p(t'))\rangle_{(\mathbf{y}_p,\mathbf{V}_p)}\, dt', \quad (20b)$$

where the notation $\langle . \rangle_{(\mathbf{y}_p,\mathbf{V}_p)}$ indicates the averaged value conditioned on the particle trajectory that "arrive" at $(\mathbf{y}_p,\mathbf{V}_p)$ at time t, which explains that the dispersion tensors are functions of $(\mathbf{y}_p,\mathbf{V}_p)$ even if the eventual dependence on $\mathbf{V}_p$ is often neglected. In these equations, $\Gamma_{jk}(t,t')$ stands for the response function

$$\Gamma_{jk}(t,t') = \frac{\delta x_{p,j}(t)}{\delta u_{f,k}(t',\mathbf{x}_p(t'))}, \quad (21)$$

that measures the effect of a perturbation of the fluctuating fluid velocity seen at an earlier time on the particle position $\mathbf{x}_p(t)$ at time t, and $\dot{\Gamma} = \frac{\partial}{\partial t}\Gamma$.

B. The dynamic-based PDF approach

In dynamic PDF models, the particle state vector is extended to include the velocity of the fluid seen, $\mathbf{Z}_p = (\mathbf{x}_p,\mathbf{U}_p,\mathbf{U}_s)$, and the PDF becomes $p(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)$, where $\mathbf{V}_s$

is used to denote the sample space variable corresponding to the stochastic process $\mathbf{U}_s$. With this extension of the particle state vector, it is seen that the particle momentum equation Eq. (2b) is now treated without approximation even with the complete formula for the particle relaxation time in Eqs. (4). This means that the corresponding terms in the PDF equation appear in a closed form. Indeed, a straightforward derivation of the evolution of $p(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)$ yields [4]

$$\frac{\partial p}{\partial t}+\frac{\partial[V_{p,i}\,p]}{\partial y_{p,i}}=-\frac{\partial}{\partial V_{p,i}}\left[\left(\frac{V_{s,i}-V_{p,i}}{\tau_p}\right)p\right]-\frac{\partial}{\partial V_{s,i}}[\langle\Theta_{s,i}|(\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)\rangle\,p], \quad (22)$$

which is still an unclosed equation but where the closure issue has been shifted to the conditional average of the "acceleration" of the fluid seen $d\mathbf{U}_s/dt=\mathbf{\Theta}_s$ which is now to be modeled. The change from the open kinetic-based PDF equation Eq. (13) to the open dynamic-based PDF equation in Eq. (22) illustrates the second hierarchy mentioned in the Introduction.

The derivation of the closed dynamic PDF equation is developed essentially in physical space by considering the evolution equation for the trajectories of the process $\mathbf{Z}_p$

$$\frac{d\mathbf{x}_p}{dt}=\mathbf{U}_p, \quad (23a)$$

$$\frac{d\mathbf{U}_p}{dt}=\frac{1}{\tau_p}(\mathbf{U}_s-\mathbf{U}_p)+\mathbf{F}_{\rm ext}, \quad (23b)$$

$$\frac{d\mathbf{U}_s}{dt}=\mathbf{\Theta}_s(t). \quad (23c)$$

The key modeling step consists in decomposing the acceleration into slow and fast variations such as

$$\mathbf{\Theta}_s(t)=\mathbf{A}_s+\mathbf{B}_s\boldsymbol{\xi}_s(t), \quad (24)$$

with $\mathbf{A}_s$ and $\mathbf{B}_s$ functions of the system and where $\boldsymbol{\xi}_s$ stands for a rapidly varying noise which, from Kolmogorov theory [2,35,36], has an integral time scale of the order of the Kolmogorov time scale τ_η. Then, for high-Reynolds-number turbulent flows, the fast part of the fluid acceleration can be replaced by a white-noise term when the system is observed at times larger than τ_η. The rationale behind the model and comprehensive discussions of the modeling steps as well as detailed presentations of the construction of the model have already been given [4–6,13,37]. For our present purpose, it suffices to express that the exact particle equations in Eqs. (23) are modeled by the following SDEs:

$$d\mathbf{x}_p=\mathbf{U}_p\,dt, \quad (25a)$$

$$d\mathbf{U}_p=\frac{1}{\tau_p}(\mathbf{U}_s-\mathbf{U}_p)\,dt+\mathbf{F}_{\rm ext}\,dt, \quad (25b)$$

$$d\mathbf{U}_s=\mathbf{A}_s(t,\mathbf{Z}_p,\langle\mathcal{F}[\mathbf{Z}_p]\rangle)\,dt+\mathbf{B}_s(t,\mathbf{Z}_p,\langle\mathcal{G}[\mathbf{Z}_p]\rangle)\,d\mathbf{W}, \quad (25c)$$

where $d\mathbf{W}$ is a vector of independent Wiener processes and where a complete notation has been introduced in the drift vector $\mathbf{A}_s$ and the diffusion matrix $\mathbf{B}_s$ to indicate that, in general, these coefficients depend not only on the particle state vector but also on some statistics of the process (this is known as McKean SDEs [38]).

In the physics literature, these equations [and in particular Eq. (25c)] are referred to as Langevin equations, which is justified by the fact that the drift vector contains usually a return-to-equilibrium term [37]. In a weak sense, whereby we are only interested in approximating the law of stochastic processes (see definitions in Refs. [15,25]), the modeled SDEs are equivalent in sample space to the well-known Fokker-Planck equation

$$\frac{\partial p}{\partial t}+\frac{\partial[V_{p,i}\,p]}{\partial y_{p,i}}=-\frac{\partial}{\partial V_{p,i}}\left[\left(\frac{V_{s,i}-V_{p,i}}{\tau_p}\right)p\right]-\frac{\partial[A_{s,i}\,p]}{\partial V_{s,i}}+\frac{1}{2}\frac{\partial^2\left[\left(B_sB_s^T\right)_{ij}p\right]}{\partial V_{s,i}\partial V_{s,j}}. \quad (26)$$

From a physical point of view, two remarks are in order. First, it is seen that this model relies on the shift from a fast but still colored noise [$\boldsymbol{\xi}_s$ in Eq. (24)] to a white-noise term in Eq. (25c). Second, since the increments of a Wiener process are normally distributed, there is also a Gaussian hypothesis built into the model. However, it is important to note that it is only the *conditional acceleration of the velocity of the fluid seen*, which we can write as $d\mathbf{U}_s|(\mathbf{Z}_p(t)=\mathbf{z}_p)$, that is assumed to be Gaussian. As emphasized repeatedly [4,6,8], the resulting process $\mathbf{Z}_p$ can deviate from Gaussianity when the drift is nonlinear or when the diffusion matrix $\mathbf{B}_s$ is space dependent and thus nonconstant in inhomogeneous flows. Numerical examples of such deviations, which can be marked depending on the choice of the stochastic process, can be found, for instance, in Ref. [2, Sec. 12] and a discussion of this question has recently been given in Ref. [6, Sec. 4.3.1].

C. Consistency issues between PDF descriptions

At this stage, a natural question is whether the marginal PDF (over $\mathbf{V}_s$) derived from a dynamic PDF model is equivalent to a kinetic one. In order to distinguish between PDFs arising from different modeling approaches, we note $p^{\rm km}(t;\mathbf{y}_p,\mathbf{V}_p)$ (where the superscript km stands for kinetic model) for the kinetic PDF model, that is, the solution of the kinetic PDF equation Eq. (14) with Eq. (19), and $p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)$ (where the superscript dm stands for dynamic model) for the solution of the dynamic PDF model, that is, the solution of Eq. (26). Indeed, since we can always extract a kinetic-based PDF description from a dynamic one by integration over the extra variable

$$p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p)=\int p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)d\mathbf{V}_s \quad (27)$$

and that we can obtain the flux closure $\langle\mathcal{P}f_i\rangle$ at the level of the kinetic PDF description as

$$\frac{1}{\tau_p^{\rm st}}\langle u'_{s,i}|\mathbf{y}_p,\mathbf{V}_p\rangle p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p)=\frac{1}{\tau_p^{\rm st}}\int[V_{s,i}-\langle U_{f,i}\rangle(t;\mathbf{y}_p)]p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)dV_{s,i} \quad (28)$$

it can be wondered whether we have

$$p^{\rm km}(t;\mathbf{y}_p,\mathbf{V}_p)\stackrel{?}{=}\int p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)d\mathbf{V}_s, \quad (29)$$

which is equivalent to asking whether we retrieve the same flux closure, that is,

$$\frac{1}{\tau_p^{\rm st}}\langle u'_{s,i}|\mathbf{y}_p,\mathbf{V}_p\rangle p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p) \stackrel{?}{=} \kappa_i\, p^{\rm dm} - \frac{\partial[\lambda_{ij}\, p^{\rm dm}]}{\partial y_{p,j}} - \frac{\partial[\mu_{ij}\, p^{\rm dm}]}{\partial V_{p,j}}. \quad (30)$$

If Eq. (30) is satisfied with dispersion tensors that have the same form as those given in Eqs. (20a) and (20b), then $p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p)$ follows the same evolution equation as $p^{\rm km}(t;\mathbf{y}_p,\mathbf{V}_p)$ from which it can be expected that the two PDFs are identical. This represents the consistency issue between the two modeling approaches.

V. THE KINETIC PDF IS THE MARGINAL OF THE DYNAMIC PDF

To investigate the consistency issue presented in Sec. IV C, we start with a dynamic PDF model and we limit ourselves to the case of a constant particle relaxation time scale, i.e., $\tau_p = \tau_p^{\rm st}$, since general expressions of τ_p cannot be treated in the kinetic PDF approach.

For the sake of clarity, the notation $\mathbf{Z}_p$ is kept to refer to the dynamic-PDF particle state vector, i.e., $\mathbf{Z}_p = (\mathbf{x}_p,\mathbf{U}_p,\mathbf{U}_s)$, while $\mathbf{Z}_p^r = (\mathbf{x}_p,\mathbf{U}_p)$ (where r stands for reduced) is used for the kinetic-PDF one. Conversely, $\mathbf{z}_p = (\mathbf{y}_p,\mathbf{V}_p,\mathbf{V}_s)$ is used to denote the sample space values of $\mathbf{Z}_p$ while $\mathbf{z}_p^r = (\mathbf{y}_p,\mathbf{V}_p)$ represents samples space values of the reduced state vector $\mathbf{Z}_p^r$. As seen in Sec. IV B, the dynamic PDF description follows a full Lagrangian approach where the velocity of the fluid seen is treated as a particle-attached variable. By integrating the closed FPE in Eq. (26) over all the sample space values $\mathbf{V}_s$, we retrieve the open kinetic PDF equation under the form

$$\frac{\partial p^r}{\partial t} + \frac{\partial[V_{p,i}\; p^r]}{\partial y_{p,i}} = -\frac{\partial[F_{{\rm ext},i}\, p^r]}{\partial V_{p,i}} + \frac{\partial}{\partial V_{p,i}}\left[\frac{V_{p,i}}{\tau_p^{\rm st}}\, p^r\right] - \frac{\partial}{\partial V_{p,i}}\left[\frac{1}{\tau_p^{\rm st}}\langle U_{s,i}|\mathbf{z}_p^r\rangle\, p^r\right] \quad (31)$$

and the closure issue is to derive a closed formula for $1/\tau_p^{\rm st}\langle U_{s,i}|\mathbf{z}_p^r\rangle\, p^r$.

For the developments to follow, it is useful to introduce specific notations to clarify the meaning of the variables that are manipulated and of the hypotheses that are made. Indeed, the notion of the velocity of the fluid seen contains a "double randomness": One is related to the intrinsic turbulence of the fluid velocity field and the other one to the random particle positions. These two sources of randomness are of course related since random particle locations are induced by the cumulative effects of the fluctuating fluid velocities seen, as emphasized by the compact notation used for the velocity of the fluid seen $\mathbf{U}_s(t) = \mathbf{U}_f(t,\mathbf{x}_p(t))$. Note that, in the dynamic PDF approach, we do not have access to the whole law of the field $(t,\mathbf{x}) \longmapsto \mathbf{U}_f(t,\mathbf{x})$ but only to the law of $\mathbf{U}_f$ along particles trajectories, that is, only to the law of $\mathbf{U}_f(t,\mathbf{y}_p)$ conditionally on the event $\{\mathbf{x}_p(t) = \mathbf{y}_p\}$.

As in the derivation of the kinetic PDF approach, we therefore assume that we are given a covariance function $R_{ij}(t,\mathbf{x};t',\mathbf{x}')$ (which is the fluid two-time two-point correlation as in Sec. IV A) and the mean velocity field $\langle\mathbf{U}_f\rangle(t;\mathbf{x})$. Then, using the same strong "kinetic assumption" that the fluid velocity field is Gaussian, we can reconstruct $(t,\mathbf{x}) \longmapsto \mathbf{U}_f(t,\mathbf{x})$ from these two moments. It is generally assumed that the correspondence between the first two moments of $\mathbf{U}_s$ coming from a dynamic PDF model and the correlations of the fluid velocity field is expressed by

$$\langle\mathbf{U}_s(t)|\mathbf{x}_p(t) = \mathbf{x}\rangle = \langle\mathbf{U}_f\rangle(t,\mathbf{x}) + \mathbf{U}_d(t,\mathbf{x}), \quad (32a)$$

$$\langle u_{s,i}(t)u_{s,j}(t')|\mathbf{x}_p(t) = \mathbf{x},\, \mathbf{x}_p(t') = \mathbf{x}'\rangle = R_{ij}(t,\mathbf{x};t',\mathbf{x}'), \quad (32b)$$

with $u_{s,i}(t) = U_{s,i}(t) - \langle U_{s,i}(t)|\mathbf{x}_p(t) = \mathbf{x}\rangle$. In Eq. (32a), it is essential to remark that the mean value of the fluid velocity seen by particles at a given point is not equal to the local mean value of the fluid velocity field, with the difference being the so-called drift velocity written as $\mathbf{U}_d$ (see Refs. [5,9,37]) and already introduced in Sec. IV A. This point was recently raised as one criterion used to assess stochastic models [37] (see in particular a discussion of the different definitions of "fluctuations" for particle-attached fluid quantities in Section V.A.3 in Ref. [37]). It is worth stressing that these relations express simply how field properties are derived from a set of instantaneous particle variables (see detailed presentations in Refs. [2,4,6]).

We can now introduce a set of deterministic functions $\omega[t,\mathbf{z}_p^r]$ that arrive at $\mathbf{z}_p^r$ at time t and consider the fluid process along these sample bridges

$$\mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]}(t') = \mathbf{U}_f(t',\omega[t,\mathbf{z}_p^r](t')), \qquad t' \leqslant t. \quad (33)$$

It is important to note that, thanks to the kinetic assumption, this construction is defined independently from the particle state vector. Then, the closure of the kinetic PDF equation relies on the tower property of conditional expectations:

$$\left\langle\mathbf{U}_s(t)\middle|\mathbf{Z}_p^r(t) = \mathbf{z}_p^r\right\rangle = \left\langle\mathbf{U}_f\left(t,\mathbf{y}_p^r\right)\middle|\mathbf{Z}_p^r(t) = \mathbf{z}_p^r\right\rangle, \quad (34)$$

$$= \left\langle\left\langle\mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]}(t)\middle|\mathbf{Z}_p^r = \omega\left[t,\mathbf{z}_p^r\right]\right\rangle\right\rangle_{\omega[t,\mathbf{z}_p^r]}, \quad (35)$$

where the second expectation on the right-hand side is taken over all the sample bridges $\omega[t,\mathbf{z}_p^r]$ arriving at $\mathbf{z}_p^r$ at time t. With this equation, it is seen that the stochasticity of $\mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]}$ is essentially due to the turbulence of the underlying fluid along that sample bridge. Note that this second averaging is written as $\langle.\rangle_{(\mathbf{y}_p,\mathbf{V}_p)}$ in Eqs. (20a) and (20b) but the notation $\langle.\rangle_{\omega[t,\mathbf{z}_p^r]}$ is kept in Eq. (35) for consistency reasons to emphasize the meaning of this second averaging operator with respect to the first one. Based on these notations and on the Gaussian hypothesis for the fluid velocity field, two equivalent roads lead to a closed kinetic PDF equation.

The first road is somewhat similar to the one followed in Ref. [18] but makes use of the FND relation in a Lagrangian framework [33]. This formulation states that the correlation between a Gaussian centered process $\boldsymbol{\zeta}$ and an adapted functional of that process $F[t;\boldsymbol{\zeta}]$ is given by

$$\langle\zeta_i(t)F[t;\boldsymbol{\zeta}]\rangle = \int_0^t \langle\zeta_i(t)\zeta_k(t')\rangle\left\langle\frac{\delta F[t;\boldsymbol{\zeta}]}{\delta\zeta_k(t')}\right\rangle dt'. \quad (36)$$

This relation is similar to the one given in Eq. (18). However, in Sec. IV A we were handling fields and following a Eulerian point of view for the fluid seen [treated as a field noise equal

to $\tau_p^{\rm st}\mathbf{f}(t,\mathbf{x})$], whereas here we are adopting a Lagrangian viewpoint. To apply Eq. (36) to the velocity of the fluid seen on a possible particle bridge, the Gaussian process $\mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]}$ must be first centered. This is done by decomposing

$$U_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t) = \left\langle U_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\right\rangle + u_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t), \tag{37}$$

where $u_{f,i}^{\omega[t,\mathbf{z}_p^r]}$ is now a centered Gaussian process. We are thus led to compute

$$\begin{aligned}&\frac{1}{\tau_p^{\rm st}}\left\langle U_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\middle|\mathbf{z}_p^{r,\omega[t,\mathbf{z}_p^r]}\right\rangle\\ &\quad=\frac{1}{\tau_p^{\rm st}}\left\langle U_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\right\rangle+\frac{1}{\tau_p^{\rm st}}\left\langle u_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\middle|\mathbf{z}_p^{r,\omega[t,\mathbf{z}_p^r]}\right\rangle,\end{aligned} \tag{38}$$

where $\mathbf{z}_p^{r,\omega[t,\mathbf{z}_p^r]}$ stands for the event $\{\mathbf{Z}_p^r=\omega[t,\mathbf{z}_p^r]\}$. By applying Eq. (36) to $\mathcal{P}$ taken as a functional of the process $\mathbf{u}_f^{\omega[t,\mathbf{z}_p^r]}$ we deduce that

$$\begin{aligned}&\left\langle u_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\middle|\mathbf{z}_p^{r,\omega[t,\mathbf{z}_p^r]}\right\rangle p^r\left(t,\mathbf{z}_p^{r,\omega[t,\mathbf{z}_p^r]}\right)\\ &\quad=\int_0^t\left\langle u_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\,u_{f,k}^{\omega[t,\mathbf{z}_p^r]}(t')\right\rangle\left\langle\frac{\delta\mathcal{P}}{\delta u_{f,k}^{\omega[t,\mathbf{z}_p^r]}(t')}\right\rangle dt'\end{aligned} \tag{39}$$

or, conversely, that

$$\begin{aligned}&\left\langle u_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\middle|\mathbf{z}_p^{r,\omega[t,\mathbf{z}_p^r]}\right\rangle p^r\left(t,\mathbf{z}_p^{r,\omega[t,\mathbf{z}_p^r]}\right)\\ &\quad=\int_0^t R_{ik}(t,\omega[t,\mathbf{z}_p^r](t);\,t',\omega[t,\mathbf{z}_p^r](t'))\left\langle\frac{\delta\mathcal{P}}{\delta u_{f,k}^{\omega[t,\mathbf{z}_p^r]}(t')}\right\rangle dt'.\end{aligned} \tag{40}$$

The functional derivative of $\mathcal{P}$ is obtained with the usual manipulations of fine-grained PDFs which give

$$\frac{\delta\mathcal{P}}{\delta u_{s,k}^{\omega[t,\mathbf{z}_p^r]}(t')}=-\Gamma_{jk}(t,t')\frac{\partial\mathcal{P}}{\partial y_{p,j}}-\dot\Gamma_{jk}(t,t')\frac{\partial\mathcal{P}}{\partial V_{p,j}} \tag{41}$$

with the response tensor $\Gamma_{jk}(t,t')$ as in Eq. (21). Finally, plugging back (41) in (40) and integrating over all the sample space trajectories that arrive at $\mathbf{z}_p^r$ at time t, we obtain that

$$\begin{aligned}\frac{1}{\tau_p^{\rm st}}\left\langle U_{s,i}\middle|\mathbf{z}_p^r\right\rangle p^r\;=\;&\frac{1}{\tau_p^{\rm st}}\left\langle\left\langle U_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\right\rangle\right\rangle_{\omega[t,\mathbf{z}_p^r]}p^r\\ &-\lambda_{ij}^r\frac{\partial p^r}{\partial y_{p,j}}-\mu_{ij}^r\frac{\partial p^r}{\partial V_{p,j}},\end{aligned} \tag{42}$$

where λ_{ij}^r and μ_{ij}^r are given respectively by

$$\begin{aligned}\lambda_{ij}^r\left(t;\mathbf{z}_p^r\right)=\frac{1}{\tau_p^{\rm st}}\int_0^t&\left\langle\Gamma_{jk}(t,t')\right.\\ &\left.\times R_{ik}\left(t,\omega\left[t,\mathbf{z}_p^r\right](t);t',\omega\left[t,\mathbf{z}_p^r\right](t')\right)\right\rangle_{\omega[t,\mathbf{z}_p^r]}dt'\end{aligned} \tag{43}$$

and by

$$\begin{aligned}\mu_{ij}^r\left(t;\mathbf{z}_p^r\right)=\frac{1}{\tau_p^{\rm st}}\int_0^t&\left\langle\dot\Gamma_{jk}(t,t')\right.\\ &\left.\times R_{ik}\left(t,\omega\left[t,\mathbf{z}_p^r\right](t);t',\omega\left[t,\mathbf{z}_p^r\right](t')\right)\right\rangle_{\omega[t,\mathbf{z}_p^r]}dt'.\end{aligned} \tag{44}$$

These formulas for the dispersion tensors can be expressed with a simpler notation as

$$\lambda_{ij}^r\left(t;\mathbf{z}_p^r\right)=\frac{1}{\tau_p^{\rm st}}\int_0^t\left\langle\Gamma_{jk}(t,t')R_{ik}(t,\mathbf{y}_p;t',\mathbf{x}_p(t'))\middle|\mathbf{z}_p^r\right\rangle dt', \tag{45}$$

$$\mu_{ij}^r\left(t;\mathbf{z}_p^r\right)=\frac{1}{\tau_p^{\rm st}}\int_0^t\left\langle\dot\Gamma_{jk}(t,t')R_{ik}(t,\mathbf{y}_p;\,t',\mathbf{x}_p(t'))\middle|\mathbf{z}_p^r\right\rangle dt', \tag{46}$$

where $\langle\cdot|\mathbf{z}_p^r\rangle$ is a more compact notation to represent an average on particle sample bridges such that $\mathbf{Z}_p^r(t)=\mathbf{z}_p^r$. Using the equivalence between the notations used in the present section and in Sec. IV A [whereby $\langle.\rangle_{\omega[t,\mathbf{z}_p^r]}\equiv\langle\cdot|\mathbf{z}_p^r\rangle\equiv\langle.\rangle_{(\mathbf{y}_p,\mathbf{v}_p)}$], it is thus seen that the two expressions for λ_{ij}^r and μ_{ij}^r in Eqs. (43) and (44), or in Eqs. (45) and (46), are strictly identical to the ones in Eqs. (20a) and (20b) in Sec. IV A.

The first term on the right-hand side of Eq. (42) represents the conditional mean velocity of the fluid seen at a given location and we can use the decomposition in Eq. (32a) to write it as the sum of the local mean fluid velocity plus the drift term $\mathbf{U}_d$. This gives the expression of the flux closure as

$$\begin{aligned}\frac{1}{\tau_p^{\rm st}}\left\langle U_{s,i}\middle|\mathbf{z}_p^r\right\rangle p^r\;=\;&\frac{1}{\tau_p^{\rm st}}\langle U_{f,i}\rangle(t,\mathbf{y}_p)p^r+\;\kappa_i^r\;p^r\\ &-\frac{\partial\left[\lambda_{ij}^r\;p^r\right]}{\partial y_{p,j}}-\frac{\partial\left[\mu_{ij}^r\;p^r\right]}{\partial V_{p,j}}\end{aligned} \tag{47}$$

with λ_{ij}^r and μ_{ij}^r as in Eqs. (43) and (44) and where

$$\kappa_i^r(t;\mathbf{z}_p^r)=\frac{1}{\tau_p^{\rm st}}U_{d,i}+\frac{\partial\lambda_{ij}^r}{\partial y_{p,j}}+\frac{\partial\mu_{ij}^r}{\partial V_{p,j}}. \tag{48}$$

Furthermore, it can be observed that using Eq. (32b), which basically states that the Lagrangian two-time correlation (the correlation between fluid velocities seen conditioned on a given particle bridge) is equal to the Eulerian two-point and two-time correlation along that trajectory, λ_{ij} and μ_{ij} take the simplified forms

$$\lambda_{ij}\left(t;\mathbf{z}_p^r\right)=\frac{1}{\tau_p^{\rm st}}\int_0^t\left\langle\langle u_{s,i}(t)u_{s,k}(t')|\mathbf{y}_p\rangle\Gamma_{jk}(t,t')\middle|\mathbf{z}_p^r\right\rangle dt', \tag{49a}$$

$$\mu_{ij}\left(t;\mathbf{z}_p^r\right)=\frac{1}{\tau_p^{\rm st}}\int_0^t\left\langle\langle u_{s,i}(t)u_{s,k}(t')|\mathbf{y}_p\rangle\dot\Gamma_{jk}(t,t')\middle|\mathbf{z}_p^r\right\rangle dt'. \tag{49b}$$

The second road towards a closed kinetic PDF equation from a dynamic-PDF one is simpler in that it avoids functional calculus. This approach is still based on the FND relation but formulated as a Gaussian integration by parts between random variables (it is interesting to note that this is the form of the FND relation given in Frisch's book [35]). In that formulation, the FND relation states that for a centered jointly Gaussian vector of random variables $(Z_i)_{i=1,d}$ and for a function $\Phi(\mathbf{Z})$ we have

$$\langle Z_i\,\Phi(\mathbf{Z})\rangle=\langle Z_i\;Z_k\rangle\left\langle\frac{\partial\Phi(\mathbf{Z})}{\partial Z_k}\right\rangle. \tag{50}$$

This second derivation is still based on the decomposition in Eq. (35) by considering first the effect of fluid velocities

$\mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]}(t')$ along sample bridges and then averaging over all possible such sample bridges. Since the particle relaxation time is taken as a constant (i.e., $\tau_p = \tau_p^{\rm st}$), it is seen from the particle governing equations Eqs. (2a) and (2b) that $\mathbf{Z}_p^r = (\mathbf{x}_p, \mathbf{U}_p)$ is obtained as a linear transformation of $\mathbf{U}_s$. Thus, the assumption that $(\mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]}(t'))$ is a Gaussian process implies that the joint process

$$\mathbf{Z}_p^{\omega[t,\mathbf{z}_p^r]}(t') = \big(\mathbf{x}_p^{\omega[t,\mathbf{z}_p^r]}(t'), \mathbf{U}_p^{\omega[t,\mathbf{z}_p^r]}(t'), \mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]}(t')\big), \tag{51}$$

which is the solution of the system

$$\frac{d\mathbf{x}_p^{\omega[t,\mathbf{z}_p^r]}}{dt} = \mathbf{U}_p^{\omega[t,\mathbf{z}_p^r]}, \tag{52a}$$

$$\frac{d\mathbf{U}_p^{\omega[t,\mathbf{z}_p^r]}}{dt} = \frac{1}{\tau_p}\big(\mathbf{U}_f^{\omega[t,\mathbf{z}_p^r]} - \mathbf{U}_p^{\omega[t,\mathbf{z}_p^r]}\big), \tag{52b}$$

forms a jointly Gaussian random vector at any time $t' \leqslant t$. With these clarifications, the simplified version of the FND relation in Eq. (50) can be applied to the fine-grained PDF $\mathcal{P}$ which yields that at time t:

$$\begin{aligned}&\frac{1}{\tau_p^{\rm st}}\big\langle U_{f,i}(t,\mathbf{y}_p)\big|\mathbf{Z}_p^{r,\omega[t,\mathbf{z}_p^r]}(t) = \mathbf{z}_p^r\big\rangle\, p^{r,\omega[t,\mathbf{z}_p^r]}\\ &\quad= \frac{1}{\tau_p^{\rm st}}\big\langle U_{f,i}^{\omega[t,\mathbf{z}_p^r]}\big\rangle\, p^{r,\omega[t,\mathbf{z}_p^r]} - \widetilde{\lambda}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}\frac{\partial p^{r,\omega[t,\mathbf{z}_p^r]}}{\partial y_{p,j}}\\ &\quad\quad- \widetilde{\mu}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}\frac{\partial p^{r,\omega[t,\mathbf{z}_p^r]}}{\partial V_{p,j}},\end{aligned} \tag{53}$$

where $p^{r,\omega[t,\mathbf{z}_p^r]}$ is the PDF of the particle vector $\mathbf{Z}_p^{r,\omega[t,\mathbf{z}_p^r]}$ and where $\widetilde{\lambda}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}$ and $\widetilde{\mu}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}$ are given by

$$\widetilde{\lambda}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}\big(t,\mathbf{z}_p^r\big) = \frac{1}{\tau_p^{\rm st}}\big\langle u_{f,i}(t,\mathbf{y}_p)\, x_{p,j}^{\omega[t,\mathbf{z}_p^r]}(t)\big\rangle, \tag{54a}$$

$$\widetilde{\mu}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}\big(t,\mathbf{z}_p^r\big) = \frac{1}{\tau_p^{\rm st}}\big\langle u_{f,i}(t,\mathbf{y}_p)\, U_{p,j}^{\omega[t,\mathbf{z}_p^r]}(t)\big\rangle. \tag{54b}$$

At the moment, distinct notations are used for $\widetilde{\lambda}_{ij}^r$ and $\widetilde{\mu}_{ij}^r$ compared to λ_{ij}^r and μ_{ij}^r in Eqs. (43) and (44) but a connection with the previous derivation can be worked out. This is done by a complementary development in which, to illustrate the above reasoning with a more physical standpoint, we consider the integration of the particle equations still using the fact that $\tau_p^{\rm st}$ is constant. By integrating Eqs. (52) we get for the particle position $\mathbf{x}_p^{\omega[t,\mathbf{z}_p^r]}(t)$

$$\begin{aligned}x_{p,j}^{\omega[t,\mathbf{z}_p^r]}(t) &= x_{p,j}^{\omega[t,\mathbf{z}_p^r]}(0) + \tau_p^{\rm st}[1 - e^{-t/\tau_p^{\rm st}}]U_{p,j}^{\omega[t,\mathbf{z}_p^r]}(0)\\ &\quad+ \int_0^t [1 - e^{(t'-t)/\tau_p^{\rm st}}]U_{f,j}^{\omega[t,\mathbf{z}_p^r]}(t')\,dt'\end{aligned} \tag{55}$$

and for the particle velocity $\mathbf{U}_p^{\omega[t,\mathbf{z}_p^r]}(t)$

$$\begin{aligned}U_{p,j}^{\omega[t,\mathbf{z}_p^r]}(t) &= U_{p,j}^{\omega[t,\mathbf{z}_p^r]}(0)\, e^{-t/\tau_p^{\rm st}}\\ &\quad+ \frac{e^{-t/\tau_p^{\rm st}}}{\tau_p^{\rm st}}\int_0^t e^{t'/\tau_p^{\rm st}}\, U_{f,j}^{\omega[t,\mathbf{z}_p^r]}(t')\,dt'.\end{aligned} \tag{56}$$

Using the expression of the response function $\Gamma_{jk}(t,t') = \tau_p^{\rm st}[1 - e^{(t'-t)/\tau_p^{\rm st}}]\delta_{jk}$ gives

$$\begin{aligned}\widetilde{\lambda}_{ij}^{r,\omega[t,\mathbf{z}_p^r]} &= \frac{1}{\tau_p^{\rm st}}\int_0^t \langle u_{f,i}(t,\mathbf{y}_p)u_{f,k}^{\omega[t,\mathbf{z}_p^r]}(t')\rangle\Gamma_{jk}(t,t')dt'\\ &= \frac{1}{\tau_p^{\rm st}}\int_0^t R_{ik}(t,\mathbf{y}_p;t',\omega[t,\mathbf{z}_p^r](t'))\Gamma_{jk}(t,t')dt',\end{aligned} \tag{57a}$$

$$\widetilde{\mu}_{ij}^{r,\omega[t,\mathbf{z}_p^r]} = \frac{1}{\tau_p^{\rm st}}\int_0^t R_{ik}(t,\mathbf{y}_p;t',\omega[t,\mathbf{z}_p^r](t'))\dot{\Gamma}_{jk}(t,t')dt'. \tag{57b}$$

Then, integrating with respect to all sample space bridges that arrive at $\mathbf{z}_p^r$ at time t, we recover the closed expression of the flux as

$$\begin{aligned}&\frac{1}{\tau_p^{\rm st}}\big\langle U_{s,i}\big|\mathbf{z}_p^r\big\rangle\, p^r\\ &\quad= \frac{1}{\tau_p^{\rm st}}\big\langle\big\langle U_{f,i}^{\omega[t,\mathbf{z}_p^r]}(t)\big\rangle\big\rangle_{\omega[t,\mathbf{z}_p^r]}p^r - \widetilde{\lambda}_{ij}^r\frac{\partial p^r}{\partial y_{p,j}} - \widetilde{\mu}_{ij}^r\frac{\partial p^r}{\partial V_{p,j}},\end{aligned} \tag{58}$$

where $\widetilde{\lambda}_{ij}^r = \langle\widetilde{\lambda}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}\rangle_{\omega[t,\mathbf{z}_p^r]}$ and $\widetilde{\mu}_{ij}^r = \langle\widetilde{\mu}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}\rangle_{\omega[t,\mathbf{z}_p^r]}$ are the averaged values of $\widetilde{\lambda}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}$ and $\widetilde{\mu}_{ij}^{r,\omega[t,\mathbf{z}_p^r]}$ over all possible particle bridges $\omega[t,\mathbf{z}_p^r]$. From Eqs. (57), it is then obvious that $\widetilde{\lambda}_{ij}^r$ and $\widetilde{\mu}_{ij}^r$ are the same as λ_{ij}^r and μ_{ij}^r given in Eqs. (43) and (44). As in the first derivation, the final step is obtained by expressing the local value of the mean fluid velocity seen at the given location $\mathbf{x}_p(t) = \mathbf{y}_p$ with Eq. (32a) and in switching λ_{ij}^r and μ_{ij}^r into the partial derivatives. Therefore, we retrieve the flux closure as in Eq. (47) and in Eq. (19), with κ_i as in Eq. (48).

A first interesting outcome of these two derivations is to yield that $\kappa_i^r \neq \partial\lambda_{ij}^r/\partial y_{p,j}$ with the difference being precisely the drift velocity [see Eq. (48)] when the assumption that $\partial\mu_{ij}^r/\partial V_{p,j} = 0$ is made. This shows that $\kappa_i^r = \partial\lambda_{ij}^r/\partial y_{p,j}$ for fluid particles or when the so-called well-mixed condition is assumed but a nonzero difference is retrieved in the general case. This is indeed the result of the analysis carried out in Ref. [18].

A second interesting outcome is provided by the second derivation which brings out a new result for the dispersion tensors. Indeed, integrating directly Eqs. (54) over all sample space bridges, the dispersion tensors λ_{ij}^r and μ_{ij}^r can also be written as

$$\lambda_{ij}^r\big(t;\mathbf{z}_p^r\big) = \frac{1}{\tau_p^{\rm st}}\Big\langle\Big\langle u_{f,i}(t,\mathbf{y}_p)x_{p,j}^{r,\omega[t,\mathbf{z}_p^r]}(t)\Big\rangle\Big\rangle_{\omega\left[t,\mathbf{z}_p^r\right]}, \tag{59a}$$

$$\mu_{ij}^r\big(t;\mathbf{z}_p^r\big) = \frac{1}{\tau_p^{\rm st}}\Big\langle\Big\langle u_{f,i}(t,\mathbf{y}_p)U_{p,j}^{r,\omega[t,\mathbf{z}_p^r]}(t)\Big\rangle\Big\rangle_{\omega\left[t,\mathbf{z}_p^r\right]}. \tag{59b}$$

These new formulations open the way for a clearer physical interpretation of the dispersion tensors appearing in the kinetic PDF equation. In physical terms, they express that $\lambda_{ij}^r(t;\mathbf{z}_p^r)$ and $\mu_{ij}^r(t;\mathbf{z}_p^r)$ represent the averaged values, over all possible particle paths arriving at time t at a given point $\mathbf{z}_p^r$ in sample space, of the correlations between the fluid velocity seen along

these paths and the positions and velocities of particles subject to these driving forces.

In homogeneous situations where the statistics of the velocity of the fluid seen are no longer space dependent, we obtain constant values of λ^r_{ij} and μ^r_{ij} which are now simply expressed as the correlations between particle positions and velocities and the variable that is eliminated from the reduced PDF description, that is, the velocity of the fluid seen:

$$\lambda^r_{ij} = \frac{1}{\tau^{\rm st}_p}\langle u_{s,i}(t)\, x_{p,j}(t)\rangle, \tag{60a}$$

$$\mu^r_{ij} = \frac{1}{\tau^{\rm st}_p}\langle u_{s,i}(t)\, U_{p,j}(t)\rangle. \tag{60b}$$

This was the form derived in Ref. [4, Sec. 7.5.7] but where the derivation was made only for such homogeneous situations, whereas it has now been extended to the general case with the expressions in Eqs. (59a) and (59b). These formulations of the dispersion tensors will be shown to be useful in Sec. VIII B.

A third interesting outcome is that these two derivations are helpful to bring out the difference between two ways to address the closure issue at the level of the kinetic PDF equation. In the first derivation, the standpoint is to fix a value of the reduced particle state vector $\mathbf{Z}^r_p$ and then try to work out where the trajectories of the velocity of the fluid seen come from. Note that this implies a "time-backward point of view" in the kinetic PDF approach that compounds the already hybrid Lagrangian-Eulerian description (since $(\mathbf{x}_p,\mathbf{U}_p)$ are simulated in a Lagrangian formulation while $(\mathbf{U}_f(t;\mathbf{x}))$ is treated as a Eulerian external field). In the second derivation, the standpoint is more to consider Gaussian copies of the velocities of the fluid along trajectories arriving at a given point $\mathbf{z}^r_p$ and analyze how particle positions and velocities respond to that fluid [see Eqs. (59a) and (59b)]. Though this second standpoint is already one step towards a formulation that is typical of dynamic PDF models, it is clear that present attempts at closing the kinetic PDF equation implies to mix a time-forward approach [for $(\mathbf{x}_p,\mathbf{U}_p)$] with a time-backward one (for $\mathbf{U}_s$). This point is at the core of the difficulties encountered by the kinetic PDF equation and will resurface in Sec. VII B.

From these developments, a threefold conclusion can be drawn:

(a) it is demonstrated that, under the same assumption of a Gaussian process for the conditional velocity of the fluid seen by particles, the marginal of the dynamic PDF $p^r = \int p d\mathbf{V}_s$ satisfies the same equation as the kinetic PDF one. In other words, it has been shown that, under the kinetic assumption of Gaussian fields, the kinetic PDF model may be recover from the dynamic PDF model or, using the notation introduced in Sec. IV C, that we have $p^{\rm dm}(t;\mathbf{y}_p,\mathbf{V}_p) = p^{\rm km}(t;\mathbf{y}_p,\mathbf{V}_p)$;

(b) the derivation of a kinetic PDF equation starting from a dynamic-PDF one leads to new expressions for the dispersion tensors in Eqs. (59a) and (59b) that allow new physical interpretations. A noteworthy result is that the dispersion tensors appearing in the kinetic PDF equation correspond to the *correlations between the variables kept in the particle state vector* [here $(\mathbf{x}_p,\mathbf{U}_p)$] *with the one treated as an external noise and eliminated* (here $\mathbf{U}_s$).

(c) Using, for example, Eqs. (49a) and (49b), an important aspect is also worth emphasizing: A dynamic PDF model yields directly *nonlocal closures* of the dispersion tensors. Indeed, the correlation of the velocity of the fluid seen is a result of a dynamic PDF description which already contains spatial effects in the present model used to simulate $\mathbf{U}_s(t)$ [cf. Eq. (25c)]. In a recent study [19], it was shown that such nonlocal closures are needed in kinetic PDF descriptions. However, such effects must be input, which was done by assuming a backward Gaussian spread of particle positions in Ref. [19]. However, it can be seen that this induces a lack of consistency between the exact treatment of the particle convection equation [in Eq. (2a)] used in the very derivation of the kinetic PDF equation and this additional assumption of a Gaussian spread of the particle location bridge $(\mathbf{x}_p(t'))_{t'\in[0,t]}|\mathbf{Z}^r_p(t) = \mathbf{z}^r_p$ to input spatially dependent expressions of the correlations of the velocity of the fluid seen $\langle u_{s,i}(t)\, u_{s,k}(t')|\mathbf{z}^r_p\rangle$. On the other hand, these space-dependent quantities are handled consistently at the dynamic-PDF level of description.

The correspondence between the two PDF levels of description has been studied here for the special case of a Gaussian process for the fluid velocity seen. When the resulting equations are proposed as models for the general case, the status of the different PDF descriptions must be analyzed. This is done in the next two sections.

VI. ANALYSIS OF THE DYNAMIC PDF MODEL AS A STAND-ALONE PDF DESCRIPTION

The dynamic PDF model presented in Sec. IV B can be addressed from a trajectory point of view, in which case it consists in the SDEs in Eqs. (25) plus an initial condition for $\mathbf{Z}_p(t=0)$, or from a PDF point of view, in which case it consists in the evolution equation for its transition PDF in Eq. (26) plus the initial PDF $p(0;\mathbf{z}_p(0))$. In the evolution equations for the particle state vector in Eqs. (25), the external noise is represented by the increments of the Wiener process in Eq. (25c). Since these increments are independent, it follows that the particle state vector $\mathbf{Z}_p$ is a Markov process. From the theory recalled in Sec. III B, it is then clear that such a dynamic-PDF description allows the complete law of the stochastic process to be reconstructed. Thus, criterion (C1) of Sec. III E is fulfilled.

To assess criterion (C2), we need to study whether the FPE is well posed. To be more precise, well-posedness means here that the Cauchy problem is satisfied (there is a solution to the initial-value problem) for any initial condition and also that the solutions of the FPE corresponding to different initial conditions tend towards the same result as $t \to \infty$. This is indeed an essential characteristic to respect as it ensures that a solution of the modeled equation reflects the modeled terms in the evolution equation, and thus the model itself, rather than the choice of an initial condition.

Detailed studies of the FPE can be found in dedicated textbooks [14,15,29] and also in the mathematical literature (see, for example, Ref. [39]) where the criterion (C2) has been positively assessed for FPEs. For our present purpose, it is, however, important to bring out the main lines of the analysis of the approach of a limit solution. The analysis is carried

out in terms of the "relative entropy" between two solutions $p_1(t;\mathbf{z}_p)$ and $p_2(t;\mathbf{z}_p)$ of a model PDF equation corresponding to two different initial conditions, and which is defined as

$$H(p_2|p_1) = -\int \ln\left(\frac{p_2}{p_1}\right) p_1(t;\mathbf{z}_p) d\mathbf{z}_p. \tag{61}$$

The functional $H(p_2|p_1)$ is always positive and is known as the "information gain" or "Kullback-Leibler divergence" in information theory where it quantifies the information lost when p_2 is used instead of p_1 to describe the system represented by $\mathbf{Z}_p$. It acts as a pseudomeasure to control the distance between two PDFs since we have [39,40]

$$\frac{1}{2}||p_1 - p_2||^2_{L^1} \leqslant H(p_2|p_1). \tag{62}$$

As a result, the relative entropy $H(p_2|p_1)$ serves as a Lyapunov functional to control the march towards a unique limit solution. In the following, we use the essential result (see the detailed derivation in Sec. 6.1 in Ref. [29]) that for an evolution equation of the "FPE form" and written as

$$\frac{\partial p}{\partial t} = -\frac{\partial [A_i(t;\mathbf{z}_p)p]}{\partial z_{p,i}} + \frac{\partial^2 [D_{ij}p]}{\partial z_{p,i}\partial z_{p,j}}, \tag{63}$$

where the matrix $\mathbf{D} = (D_{ij})_{i,j=1,d}$ can be any second-order tensor, we have that

$$\dot{H}(p_2|p_1) = -\int p_1 D_{ij} \frac{\partial \ln(R)}{\partial z_{p,i}} \frac{\partial \ln(R)}{\partial z_{p,j}} d\mathbf{z}_p, \tag{64}$$

with $R = p_1/p_2$. For a genuine FPE, the second-order diffusion matrix $(D_{ij})_{i,j=1,d}$ is positive definite which entails that $\dot{H}(p_2|p_1) \leqslant 0$ and, thus, that a unique solution is obtained [since $H(p_2|p_1) \to 0$ as $t \to \infty$]. This is the expected result as there should be no information gain or loss in the long-time limit when we switch between two solutions corresponding to two different initial conditions.

In the case of the dynamic PDF model represented by the FPE in Eq. (26), the general diffusion matrix $(D_{ij})_{i,j=1,d}$ corresponding to $\mathbf{Z}_p$ is not strictly positive definite [it is actually degenerate for the components corresponding to the variables $\mathbf{x}_p$ and $\mathbf{U}_p$ since there are no white-noise explicit terms in Eqs. (25a) and (25b)] but the same conclusion applies (note that the situation is similar to the Kramers equation studied with the same approach in Sec. 6.1 in Ref. [29]). This results guarantees that for the dynamic PDF model two solutions corresponding to two different initial conditions tend towards the same limit solution and, consequently, that this limit solution manifest properties related to the model and not to the choice of an initial PDF. In that sense, the FPE is well posed and the criterion (C2) is fulfilled.

From these results, we conclude that dynamic PDF models constitute complete PDF stand-alone descriptions of particle dynamics in turbulent fluid flows.

VII. ANALYSIS OF THE KINETIC PDF MODEL AS A STAND-ALONE PDF DESCRIPTION

As presented in Sec. IV A, the kinetic PDF model is only formulated in sample space where it consists of Eq. (14) with Eq. (19). The status of this PDF description is now addressed.

A. A non-Markovian particle state vector

The particle state vector is reduced to $\mathbf{Z}^r_p = (\mathbf{x}_p, \mathbf{U}_p)$ and the velocity of the fluid seen $\mathbf{U}_s(t) = \mathbf{U}_f(t;\mathbf{x}_p(t))$ is an external variable. To assess the status of that particle state vector, the remark made in Sec. III B about the nature of the "information known at the present time" is of direct application. Indeed, if the whole *instantaneous fluid velocity field* $\mathbf{U}_f(t;\mathbf{x})$ is known at each instant, then $\mathbf{U}_s(t)$ is completely determined by the knowledge of the particle positions $\mathbf{x}_p(t)$ at the present time t, which means that the particle evolution equations Eqs. (2) are closed. This corresponds to the situation where the fluid flow is obtained from a DNS and, in that case, $\mathbf{Z}^r_p$ is a Markov process. However, in the general situation where only statistics of the fluid velocity field are available (such as the mean values, time and space correlations, etc.), then clearly $\mathbf{U}_s(t)$ cannot be determined by the sole knowledge of the particle position and velocity. This is due to the nonzero time correlation of the velocity of the fluid seen which is treated as an external noise (this corresponds exactly to the example put forward in Sec. III D).

From the discussions in Sec. III C, it is seen that the solution of such a kinetic PDF equation does not allow the complete law of the stochastic process to be reconstructed. In that sense, the criterion (C1) is not respected and it is concluded that the kinetic PDF approach is an incomplete stand-alone PDF description of particle dynamics in turbulent fluid flows.

B. An ill-posed PDF equation

If the main objective of a PDF approach is to derive one-point statistical moments, failure to meet the criterion (C1) is not necessarily a major obstacle. Far more relevant are the issues related to the criterion (C2). To analyze these aspects for the kinetic PDF model, Eq. (14) and Eq. (19) are combined into the compact form

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial Z^r_l}\left[A^{\mathrm{KE}}_l p\right] + \frac{1}{2}\frac{\partial^2}{\partial Z^r_l \partial Z^r_m}\left[B^{\mathrm{KE}}_{\mathrm{lm}} p\right], \tag{65}$$

where the components of the drift vector $(A^{\mathrm{KE}}_l)_{l=1,6}$ are

$$A^{\mathrm{KE}}_l = \begin{cases} V_{p,l} & l = 1,3, \\ F_{\mathrm{ext},l-3} + \dfrac{\langle U_{f,l-3}\rangle - V_{p,l-3}}{\tau^{\mathrm{st}}_p} + \kappa_{l-3} & l = 4,6. \end{cases} \tag{66}$$

and where the symmetrical matrix in the second-order derivative $(B^{\mathrm{KE}}_{\mathrm{lm}})$ is (using a bloc notation with $i,j = 1,3$)

$$\mathbf{B}^{\mathrm{KE}} = \begin{pmatrix} 0 & | & (\lambda_{ij}) \\ -- & & --- \\ (\lambda_{ji}) & | & (\mu_{ij}) + (\mu_{ji}) \end{pmatrix}. \tag{67}$$

At first sight, Eq. (65) looks like a classical convection-diffusion equation. However, it is straightforward to show that the symmetrical matrix $(B^{\mathrm{KE}}_{\mathrm{lm}})$ is actually negative definite. Indeed, the determinant of the matrix is $\det(\mathbf{B}^{\mathrm{KE}}) = -(\det(\boldsymbol{\lambda}))^2 < 0$ and, consequently, the matrix $\mathbf{B}^{\mathrm{KE}}$ has always at least one negative eigenvalue. This point was put forward in Ref. [13] where it was qualitatively connected to an "antidiffusive" behavior and to the non-Markovian nature of the reduced state vector $\mathbf{Z}^r_p$. These aspects are taken up here and further investigated.

For the analysis of the well-posed nature of Eq. (65), it suffices to consider the 1D case where $\mathbf{Z}_p^r = (x_p, U_p)$ and constant values of $\lambda = \lambda_{11}$ and $\mu = \mu_{11}$ (physically speaking, this corresponds for instance to the isotropic stationary turbulence case). Note that $\kappa = \kappa_1$ is either zero when drift velocities are neglected or enters the drift term where it does not play any significant role in the resulting properties. Thus, $\mathbf{B}^{\mathrm{KE}}$ is now the (2×2) matrix

$$\mathbf{B}^{\mathrm{KE}} = \begin{pmatrix} 0 & \lambda \\ \lambda & 2\mu \end{pmatrix}, \tag{68}$$

whose two eigenvalues e_{v1} and e_{v2} are

$$e_{v1} = \mu + \sqrt{\mu^2 + \lambda^2} \geqslant 0, \tag{69a}$$

$$e_{v2} = \mu - \sqrt{\mu^2 + \lambda^2} \leqslant 0. \tag{69b}$$

We set $\mathbf{x}_p = (x_p, 0)$ and $\mathbf{U}_p = (0, U_p)$. The two eigenvectors, noted here $\mathbf{Z_1}$ and $\mathbf{Z_2}$, are

$$\mathbf{Z_1} = \frac{1}{\sqrt{1 + \lambda^2/e_{v1}^2}} \left(\frac{\lambda}{e_{v1}} \mathbf{x}_p + \mathbf{U}_p \right), \tag{70a}$$

$$\mathbf{Z_2} = \frac{1}{\sqrt{1 + \lambda^2/e_{v2}^2}} \left(\frac{\lambda}{e_{v2}} \mathbf{x}_p + \mathbf{U}_p \right). \tag{70b}$$

This suggests introducing the two random variables Z_1 and Z_2 obtained by the combination of x_p and U_p as

$$Z_1 = \frac{1}{\sqrt{1 + \lambda^2/e_{v1}^2}} \left(\frac{\lambda}{e_{v1}} x_p + U_p \right), \tag{71a}$$

$$Z_2 = \frac{1}{\sqrt{1 + \lambda^2/e_{v2}^2}} \left(\frac{\lambda}{e_{v2}} x_p + U_p \right). \tag{71b}$$

Using these transformed variables, the kinetic PDF equation Eq. (65) becomes for $\widetilde{p}(t; z_1, z_2)$

$$\frac{\partial \widetilde{p}}{\partial t} = -\frac{\partial (\widetilde{A}_1 \widetilde{p})}{\partial z_1} - \frac{\partial (\widetilde{A}_2 \widetilde{p})}{\partial z_2} + \frac{1}{2} e_{v1} \frac{\partial^2 \widetilde{p}}{\partial z_1^2} - \frac{1}{2} |e_{v2}| \frac{\partial^2 \widetilde{p}}{\partial z_2^2}, \tag{72}$$

where $\widetilde{A}_1$ and $\widetilde{A}_2$ are the transformed drift coefficients (see the transformation rules for instance in Ref. [29]) and where the second-order derivatives have been re-expressed to bring out the antidiffusion coefficient written as $-|e_{v2}|$. A first remark is that the usual terminology of "dispersion tensors" used for λ_{ij} and μ_{ij} is thus inappropriate, though, for the sake of convenience, these names will be retained in the rest of the paper (this restriction is indicated by keeping the apostrophes).

At this stage, the question of the physical origin of such an antidiffusive behavior can be raised. The answer to that question is provided by using that $\lambda = \langle x_p\, U_s \rangle / \tau_p^{\mathrm{st}}$ and $\mu = \langle U_p\, U_s \rangle / \tau_p^{\mathrm{st}}$ and by considering the correlations of the transformed variables Z_1 and Z_2 with the velocity of the fluid seen. With Eqs. (69) and Eqs. (71), we get

$$\frac{1}{\tau_p^{\mathrm{st}}} \langle Z_1\, U_s \rangle = \frac{1}{\sqrt{1 + \lambda^2/e_{v1}^2}} \sqrt{\mu^2 + \lambda^2} > 0, \tag{73a}$$

$$\frac{1}{\tau_p^{\mathrm{st}}} \langle Z_2\, U_s \rangle = \frac{-1}{\sqrt{1 + \lambda^2/e_{v2}^2}} \sqrt{\mu^2 + \lambda^2} < 0. \tag{73b}$$

In the kinetic description, the velocity of the fluid seen is eliminated but since (Z_1, Z_2, U_s) forms a jointly Gaussian process with Z_2 negatively correlated with U_s, the effect of the elimination appears as an "antidiffusion" for Z_2.

To bring further physical insight into that point and in anticipation of the more detailed developments presented in Sec. VIII B, it is interesting to make the following remark: If we could regard λ as a small parameter (actually, λ/μ is equal to the time scale of the eliminated variable, that is, the velocity of the fluid seen, see the demonstration in Sec. VIII B), then by taking the limit $\lambda \to 0$ we would have $\langle Z_2\, U_s \rangle / \tau_p^{\mathrm{st}} \simeq -\lambda/2 \ll 1$ while $\langle Z_1\, U_s \rangle / \tau_p^{\mathrm{st}} \simeq \mu$ remains finite so, in the limit of vanishing λ, this antidiffusive behavior disappears. Note that μ has been treated as a positive coefficient. Indeed, a simple equilibrium argument from the particle momentum equation and the drag force term, cf. Eq. (2b), shows that $\tau_p^{\mathrm{st}} \mu = \langle U_p\, U_s \rangle \simeq \langle U_p^2 \rangle > 0$ (similarly, λ is also positive). These arguments are pursued in Sec. VIII B.

To analyze the status of the kinetic PDF equation, Eq. (65), we consider the equation satisfied by the marginal PDF $\widetilde{p}_r(t; z_2) = \int \widetilde{p}(t; z_1, z_2) dz_1$ which is obtained from Eq. (72)

$$\begin{cases} \dfrac{\partial \widetilde{p}_r}{\partial t} = -\dfrac{\partial (\widetilde{A}_2^r \widetilde{p}_r)}{\partial z_2} - \dfrac{1}{2} |e_{v2}| \dfrac{\partial^2 \widetilde{p}_r}{\partial z_2^2}, \\ \widetilde{p}_r(0; z_2) = \phi(z_2), \end{cases} \tag{74}$$

where $\phi(z_2)$ is the initial condition for the time-forward equation for $\widetilde{p}_r(t; Z_2)$. We then rewrite Eq. (74) as

$$\frac{\partial \widetilde{p}_r}{\partial t} + \widetilde{A}_2^r \frac{\partial \widetilde{p}_r}{\partial z_2} + \frac{1}{2} |e_{v2}| \frac{\partial^2 \widetilde{p}_r}{\partial z_2^2} + \frac{\partial \widetilde{A}_2^r}{\partial z_2} \widetilde{p}_r = 0, \tag{75}$$

where we can use the celebrated Feynman-Kac formula. This is done by introducing a terminal condition $\widetilde{p}_r(T; z_2) = \Psi(z_2)$ at a time T, which translates the fact that this equation is actually associated with the backward Kolmogorov equation of a stochastic diffusion process [15,25]. In that sense, it be can already be guessed that present difficulties with Eq. (74) come from the attempt to treat as a forward equation what is basically a backward one. The application of the Feynman-Kac formula [41, Sec. 5.7] shows that the initial condition associated to Eq. (74) cannot be any function but should be of the special following form:

$$\widetilde{p}_r(0; z_2) = \langle I[T; X] \Psi(X(T)) | (X(0) = z_2) \rangle, \tag{76}$$

with $I[T; X]$ given by

$$I[T; X] = \exp \left[\int_0^T \frac{\partial \widetilde{A}_2^r}{\partial z_2}(s, X(s)) ds \right] \tag{77}$$

and where X is the properly defined stochastic diffusion process whose trajectory equations are

$$dX = \widetilde{A}_2^r(X)\, dt + \sqrt{|e_{v2}|}\, dW. \tag{78}$$

Basically, this says that Eq. (74) can only be solved if the initial condition $\widetilde{p}_r(0; z_2)$ can be written as the "end solution" (at a given later time T) of the stochastic process X defined in Eq. (78). Clearly, the Cauchy problem is not respected,

apart from some very special classes of initial conditions, which demonstrates that Eq. (74) is an unstable and ill-posed equation. This is actually a typical example of such ill-posed problems (see, for instance, the survey in Ref. [42]). Since this is obtained as the marginal of the initial kinetic PDF equation, these shortcomings are also associated with the original PDF equation. Consequently, the criterion (C2) is not met and it is concluded that the kinetic PDF equation is ill posed and cannot be solved (apart from some special cases).

The ill-posed nature of the kinetic PDF equation has been demonstrated in the general case but can be exemplified in the specific case considered here. Indeed, using Eqs. (71), the transformed drift term $\widetilde{A}_2^r$ is given by

$$\widetilde{A}_2^r(z_2) = -\alpha_{Z_2}\left(\frac{\lambda}{|e_{v2}|}+\frac{1}{\tau_p^{\rm st}}\right) \times\left\{\alpha_{Z_1}|e_{v2}|z_2+\alpha_{Z_1}e_{v1}\langle Z_1|z_2\rangle\right\}, \tag{79}$$

where α_{Z_1} and α_{Z_2} stand for

$$\alpha_{Z_1}=\frac{1}{\sqrt{1+\lambda^2/e_{v1}^2}},\quad \alpha_{Z_2}=\frac{1}{\sqrt{1+\lambda^2/e_{v2}^2}}. \tag{80}$$

For centered Gaussian joint random variables, we can use that $\langle Z_1|z_2\rangle = \langle Z_1Z_2\rangle/\langle Z_2^2\rangle z_2$, from which $\widetilde{A}_2^r$ becomes

$$\widetilde{A}_2^r(z_2) = -\alpha_{Z_2}\left(\frac{\lambda}{|e_{v2}|}+\frac{1}{\tau_p^{\rm st}}\right) \times\left\{\alpha_{Z_1}|e_{v2}|+\alpha_{Z_1}e_{v1}\frac{\langle Z_1Z_2\rangle}{\langle Z_2^2\rangle}\right\}z_2, \tag{81}$$

In the long-time limit, it is clear from Eqs. (71) that $\langle Z_1Z_2\rangle$ and $\langle Z_2\rangle$ are governed by $\langle x_p^2\rangle$ (in the diffusive regime $\langle x_p^2\rangle$ increases linearly in time whereas $\langle U_p^2\rangle$ and $\langle x_pU_p\rangle$ tend to constant values). Then, simple calculations show that $\widetilde{A}_2^r=0$. This means that, in the present situation, the PDF equation for the marginal $\widetilde{p}_r(t;z_2)$ in Eq. (74) becomes the "anti-heat-diffusion" equation

$$\begin{cases}\dfrac{\partial\widetilde{p}_r}{\partial t}=-\dfrac{1}{2}|e_{v2}|\dfrac{\partial^2\widetilde{p}_r}{\partial z_2^2},\\ \widetilde{p}_r(0;z_2)=\phi(z_2),\end{cases} \tag{82}$$

which is one of the canonical examples of an ill-based equation, see Ref. [42, p. 333]. More precisely, this equation can only be "solved" on a finite-time interval, say, $[0,T]$, and when the initial condition is of the special form where it is expressed as

$$\widetilde{p}_r(0;z_2)=\int g(y)\frac{1}{\sqrt{2\pi|e_{v2}|T}}\exp\left[-\frac{(z_2-y)^2}{2|e_{v2}|T}\right]dy, \tag{83}$$

with $g(y)$ a PDF so $g(y)\geqslant 0$ with $\int g(y)\,dy=1$. In particular, we have the T-dependent bound

$$\widetilde{p}_r(0;z_2)\leqslant\frac{1}{\sqrt{2\pi|e_{v2}|T}},$$

which implies that the initial condition should be very flat if one wants a solution that exists on a large time interval. In particular, letting $T\to+\infty$, we deduce that the only solution defined for any time $t\in[0,+\infty[$ is the null solution. Equation (83) illustrates that the "initial condition" corresponds in fact to an "end condition" as we have then

$$\widetilde{p}_r(t;z_2)=\int g(y)\frac{1}{\sqrt{2\pi|e_{v2}|(T-t)}} \times\exp\left[-\frac{(z_2-y)^2}{2|e_{v2}|(T-t)}\right]dy, \tag{84}$$

implying that $\widetilde{p}_r(T;z_2)=g(z_2)$ which is the very function we had to give ourselves through the expression of the initial condition. For example, if we expect the "antidiffusion" to lead to a PDF that is a Dirac function at the "final time" T, then $g(y)=\delta(y-z_0)$ with z_0 a constant value in sample space, which shows that the initial condition becomes $\widetilde{p}_r(0;z_2)=1/\sqrt{2\pi|e_{v2}|T}\exp[-\frac{(z_2-z_0)^2}{2|e_{v2}|T}]$ and, therefore, must be a Gaussian PDF. The kinetic PDF equation is then of little interest as we are forced to remain in the Gaussian context and, obviously, writing an evolution equation is not needed when we already know the form of the PDF.

Concerning this problem, there is a strong similarity with a classical issue in PDF scalar modeling in single-phase turbulent flows (see the detailed account in Ref. [2, Sec. 12.7.4] for the case of the conserved scalar PDF equation in isotropic turbulence). It is shown in Ref. [2] that the assumption of Gaussian scalar fields leads to an exact scalar-PDF equation that has an "antidiffusive" nature (see Eq. (12.347) in Ref. [2, Sec. 12.7.4]). Then, the conclusion is also that, to avoid solutions from blowing up in finite time, one has to consider only initial PDFs that are Gaussian [2] and, thus, that such formulations are flawed as general models.

The ill-based nature of Eq. (74) is further demonstrated by considering the relative entropy $H(\widetilde{p}_{r,2}|\widetilde{p}_{r,1})$ between two solutions $\widetilde{p}_{r,1}$ and $\widetilde{p}_{r,2}$ corresponding to two different initial conditions. Since Eq. (74) is of the same form as Eq. (63), the reasoning presented in Sec. VI can be directly applied to give that

$$\dot{H}(\widetilde{p}_{r,2}|\widetilde{p}_{r,1})=|e_{v2}|\int\widetilde{p}_{r,1}(t;z_2)\left[\frac{d\ln(R)}{dz_2}\right]^2dz_2>0 \tag{85}$$

still with $R=\widetilde{p}_{r,1}/\widetilde{p}_{r,2}$. This shows that the functional $H(\widetilde{p}_{r,2}|\widetilde{p}_{r,1})$ measuring the "distance" between two solutions of the same equation is increasing and, thus, that solutions are diverging. Note that this remains true even if we consider two Gaussian distributions (with different variances) as initial conditions.

The failure of the kinetic PDF equation to respect basic criteria for well-posedness may come as some surprise since this equation is an exact result for Gaussian processes. Yet, present results reveal that the kinetic PDF equation can indeed be solved (without solutions blowing up in finite time) but provided that we consider only very special initial conditions, for example, with a Gaussian form. Furthermore, solutions reflect the choice of an initial condition rather than the true properties of the modeled equation. For the complete kinetic PDF equation in Eq. (65) or in Eq. (72), this behavior can be sometimes hidden by the effects of the positive eigenvalue and specific (or "lucky") choices of initial conditions but they remain potentially present.

At this stage, a remark is in order. It has been recognized in recent years that, even in statistically homogeneous turbulent flows, particles can correlate with special fluid structures embedded in the flow and, as a result, concentrate in some specific areas. This is known as the "particle preferential concentration effect" in turbulent flows [6,43,44]. This is a physical effect due to the existence of so-called fluid coherent structures that can "capture" particles, which leads to possible local concentration build-ups for particles whose relaxation time scales are tuned to the characteristic lifetimes of these structures. Yet, this physical effect must not be confused with the antidiffusive behavior exhibited by a model. For example, the negative eigenvalue of the kinetic PDF equation can produce particle concentration build-ups. However, this is the result of the ill-based nature of the formulation and in no way can it be said that this represents the physical effect of particle preferential concentration effect.

In conclusion, the results put forward in this section invalidates the kinetic PDF approach as an acceptable PDF description of disperse two-phase flows.

VIII. STOCHASTIC MODELING PRINCIPLES

A. The key issue of the particle state vector

Given the well-posed nature of the dynamic PDF model and the ill-based formulation of the kinetic PDF model, it may be believed that the difference is due solely to the addition of an extra variable in the particle state vector. However, it is worth emphasizing that it is the combination of a relevant choice of this state vector with fast-variable elimination that really paves the way towards physically meaningful and mathematically correct PDF formulations. A second misleading belief is that differences in the model expressions are due to the approach used to obtain the closed fluxes: Langevin models versus the Furutsu-Novikov-Donsker relation. This is not so and the purpose of the present section is to show how this interplay between physical choices and mathematical guidelines is working.

To demonstrate that including the velocity of the fluid seen is not sufficient by itself or that the FND relation is not to blame, we address the situation where $\mathbf{Z}_p = (\mathbf{x}_p, \mathbf{U}_p, \mathbf{U}_s)$ is influenced by a general Gaussian noise.

B. The Furutsu-Novikov approach for the velocity of the fluid seen

For this purpose, a 1D formulation with no mean velocity field and no external forces is sufficient to concentrate on the treatment of "external noises." We consider the particle state vector $\mathbf{Z}_p = (x_p, U_p, U_s)$ whose trajectories are given by the following equations:

$$\frac{dx_p}{dt} = U_p, \tag{86a}$$

$$\frac{dU_p}{dt} = \frac{U_s - U_p}{\tau_p}, \tag{86b}$$

$$\frac{dU_s}{dt} = -\frac{U_s}{T_L} + \xi_s, \tag{86c}$$

where the relaxation time scales τ_p and T_L are taken as constant. As an example, we can take $\tau_p = \tau_p^{\rm st}$ but we retain the more general expression to indicate that any constant value can be used in the following. Compared to the SDEs in Eqs. (25), it is seen that the white-noise term in Eq. (25c) has been replaced by a colored noise ξ_s and U_s is thus differentiable. This explains that the trajectory equations in Eqs. (86) are written in a differentiable form: They correspond to Eqs. (23) in Sec. IV B with $\Theta_s(t) = -U_s/T_L + \xi_s(t)$, a decomposition in line with the one used in Eq. (24). For ξ_s, we consider a stationary Gaussian process with a nonzero correlation time scale, which is easily simulated as an OU process

$$d\xi_s = -\frac{\xi_s}{\tau}\,dt + \sqrt{K}\,dW, \tag{87}$$

where τ is the time scale of ξ_s and K a constant (equal to $K = 2\langle\xi_s^2\rangle/\tau$ from the classical fluctuation-dissipation theorem [14]). The autocorrelation is an exponential function $\langle\xi_s(t)\xi_s(t')\rangle = \langle\xi_s^2\rangle \exp(-|t-t'|/\tau)$ and is not δ correlated when $\tau \neq 0$.

From Eqs. (86) it is clear that, although the fluid velocity seen U_s has been included, the particle state vector $\mathbf{Z}_p$ does not constitute a Markov process. Thus, this formulation is an incomplete stand-alone PDF description [note that, of course, a Markov process would be retrieved by considering the extended state vector (x_p, U_p, U_s, ξ_s)]. Conversely, the PDF equation for $p(t; y_p, V_p, V_s)$ is open and is [cf. Eq. (22)]

$$\frac{\partial p}{\partial t} + \frac{\partial\big[V_p\, p\big]}{\partial y_p} = -\frac{\partial}{\partial V_p}\left[\left(\frac{V_s - V_p}{\tau_p}\right) p\right] + \frac{\partial}{\partial V_s}\left[\frac{V_s}{T_L} p\right] - \frac{\partial}{\partial V_s}\big[\langle \xi_s | (y_p, V_p, V_s)\rangle\, p\big]. \tag{88}$$

Since the external noise ξ_s to the system represented by $\mathbf{Z}_p$ is Gaussian, we can work out an exact closed PDF equation by applying the same FND relation used in Secs. IV A and IV B. This is done, for example, with the formulation in Eq. (36) or with Eq. (50) for the jointly Gaussian process $\mathbf{Z}_p = (x_p, U_p, U_s, \xi_s)$, which yields

$$\langle \xi_s \,|\, (y_p, V_p, V_s)\rangle\, p = -\Lambda_{y_p}\frac{\partial p}{\partial y_p} - \Lambda_{U_p}\frac{\partial p}{\partial V_p} - \Lambda_{U_s}\frac{\partial p}{\partial V_s}, \tag{89}$$

where the coefficients Λ_{y_p}, Λ_{U_p}, and Λ_{U_s} are given below. For the moment, we are concerned with the structure of the resulting PDF equation obtained by inserting the above closure flux in Eq. (88), which gives a closed PDF formulation written in a compact form as

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial Z_{p,l}}[\widetilde{A}_l\, p] + \frac{1}{2}\frac{\partial^2}{\partial Z_{p,l}\partial Z_{p,m}}\big[\widetilde{B}_{\rm lm}\, p\big], \tag{90}$$

where the drift vector $(\widetilde{A}_l)_{l=1,3}$ follows directly from Eq. (88). From the analysis in Sec. VII B, we know that the well-posed property of this PDF equation is governed by the second-order tensor which is a (3×3) matrix

$$\widetilde{\mathbf{B}} = \begin{pmatrix} 0 & 0 & \Lambda_{y_p} \\ 0 & 0 & \Lambda_{U_p} \\ \Lambda_{y_p} & \Lambda_{U_p} & 2\,\Lambda_{U_s} \end{pmatrix}. \tag{91}$$

Its characteristic polynomial, $\mathbb{P}_{\widetilde{\mathbf{B}}}(x) = det(x\mathbb{1} - \widetilde{\mathbf{B}})$, is

$$\mathbb{P}_{\widetilde{\mathbf{B}}}(x) = x\big(x^2 - 2x\Lambda_{U_s} - \Lambda_{U_p}^2 - \Lambda_{x_p}^2\big), \tag{92}$$

from which it follows that the eigenvalues are

$$e_{v1} = 0, \tag{93a}$$

$$e_{v2} = \Lambda_{U_s} - \sqrt{\Lambda_{U_s}^2 + \Lambda_{U_p}^2 + \Lambda_{x_p}^2}, \tag{93b}$$

$$e_{v3} = \Lambda_{U_s} + \sqrt{\Lambda_{U_s}^2 + \Lambda_{U_p}^2 + \Lambda_{x_p}^2}, \tag{93c}$$

and, thus, that there is always a negative eigenvalue (here e_{v2}). The analysis of Sec. VII B is directly applicable and shows that Eq. (90) is ill posed.

Therefore, the sole inclusion of the velocity of the fluid seen in the particle state vector does not change the situation for an external colored Gaussian noise and we end up again with an incomplete and ill-based PDF formulation. The way out of the this dead end is provided by physical insight. We use the important result obtained in Sec. V [see point (b) in the conclusion of that section] that the coefficients in the flux closure formula represent the correlations between the variables kept in the state vector and the noise that is eliminated. Thus, for our example, we have

$$\Lambda_{U_s} = \langle U_s\, \xi_s\rangle, \quad \Lambda_{U_p} = \langle U_p\, \xi_s\rangle, \quad \Lambda_{x_p} = \langle x_p\, \xi_s\rangle. \tag{94}$$

These correlations are derived from Eqs. (86) and Eq. (87) by a straightforward application of stochastic calculus to yield

$$\Lambda_{U_s} = \frac{\tau\, T_L}{\tau + T_L}\big\langle \xi_s^2\big\rangle, \tag{95a}$$

$$\Lambda_{U_p} = \frac{\tau}{\tau + \tau_p}\langle U_s\, \xi_s\rangle = \frac{\tau^2\, T_L}{(\tau + T_L)(\tau + \tau_p)}\big\langle \xi_s^2\big\rangle, \tag{95b}$$

$$\Lambda_{x_p} = \tau\langle U_p\, \xi_s\rangle = \frac{\tau^3\, T_L}{(\tau + T_L)(\tau + \tau_p)}\big\langle \xi_s^2\big\rangle. \tag{95c}$$

This reveals that the "dispersion coefficients," which appear in the PDF equation as the result of the elimination of the external noise ξ_s, *scale as the successive powers in the time scale of that noise*.

Next, the physically meaningful step is to search for a rapidly varying noise having the same total energy [4,14]. This means that τ becomes small (with respect to T_L) but that the total energy, $\tau\langle \xi_s^2\rangle$, remains constant. Scaling of the "dispersion coefficients" can then be expressed in terms of the small parameter τ (or, more rigorously, in terms of τ/T_L) to show that

$$\langle U_s\, \xi_s\rangle \sim 1, \text{ while } \langle U_p\, \xi_s\rangle \sim \tau \text{ and } \langle x_p\, \xi_s\rangle \sim \tau^2. \tag{96}$$

However, to obtain a mathematically correct result for the PDF equation, we must go to the white-noise limit. Indeed, when $\tau \to 0$ with $\langle \xi_s^2\rangle \to \infty$, such that $\lim_{\tau\to 0}(\tau\langle \xi_s^2\rangle) = \widetilde{K}$ where $\widetilde{K}$ is a positive constant, the dispersion coefficients tend to

$$\Lambda_{U_s} \underset{\tau\to 0}{\longrightarrow} \widetilde{K},\ \Lambda_{U_p} \underset{\tau\to 0}{\longrightarrow} 0,\ \Lambda_{x_p} \underset{\tau\to 0}{\longrightarrow} 0. \tag{97}$$

In that limit, the second-order tensor $\widetilde{\mathbf{B}}$ becomes

$$\lim_{\tau\to 0}\widetilde{\mathbf{B}} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 2\,\widetilde{K} \end{pmatrix} \tag{98}$$

with eigenvalues equal to $e_{v1} = e_{v2} = 0$ while $e_{v3} > 0$. Thus, $\widetilde{\mathbf{B}}$ is now degenerate but with a strictly positive submatrix (here the positive coefficient $\widetilde{K}$) and we retrieve a well-posed FPE for the PDF equation in Eq. (90). The corresponding trajectory equations are

$$dx_p = U_p\, dt, \tag{99a}$$

$$dU_p = \frac{U_s - U_p}{\tau_p}\, dt, \tag{99b}$$

$$dU_s = -\frac{U_s}{T_L}\, dt + \sqrt{2\widetilde{K}}\, dW, \tag{99c}$$

which are also obtained by (loosely) writing directly that $\lim_{\tau\to 0}\xi_s\, dt \simeq \sqrt{2\widetilde{K}}\, dW$ in the original trajectory equations, Eqs. (86). In short, this shows that we retrieve exactly the Langevin model by the combined application of the FND relation and of fast-variable elimination techniques [4,14].

As indicated in Sec. IV B, the elimination of the acceleration of the velocity of the fluid seen is supported by the Kolmogorov theory [2,4,36] which indicates that τ is of the order of the Kolmogorov time scale τ_η while T_L is of the order of the time scale of the large scales, with $\tau_\eta \ll T_L$ for high Reynolds-number flows. This provides interesting insights into the nature of the modeling steps made in the dynamic PDF approach that were first mentioned at the end of Sec. IV B. Going back to the general case, we can write the increments of $\mathbf{U}_s(t)$ [cf. Eq. (23c) with Eq. (24)] as

$$\mathbf{U}_s(t + \Delta t) - \mathbf{U}_s(t) = \int_t^{t+\Delta t} \mathbf{A}_s(t', \mathbf{Z}_p(t'))\, dt' + \int_t^{t+\Delta t} \mathbf{B}_s(t', \mathbf{Z}_p(t'))\, \boldsymbol{\xi}_s(t')\, dt'. \tag{100}$$

When $\Delta t \gg \tau_\eta$, the integral of this rapid part can be split into the sum of several integrals, using a partition $(t_i)_{i=1,n}$ of the interval $[t; t + \Delta t]$ such that $t_1 = t$, $t_n = t + \Delta t$, and $t_{i+1} - t_i = k\tau_\eta$ (with $k \gg 1$, for $i = 1, \ldots, n-1$)

$$\int_t^{t+\Delta t} \mathbf{B}_s(t', \mathbf{Z}_p(t'))\, \boldsymbol{\xi}_s(t')\, dt' = \sum_{i=1}^{n-1} \int_{t_i}^{t_{i+1}} \mathbf{B}_s(t', \mathbf{Z}_p(t'))\, \boldsymbol{\xi}_s(t')\, dt'. \tag{101}$$

In the dynamic PDF approach, we are considering the conditional increments $d\mathbf{U}_s|(\mathbf{Z}_p(t) = \mathbf{z}_p)$. The modeling steps consist in freezing the slowly varying functions $\mathbf{A}_s(t', \mathbf{Z}_p(t'))$ and $\mathbf{B}_s(t', \mathbf{Z}_p(t'))$ at $\mathbf{A}_s(t, \mathbf{Z}_p(t))$ and $\mathbf{B}_s(t, \mathbf{Z}_p(t))$ in Eq. (100) and in regarding each of the integrals on the right-hand side of Eq. (101) as being nearly independent. Then the central limit theorem (CLT) for $n \gg 1$, that is, when $\Delta t \gg \tau_\eta$, allows us to approximate this sum as a Gaussian random variable. This reasoning clarifies the meaning of the nature of the Gaussian hypothesis made in the dynamic-PDF description

where $d\mathbf{U}_s|(\mathbf{Z}_p(t)=\mathbf{z}_p)$ is assumed to be Gaussian over small time increments Δt (small with respect to T_L, $\Delta t \ll T_L$, to justify freezing the functions $\mathbf{A}_s$ and $\mathbf{B}_s$ but large with regard to τ_η, $\Delta t \gg \tau_\eta$, to resort to the CLT). It also justifies the consideration of Gaussian noises in the present section as with the original form in Eqs. (86). Then, as explained above, the meaningful form of the dynamic PDF model is obtained by taking the limit of vanishing τ (the white-noise limit). This yields the formulation given in Eq. (25c) in Sec. IV B. Note that, in the fluid case, the limit $\lim_{\tau\to 0}(\tau\langle\xi_s^2\rangle)$ is indeed predicted as being finite by the Kolmogorov theory since, for $\mathrm{Re}\to\infty$ (or $\tau_\eta\to 0$), this limit is the nonvanishing mean turbulent kinetic energy dissipation $\lim_{\tau\to 0}(\tau\langle\xi_s^2\rangle)\simeq\langle\epsilon\rangle(t,\mathbf{x}_p)$ [2,36]. This point has been covered in detailed studies for Lagrangian stochastic approaches to single-phase turbulent flows [1,2,45] as well as to disperse turbulent two-phase flows [4,6].

Unfortunately, these ideas cannot be carried to velocities governed by the fluid large scales that is when the same elimination procedure is attempted on the velocity of the fluid seen, as done in the kinetic PDF approach. Indeed, using Eqs. (99) and applying the same procedure, we get for the "dispersion coefficients" λ and μ

$$\mu=\frac{1}{\tau_p}\langle U_p\,U_s\rangle=\frac{1}{\tau_p}\frac{T_L}{\tau_p+T_L}\langle U_s^2\rangle, \tag{102a}$$

$$\lambda=\frac{1}{\tau_p}\langle x_p\,U_s\rangle=\frac{1}{\tau_p}\frac{T_L^2}{\tau_p+T_L}\langle U_s^2\rangle, \tag{102b}$$

which, of course, scale as successive powers in the time scale T_L of the eliminated variable $U_s(t)$ with, in particular, $\lambda/\mu=T_L$. Note that, by taking $\tau_p=\tau_p^{\mathrm{st}}$, the usual expressions of λ and μ for the homogeneous turbulence case are retrieved. Yet, in that case, the proper dimensionless parameter that appears is T_L/τ_p which is the inverse of the particle Stokes number $\mathrm{St}_p=\tau_p/T_L$. For high-inertia particles with $\mathrm{St}_p\gg 1$, the elimination procedure can be applied and leads to a FPE since λ can be neglected. Note that we retrieve then a well-defined and closed PDF description since for such "bullet particles" the fluid seen can be regarded as a weak white-noise term (that is, we find again that descriptions are complete when the external forces are fully determined, here as white noise). This is not, however, an interesting limit and the issue is mostly to describe particles having low and moderate Stokes numbers. Yet, in this range of values, the elimination of the velocity fluid seen is no longer possible, leaving the ill-posed kinetic PDF equation.

To summarize, these developments show that Gaussian colored noise can always be eliminated but that the resulting PDF equation is ill based and cannot be applied outside the Gaussian world. Only the elimination of white-noise terms leads to a closed and well-posed formulation and an acceptable general PDF description.

C. Two stochastic modeling approaches

The two specific examples studied in Sec. IV A for the kinetic PDF model and in Sec. IV B for the dynamic one reflect two different modeling approaches.

In the first modeling approach, the particle state vector is fixed right from the outset to $\mathbf{Z}_p^r=(\mathbf{x}_p,\mathbf{U}_p)$. The analysis carried out in the preceding sections indicates that this description has a chance to be physically relevant and mathematically well posed only if the actions of force fields external to this fixed particle system are fully known. Otherwise, the example of the actions of a turbulent fluid flow described by limited information (a few statistical moments) reveals that the resulting probabilistic descriptions are restrained (to a Gaussian world) or suffer from an ill-based behavior of their solutions.

The second modeling approach adopts a more flexible point of view and seeks to adjust the state vector to each problem and to the nature of the information at hand. In that case, the mathematical toolbox consists of Markov processes and well-defined stochastic diffusion processes (only continuous physical phenomena in the frame of fluid mechanics have been considered in this work). Then, the "art of modeling" consists in separating the slow and fast modes between the degrees of freedom of a system and in replacing rapidly varying ones by white-noise terms to be able to apply the well-defined mathematical machinery. Depending on the available information, different descriptions can then be chosen for the same system. Note that this second modeling approach is typical in synergetics [46].

D. A thermodynamic interpretation

It is possible to give a thermodynamical flavor to the previous arguments, which is helpful to outline the difference between the two modeling approaches. It was shown that the descriptions considered in this work can be formulated in a compact notation by a general equation of the following form [cf. for example Eq. (8) and Eq. (65)]:

$$\frac{\partial p}{\partial t}=-\frac{\partial[A_i\,p\,]}{\partial z_i}+\frac{\partial^2[B_i\,p]}{\partial z_i^2}=-\frac{\partial J_i}{\partial z_i}, \tag{103}$$

where the matrix $\mathbf{B}$ has been written here in a diagonal form (note that this is always possible since $\mathbf{B}$ is symmetrical) and with the flux J_i given by

$$J_i(t;\mathbf{z}_p)=A_i\,p-\frac{\partial[B_i\,p]}{\partial z_i}. \tag{104}$$

Then the entropy of the system is defined by [47–49]

$$S(t)=-\int p(t;\mathbf{z}_p)\,\ln p(t;\mathbf{z}_p)\,d\mathbf{z}_p, \tag{105}$$

thus as the equivalent of the Gibbs entropy, and it can be shown that entropy production can be identified with [47–49]

$$\Pi=\int\frac{|J_i(t;\mathbf{z}_p)|^2}{B_i\,p(t;\mathbf{z}_p)}\,d\mathbf{z}_p. \tag{106}$$

In the dynamic PDF model, the second-order tensor is definite positive (for the reduced set of variables on which white-noise terms are acting) and we have $\Pi>0$. On the other hand, in the kinetic PDF model, there is typically at least one negative eigenvalue and thus, potentially, one can have a negative entropy production $\Pi<0$.

Although surprising at first sight, this is in line with the previous accounts. In the kinetic PDF description, one is considering an *open thermodynamical system* (made up by $\mathbf{Z}_p^r$) in contact with another system represented by the velocity

of the fluid seen $\mathbf{U}_s$ but which is treated as the outside world. However, unless we are limited to very high inertia particles for which the velocity of the fluid can be seen as acting as a heat bath, the velocity of the fluid seen $\mathbf{U}_s$ is varying with time scales comparable to the particle one. This is then reflected in the nonzero λ_{ij} terms appearing in the kinetic PDF equation. Yet, as demonstrated above, this characteristic feature is also the very same one that makes the description ill posed. In thermodynamical terms, this translates the fact that we are considering a system in contact with another one but that is also slowly varying. As for any open subsystem of a larger one, nothing much can be assessed for the evolution of its entropy. It can also be seen that attempts at describing such open systems as closed ones are not likely to succeed.

On the other hand, in the dynamic PDF model, the extended-particle system $\mathbf{Z}_p$ is now described as a *thermodynamical system in contact with a heat bath* represented by the fast variables eliminated and the resulting white-noise terms, and this whatever the particle inertia. In that sense, the two approaches clearly differ with the dynamic PDF approach following a classical thermodynamic road.

IX. CONSEQUENCES FOR PRACTICAL SIMULATIONS

While the previous analyses clarify the content of the kinetic and dynamic PDF approaches, they are also relevant for practical simulations. This corresponds to the twofold interest of PDF formulations mentioned in the Introduction, both as stand-alone approaches and as roads towards closed macroscopic models.

The first consequence is related to the direct simulation of the chosen PDF. One possibility is to consider the PDF equation in sample space and use classical numerical methods (e.g., finite volume schemes). In the kinetic PDF approach, this has been used in some works [17]. Yet, even with the reduced particle state vector $\mathbf{Z}_p^r$, we are considering an equation in a six-dimensional space since $\mathbf{z}_p^r = (\mathbf{y}_p, \mathbf{V}_p)$ and this can be attempted only for some special configurations where the dimension can be reduced to typically one spatial direction and two or three independent variables. Furthermore, it is now clear from the ill-posed nature of the kinetic PDF equation demonstrated in Sec. VII B that attempts at solving this equation in the general case are doomed to fail. In the dynamic PDF formulation, the resulting PDF equation is well posed and a stand-alone approach is possible. The particle state vector is extended to $\mathbf{Z}_p$, which means that the sample space dimension of $\mathbf{z}_p = (\mathbf{y}_p, \mathbf{V}_p, \mathbf{V}_s)$ is already equal to 9 for a general three-dimensional fluid flow (in physical space). This dimension accounts only for dynamical variables and, as PDF approaches are typically attractive for more complex problems (involving for instance chemical reactions, evolving particle diameter, etc.), we are thus dealing with equations in high-dimensional spaces. Obviously, classical numerical schemes cannot be applied anymore and numerical solutions of the PDF equation rely on Monte Carlo methods, which indicates that one is simulating a large number of stochastic particle realizations and approximating the actual PDF in a weak sense [1,4,15]. This has become a well-established simulation method and presentations can be found in several works, see Refs. [1,50,51] for comprehensive discussions of stochastic numerical schemes for dynamic PDF approaches based on Langevin models and assessment of numerical costs. Detailing the numerical aspects of simulations carried out with dynamic PDF models is outside the scope of this article but various validation cases and applications can be found in Refs. [4,5,51] and in a recent survey [6].

The second consequence is of importance for the derivation of macroscopic descriptions made up by two-fluid models. Indeed, one application of PDF models is to use PDF equations as an intermediate step to extract a set of moment equations [typically, the set of partial differential equations (PDE) for particle first and second one-point moments]. At first sight, this appears as particularly interesting with the kinetic PDF description since the reduced state vector implies that the first two moments yield a closed formulation in terms of the particle mean velocity field and kinetic stresses only. This corresponds to a set of 10 PDEs, which represents a numerical effort that remains tractable for monodisperse particles and to a macroscopic description on a par with classical Reynolds-stress models (cf. $R_{ij} - \epsilon$ formulations) that still make up the mainstream of classical models for single-phase turbulent flows. However, the status of such a moment (or Eulerian) description can then be pondered. Indeed, the ill-based nature of the kinetic PDF equation casts some doubts on the validity of these moment equations. On the other hand, it does not necessarily invalidate them. From the present analysis, it appears more justified to consider the set of moment equations derived from the dynamic PDF approach. The resulting Eulerian description and two-fluid model was derived in Refs. [4,10] and it is worth emphasizing that, for the same level of physics contained in the model (typically, in the Langevin equation used for the velocity of the fluid seen), the corresponding two-fluid model consists in a set of at least 19 coupled PDEs since transport equations must also be written for the fluid-particle velocity tensor $\langle u_{p,i} u_{s,j} \rangle$ which is not symmetrical. This represents already a considerable computational effort for monodisperse particles. When particles are polydisperse, the closure issues of the particle relaxation time scale and momentum exchange term [4,6] means that Eulerian simulations must be run for a set of particle diameter classes, implying that the total numerical task is now to solve a set of $19N_{\text{class}}$ coupled PDEs where N_{class} is the number of particle classes in a simulation. This is clearly a daunting challenge which explains that Lagrangian stochastic methods, or Monte Carlo simulations of the dynamic PDF equation, are of direct interest for practical applications.

These considerations do not mean that Lagrangian stochastic and two-fluid models should be regarded as competitive methods. For example, new hybrid PDF-moment formulations have recently surfaced (see an introduction to these new ideas in Ref. [6, Sec. 5.3]). Yet, it is important that closed moment formulations be properly obtained and be shown to represent realizable models, which points to the use of the complete set of second-order transport equations for the particle mean velocity, particle kinetic stresses, and full fluid-particle velocity tensor. However, if the derivation of a reduced set of moment equations is still deemed useful, it must be put on a sound footing. For that purpose, the following procedure is proposed: Embed the kinetic description into a dynamic one, derive the extended set of moment equations at this

particle-fluid-velocity-seen level (this is done, for example, in Refs. [4,10]), and then work our way "downwards" by injecting local equilibrium assumptions for the various correlations with $\mathbf{U}_s$ that appear to derive a reduced and closed set of equations. If such equilibrium simplifications are not physically justified, then the conclusions of this work support the argument that the physics of disperse two-phase flows is best addressed at the level of dynamic PDF approaches and, thus, with the full set of moment equations that include transport equations for particle-fluid-seen correlations.

X. CONCLUSIONS

The aim of this article was to assess the kinetic and dynamic PDF models proposed in the literature for disperse two-phase flows. In a broader perspective, its objective was also to bring out a consistent approach to one-particle PDF descriptions of the dynamics of particles in turbulent flows. Drawing on the developments presented in this article, several conclusions emerge. They are gathered and presented along two main lines.

First, in the framework of the hypothesis of a Gaussian distribution for the velocity of the fluid seen and of constant particle relaxation time scale (for monodisperse particles) imposed by the limitation of the kinetic PDF state vector:

(G1) a new derivation of the kinetic PDF equation has been obtained by integrating a dynamic PDF description over the velocity of the fluid seen and new physically meaningful expressions of the "dispersion tensors" have been given as the correlations with the eliminated noise (cf. Sec. V);

(G2) these results demonstrate that the kinetic PDF equation is retrieved as the marginal of the dynamic PDF approach and that present kinetic PDF descriptions are always contained in dynamic-PDF ones when the velocity of the fluid seen has a Gaussian distribution (cf. Sec. V).

Second, when the two approaches are analyzed as general models, and therefore outside of the Gaussian context, two very different pictures are revealed:

(NG1) the dynamic-PDF approach, which is based on Langevin or FPE types of models, benefit directly from the great body of work devoted to their analysis that has shown that such approaches are well posed and form acceptable stand-alone PDF descriptions, in the sense that the criteria (C1) and (C2) are met (cf. Sec. VI);

(NG2) on the contrary, neither criterion (C1) nor (C2) are satisfied by the kinetic-PDF approach. One of the key results is that, whatever the closure of the "dispersion tensors," the kinetic PDF equation is ill posed (as soon as $\lambda_{ij} \neq 0$) and cannot be applied outside of special cases and special initial conditions, which represents a serious shortcoming for a general modeling proposal (cf. Sec. VII);

(NG3) as the main modeling issues concern space-dependent closures for inhomogeneous flows (related to non-Gaussian fluid statistics), it has been shown that the dynamic PDF approach represents not only a safe mathematical road but yields naturally nonlocal closures at the level of particle statistics since the space-dependent correlations of the fluid velocity seen along particle trajectories are predicted by its formulation. Within the limits of each model chosen for the velocity of the fluid seen, it is thus seen that the dynamic PDF description offers a consistent approach to derive nonlocal effects on particle statistics (cf. Sec. V).

One of the most important outcome is that the status of a PDF formulation is not related to the choice of the modeling approach used to derive flux closures in sample space but to a physically sound selection of the variables retained in the particle state vector combined with the use of fast-variable elimination techniques. In that respect, an interesting aspect of the exact Furutsu-Novikov-Donsker relation (for Gaussian processes) is to demonstrate that attempts at formulating consistent PDF descriptions for systems influenced by external colored noise are likely to fail (cf. Sec. VIII B). This is helpful in that it points to a clear and mathematically correct approach to guide necessary physically oriented improvements of PDF descriptions of particle-laden turbulent flows. The importance of a relevant choice of the particle state vector has been emphasized repeatedly, mostly with physical arguments [4–6,13], and is now further supported by the present theoretical analyses as a central issue if one is to obtain acceptable and well-posed PDF descriptions. More precisely, the demonstrations given in this work point to the requirement of addressing disperse two-phase flow PDF modeling from a dynamical PDF standpoint. Present state-of-the-art Langevin models have been shown to be able to yield interesting results in practical applications [6] but much work remains to be done to improve current PDF formulations. In that sense, the conclusions developed in this paper complement the recent analysis of Langevin models for the dynamic PDF approach [37], in which requirements and guidelines were established, and are helpful to orient future modeling works towards well-posed and applicable PDF models.

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