Spatially inhomogeneous transition probabilities as memory effects for diffusion in statistically homogeneous random velocity fields

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Whenever one uses translation invariant mean Green's functions to describe the behavior in the mean and to estimate dispersion coefficients for diffusion in random velocity fields, the spatial homogeneity of the transition probability of the transport process is implicitly assumed. This property can be proved for deterministic initial conditions if, in addition to the statistical homogeneity of the space-random velocity field, the existence of unique classical solutions of the transport equations is ensured. When uniqueness condition fails and translation invariance of the mean Green's function cannot be assumed, as in the case of nonsmooth samples of random velocity fields with exponential correlations, asymptotic dispersion coefficients can still be estimated within an alternative approach using the Itô equation. Numerical simulations confirm the predicted asymptotic behavior of the coefficients, but they also show their dependence on initial conditions at early times, a signature of inhomogeneous transition probabilities. Such memory effects are even more relevant for random initial conditions, which are a result of the past evolution of the process of diffusion in correlated velocity fields, and they persist indefinitely in case of power law correlations. It was found that the transition probabilities for successive times can be spatially homogeneous only if a long-time normal diffusion limit exits. Moreover, when transition probabilities, for either deterministic or random initial states, are spatially homogeneous, they can be explicitly written as Gaussian distributions.

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I. INTRODUCTION

Techniques from classical field theory used to calculate mean values of passive scalars transported in random velocity fields and to estimate dispersion coefficients, as for instance, in applications to turbulence [1] or to contaminant transport in groundwater [2,3], often assume the translation invariance of the ensemble mean Green's function. The central role of the one-particle dispersion in predicting the behavior in the mean of the transport process [4,5] also relies on the spatial homogeneity of the mean Green's function. Since this property is postulated in many cases [1–3,6], specifying the premises which allow rigorous proofs can be helpful in identifying the limits of applicability of the theoretical results.

The Green's function $g(\mathbf{x},t|\mathbf{x}_0)$ is the fundamental solution [Cauchy problem $g(\mathbf{x},0|\mathbf{x}_0)=\delta(\mathbf{x}-\mathbf{x}_0)$] of the Fokker-Planck equation

$$\partial_t g + \mathbf{u}\,\boldsymbol{\nabla}\, g = D_0 \nabla^2 g, \tag{1}$$

with space variable drift $\mathbf{u}(\mathbf{x})$ which is a sample of a random velocity field, and a local diffusion coefficient D_0 which is assumed constant in many theoretical investigations. The Green's function g is the density of the transition probability $P(d\mathbf{x},t,\mathbf{x}_0)=g(\mathbf{x},t|\mathbf{x}_0)d\mathbf{x}$ of the Itô process,

$$X_i(t) = x_{0i} + \int_0^t u_i[\mathbf{X}(t')]dt' + W_i(t), \tag{2}$$

where $i=1,2,3$, $x_{0i}=X_i(0)$ is a deterministic initial position, and W_i are the components of a Wiener process of mean zero and variance $\langle W_i^2(t)\rangle = 2D_0 t$. The Itô process is a Markov process [7], often called Itô diffusion [8]. The process is completely characterized by its transition probabilities, which in general are not spatially homogeneous, the only diffusion processes with space-homogeneous transition probabilities being the Brownian motion with drift and diffusion coefficients constant in space [9]. An intriguing question is whether the average with respect to the homogeneous statistics of the velocity field renders the transition probabilities translation invariant.

Some authors, for instance, Phythian and Curtis [1], state that, because of the homogeneous nature of the probability distribution of the velocity field, the ensemble mean Green's function $\bar{g}$ is invariant to space translations, i.e., $\bar{g}(\mathbf{x},t|\mathbf{x}_0) = \bar{g}(\mathbf{x}-\mathbf{x}_0,t|\mathbf{0})$. More recently, Dentz and Tartakovsky [2] based their argumentation on the statement that $\bar{g}$ should be invariant to translations in space of the statistically homogeneous velocity field or, as one can see in the literature [10,11], this is equivalent to postulating the translation invariance of the mean Green's function. Within the "usual setup for homogeneity" formulated by Zirbel [12], the statistical homogeneity of the displacement $\tilde{X}_i(t)=X_i(t)-x_{0i}$, and implicitly of the Green's function, can be proved for deterministic initial conditions, homogeneous Eulerian velocity fields, and unique classical solutions of the transport equations.

With rare exceptions, e.g., [1], it is not emphasized that estimations of mean values by translation invariant mean Green's functions are only done for nonrandom initial conditions. Therefore, one might be tempted to assume homogeneous transition probabilities for successive states of the process describing the mean transport, i.e., translation invariant mean Green's functions for random initial conditions. This

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assumption is, as shown in Sec. IV of this paper, equivalent in a wide sense with assuming normal diffusion behavior, described by Gaussian transition probabilities. In fact, we will see that spatially homogeneous transition probabilities are rather the exception than the rule. In many situations of practical interest inhomogeneous transition probabilities are associated to persistent memory of the diffusion in random velocity fields.

The paper is organized as follows. After discussing the conditions for statistical homogeneity properties of transport observables in Sec. II, a situation where these conditions are not fulfilled is investigated in Sec. III through approximations based on Itô equation and numerical simulations. Memory effects and inhomogeneous transition probabilities for deterministic and random initial conditions are investigated in Sec. IV. Section V provides concluding remarks.

II. HOMOGENEITY PROPERTIES

The statistical homogeneity of some random function $B(A)$ is implied by the homogeneity of its argument A only if B is function of a single variable [13] or if it has a particular dependence on A. To be specific, within the usual setup for homogeneity [10–12,14], the random function is identified with an element ω of the canonical probability space, $A(\omega,\mathbf{x})=\omega(\mathbf{x})$, and the homogeneity is the measure preserving property of the shift maps, $(\tau_{\mathbf{x}_0}\omega)(\mathbf{x})=\omega(\mathbf{x}+\mathbf{x_0})$. In order for B to be homogeneous it must depend on ω and $\mathbf{x}_0$ only through measure preserving shifts, $B=B(\tau_{\mathbf{x}_0}\omega)$ ([12], Remark 2.1.).

To see that g has the desired dependence on the statistics of $\mathbf{u}$, let us translate the system of coordinates by $\mathbf{x}_0$. Equation (1) written in the new variables $\tilde{\mathbf{x}}=\mathbf{x}-\mathbf{x}_0$,

$$\partial_t g(\tilde{\mathbf{x}},t|\mathbf{0}) + \nabla[\mathbf{u}(\tilde{\mathbf{x}}+\mathbf{x}_0)g(\tilde{\mathbf{x}},t|\mathbf{0})] = D_0\nabla^2 g(\tilde{\mathbf{x}},t|\mathbf{0}), \quad (3)$$

shows that g depends on velocity statistics only through measure preserving shifts $\mathbf{u}(\tilde{\mathbf{x}}+\mathbf{x}_0)=\tau_{\mathbf{x}_0}\omega$. If the solution $g(\mathbf{x},t|\mathbf{x}_0,\omega)$ to Eq. (1) for the deterministic initial condition $g(\mathbf{x},0|\mathbf{x}_0)=\delta(\mathbf{x}-\mathbf{x}_0)$ is unique in a classical sense, then it is identical with the solution $g(\tilde{\mathbf{x}},t|\mathbf{0},\tau_{\mathbf{x}_0}\omega)$ to Eq. (3) for the initial condition $g(\tilde{\mathbf{x}},0|\mathbf{0})=\delta(\tilde{\mathbf{x}})$. Thus, $g(\mathbf{x},t|\mathbf{x}_0,\omega)=g(\mathbf{x}-\mathbf{x}_0,t|\mathbf{0},\tau_{\mathbf{x}_0}\omega)$ and the measure preserving property of the shift $\tau_{\mathbf{x}_0}$ implies the translation invariance of the ensemble mean Green's function, $\bar{g}(\mathbf{x},t|\mathbf{x}_0)=\bar{g}(\mathbf{x}-\mathbf{x}_0,t|\mathbf{0})$.

We can see that the argument of invariance of the mean Green's function to spatial translations of the random field used in Refs. [2,3] is in fact a postulate of the translation invariance of the mean Green's function. Instead, standard approaches within the usual setup for homogeneity first infer the equality of the nonaveraged Green's functions for the original and for the shifted field from the uniqueness of the solutions, and then use the homogeneity of the random field to complete the proof [10,11]. Disregarding the uniqueness condition and the necessity to use deterministic initial conditions can induce the wrong perception that transition probabilities are spatially homogeneous for arbitrary initial states, whenever the velocity field is statistically homogeneous or, as it will be shown in Sec. IV, this is true only in the limit of normal diffusive behavior if such a limit exists.

The solutions of the Fokker-Planck equation (3) are probability densities of the displacements $\tilde{X}_i(t)=X_i(t)-x_{0i}$, which, according to Eq. (2), solve the Itô equation

$$\tilde{X}_i(t) = \int_0^t u_i[\tilde{\mathbf{X}}(t') + \mathbf{x}_0]dt' + W_i(t). \quad (4)$$

If Eq. (4) has pathwise unique solutions, then for every fixed realization of the Wiener process, the trajectory $\mathbf{X}(t;\mathbf{x}_0)$ has the flow property $\mathbf{X}(t;\mathbf{x}_0,\omega)-\mathbf{x}_0=\tilde{\mathbf{X}}(t;\mathbf{0},\tau_{\mathbf{x}_0}\omega)$; hence the displacement field $\tilde{\mathbf{X}}$ is homogeneous [12]. This implies the homogeneity of the Lagrangian velocity field $\mathbf{u}^L(t;\mathbf{x}_0,\omega)=\mathbf{u}[\tilde{\mathbf{X}}(t;\mathbf{0},\tau_{\mathbf{x}_0}\omega)]$. Conversely, assuming the homogeneity of the Lagrangian velocity field, Eq. (4) implies the homogeneity of $\tilde{\mathbf{X}}$. Obviously, translation invariance of $\bar{g}$ and homogeneity of $\tilde{\mathbf{X}}$ are equivalent. Concluding, we see that translation invariance of $\bar{g}$ and homogeneity of $\tilde{\mathbf{X}}$ and $\mathbf{u}^L$ are equivalent properties, provided Eqs. (3) and (4) admit unique solutions. Nevertheless, the last two are more general and hold even if the coefficients of the Fokker-Planck equation do not possess the smoothness required to ensure the existence of the classical solutions [8], as well as in case of discrete-space processes, when the probability distribution of $\tilde{\mathbf{X}}$ has no density [12].

The discussion above emphasizes the key role of the uniqueness of the solutions and implicitly of the smoothness of the velocity samples. When the velocity field has a Gaussian shaped correlation, the velocity samples are analytical functions [13]. But for exponential correlations, e.g., Kraichnan fields consisting of a superposition of a finite number of random periodic modes [4], the sufficient conditions for sample smoothness (i.e., differentiability of the correlation function at the origin) are not fulfilled [13]. It is generally accepted that these sufficient conditions can hardly be further narrowed and they practically coincide with the necessary conditions (see [13] and references therein).

Exponential correlations occur in applications when the velocity is an Ornstein-Uhlenbeck process [15]. For instance, samples of a homogeneous one-dimensional velocity field with mean zero and correlation function $\overline{V(x)V(0)}=(1/2)\exp(-|x|)$ are solutions of the Langevin equation $dV(x)=-V(x)dx+dW(x)$. Because the Wiener process scales as $dW\sim(dx)^{1/2}$, the limit of $\Delta V/\Delta x$ for $\Delta x\to 0$ does not exist and $V(x)$ is not differentiable almost everywhere [8].

Since Lipschitz continuity is a requirement for pathwise unique solutions to Itô equation [8], we estimated numerically the Lipschitz constant $|\Delta V|/|\Delta x|$ for a sample of the transverse component of the two-dimensional Kraichnan velocity field with exponential correlation used in [4]. Figure 1 shows that the estimated Lipschitz constant behaves similarly to that of the nondifferentiable Ornstein-Uhlenbeck process. Similar tests show that, while for Gaussian correlations the estimated Lipschitz constants are smaller than 0.5, for exponential correlations they increase with increasing number of periodic modes. Thus, very likely, the samples of the exponentially correlated Kraichnan field are not Lipschitz continuous.

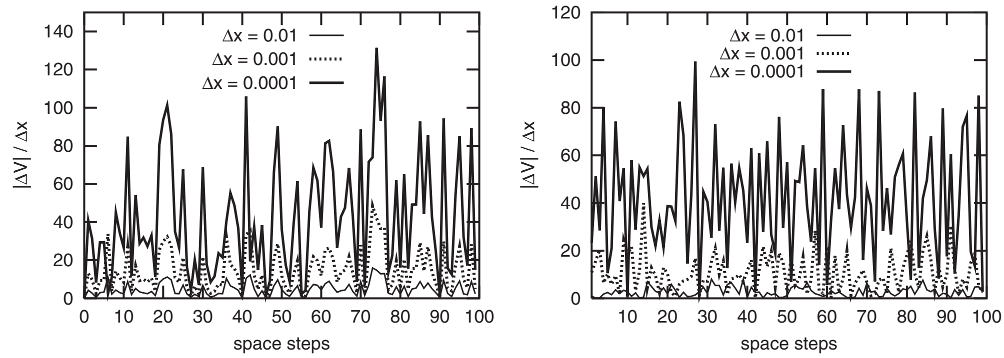

FIG. 1. Behavior of the estimated Lipschitz constant as function of Δx for a sample of the Ornstein-Uhlenbeck process (left) and a Kraichnan velocity sample with exponential correlation generated with 6400 random periodic modes (right).

When Lipschitz continuity does not hold, the proof of uniqueness for the solutions of the Itô equation (2) fails. As for the Fokker-Planck equation, it cannot even be written in the form of Eq. (1) for divergence-free velocity. Hence, the approximation methods based on Eq. (1) cannot be used for exponentially correlated velocity fields. If instead of classical solutions of the Fokker-Planck equation one looks for weak solutions, e.g., by using discrete operators [11], then the formalism has to be modified in major ways.

III. APPROXIMATIONS OF DISPERSION COEFFICIENTS

In the following it will be shown that even if the velocity samples do not possess the smoothness required for unique solutions, the Itô equation can still be used to approximate the leading terms in the expansion of the dispersion coefficients for small variance σ^2 of the velocity field. To this end we write Eq. (2), for $\mathbf{x}_0=\mathbf{0}$, in nondimensional form,

$$X_i(t) = \delta_{i,1}t + \epsilon\int_0^t u_i[\mathbf{X}(t')]dt' + \mathrm{Pe}^{-1/2}\, W_i(t), \qquad (5)$$

where $\mathrm{Pe}=U\lambda/D_0$ is the Péclet number, $\epsilon=\sigma/U$ describes the velocity fluctuations, $U=|\bar{\mathbf{u}}|$ is the modulus of the mean velocity, and $\lambda=\lambda_{11}$ is the correlation length of the velocity field in the mean flow direction. We used the scaling relations $\hat{X}_i=X_i\lambda$, $\hat{t}=t\lambda/U$, $\hat{u}_i=\delta_{i,1}U+\epsilon u_i U$, and $\hat{W}_i=W_i\lambda\mathrm{Pe}^{-1/2}$ (with hats for dimensional quantities) [16]. The diagonal dispersion coefficients are defined by the long time limit of $D_{ii}(t)=\frac{1}{2}d\Sigma_{ii}(t)/dt$, where

$$\Sigma_{ii}(t) = \overline{\langle [X_i(t) - \overline{\langle X_i(t)\rangle}]^2\rangle}$$

are the second centered moments of the ensemble mean concentration [5]. With the order of magnitude hypothesis $\mathrm{Pe}^{-1/2}=\epsilon^{\alpha}$, from Eq. (5) one obtains

$$X_i(t) - \overline{\langle X_i(t)\rangle} = \epsilon\int_0^t v_i[\mathbf{X}(t')]dt' + \epsilon^{\alpha}W_i(t), \qquad (6)$$

where $v_i[\mathbf{X}(t')]=u_i[\mathbf{X}(t')]-\overline{\langle u_i[\mathbf{X}(t')]\rangle}$. For statistically homogeneous Lagrangian velocity $\overline{u_i[\mathbf{X}(t')]}=0$ and $v_i[\mathbf{X}(t')]=u_i[\mathbf{X}(t')]$. Consequently, the cross correlations

$$\overline{\langle W_i(t)v_i[\mathbf{X}(t')]\rangle} = \langle W_i(t)\overline{u_i[\mathbf{X}(t')]}\rangle$$

vanish. By averaging the square of Eq. (6) and taking the time derivative one obtains the dispersion coefficient as an exact sum of the local dispersion coefficient and the contribution of the Lagrangian velocity correlation,

$$D_{ii}(t) = \epsilon^{2\alpha} + \epsilon^2\int_0^t \overline{\langle u_i[\mathbf{X}(t)]u_i[\mathbf{X}(t')]\rangle}dt'. \qquad (7)$$

Compared with equivalent representations of dispersion coefficients derived by methods of classical field theory [1,2], Eq. (7) has a straightforward physical interpretation: it is a Kubo formula relating dispersion coefficients to velocity correlations. The convergence of the integral in Eq. (7) for $t\to\infty$ ensures finite correlation times τ_{ii} of the Lagrangian velocity, which correspond to the criterion for diffusive limit formulated by Fannjiang and Komorowski [14].

Equation (7) generalizes the additivity relation occurring in diffusion limit theorems [14,17] and in formal first-order approximations in ϵ^2 [16]. The latter can be derived from Eq. (7) for trajectory Eq. (5) written in vectorial form $\mathbf{X}(t)=\mathbf{X}^{(0)}(t)+\epsilon\Delta\mathbf{X}(t)$, with $\mathbf{X}^{(0)}(t)=(t,0,0)$, by formally expanding u_i as

$$u_i(\mathbf{X}) = u_i(\mathbf{X}^{(0)}) + \epsilon u_i'(\mathbf{X}^{(0)})\Delta\mathbf{X}, \qquad (8)$$

where u_i' denotes the Fréchet derivative. Then, assuming $\alpha\geq 1$, Eqs. (7) and (8) yield the following asymptotic behavior ($t\to\infty$):

$$D_{ii} \sim \epsilon^{2\alpha} + \epsilon^2\tau_{ii}^{(0)} + \epsilon^4 F[u_i(\mathbf{X}^{(0)}),u_i'(\mathbf{X}^{(0)})] + \cdots, \qquad (9)$$

where $\tau_{ii}^{(0)}$ are the Lagrangian correlation times and F is a functional of the Lagrangian velocity and of its Fréchet derivative evaluated along the unperturbed trajectory $\mathbf{X}^{(0)}$. If the velocity samples are bounded and are at least Lipschitz continuous, then F can be constructed by using integral representations of functionals of Itô processes (see, e.g., [18] and references therein). This could be a promising alternative to higher-order approximation methods based on the Fokker-Planck equation [1,2,6].

The correlation time $\tau_{ii}^{(0)}$ in Eq. (9) is the long time limit of the integral in Eq. (7) evaluated for $\mathbf{X}=\mathbf{X}^{(0)}$ and corresponds to the well-known consistent approximation by the first iteration of the Itô equation (5) about the mean flow

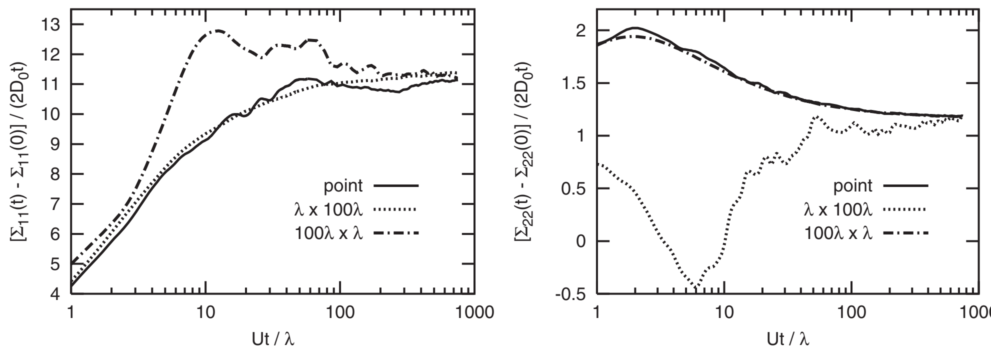

FIG. 2. Time behavior of the dispersion coefficients, represented as mean slopes of the longitudinal (left) and transverse (right) second moments of the ensemble mean concentration.

trajectory [16]. To the first order, $v_i[\mathbf{X}^{(0)}(t)] \equiv u_i[\mathbf{X}^{(0)}(t)]$ and Eq. (7) holds true for integrable velocity samples [condition required for the existence of the first-iteration solutions to Eq. (5)]. No Lipschitz or other smoothness assumptions are necessary. Such weak conditions are fulfilled not only by exponentially correlated fields but also by δ correlated white noise in space velocity fields. For velocity fields with finite correlation range, the existence of $\tau_{ii}^{(0)}$ is implied by the finiteness of the correlation lengths λ_{ii} and the first-order approximation in Eq. (9), reverted to dimensional variables, reads

$$D_{ii} \sim D_0 + \sigma^2 \lambda_{ii}/U. \tag{10}$$

If the Itô equation (5) is iterated once about the trajectory of the diffusion with drift equal to the mean flow velocity $\mathbf{Z}^{(0)}(t) = \mathbf{X}^{(0)}(t) + \epsilon^{\alpha}\mathbf{W}(t)$, Eq. (7) yields

$$D_{ii}(t) = D_0 + \int_0^t dt' \int \int \overline{u_i(\mathbf{x}) u_i(\mathbf{x}')} p(\mathbf{x}, t; \mathbf{x}', t') d\mathbf{x} d\mathbf{x}' \tag{11}$$

where $p(\mathbf{x},t;\mathbf{x}',t') = \langle \delta[\mathbf{x} - \mathbf{Z}^{(0)}(t)] \delta[\mathbf{x}' - \mathbf{Z}^{(0)}(t')] \rangle$ is the Gaussian joint probability density of the diffusion process $\mathbf{Z}^{(0)}(t)$. This inconsistent approximation, which considers the contribution ϵ^{α} in the zeroth order $\mathbf{Z}^{(0)}$, leads to the same asymptotic behavior as in Eq. (10) and, in addition, it accounts for enhanced diffusion in single realizations of the velocity field [Eq. (11) without overbar], not accounted for by the consistent approximation [16].

Going beyond formal expansions, limit theorems prove the weak convergence ($\varepsilon \to 0$, $t \to \infty$, and $\varepsilon^2 t = \text{const}$) of the rescaled process $X_i^{\epsilon}(t) = \epsilon X_i(t/\epsilon^2)$ to a diffusion process with coefficients given by Eq. (10) [14,17]. Since the proof given in [14] does not assume smooth velocity fields, one expects that the upscaled diffusion process also exists in case of exponentially correlated velocity fields.

Figure 2 presents some results of the numerical experiment described in [4,5]. For the transport parameters used in simulations, Eq. (10) gives the asymptotic dispersion coefficients $D_{11}/D_0 = 11$ and $D_{22}/D_0 = 1$ (because, by the construction of the Kraichnan field, $\lambda_{22} = 0$). Several initial instantaneous injection conditions were considered: point sources and injections in slabs with dimensions of λ times 100λ, oriented parallel and transverse to the mean flow direction. The simulations were carried out for an ensemble of 1 024 realizations of the Kraichnan velocity field, generated with 6 400 modes, by simultaneously tracking in each realization 10^{10} particles with the global random walk algorithm [19]. For these parameters, fairly larger than those used in previous simulations for similar problems (e.g., [2]), numerical tests indicated the reliability of the statistical estimates of the second moments [4]. The long time behavior of the slope of $\Sigma_{ii}(t)$ shown in Fig. 2 confirms the asymptotic dispersion coefficients [Eq. (10)] predicted by formal expansions and diffusion limit theorems. As one can see in Fig. 2, at finite times there is a significant dependence on initial conditions of the second moment $\Sigma_{ii}(t)$ of the mean concentration, which, as shown in the next section, provides a numerical evidence for the spatial inhomogeneity of the mean Green's function.

IV. DETERMINISTIC VERSUS RANDOM INITIAL CONDITIONS

The second moment of the mean concentration is the variance of the process $\mathbf{X}(t)$ defined by the ensemble of the diffusion trajectories [Eq. (2)], starting from all the initial positions, in all the realizations of the velocity field, $\Sigma_{ii}(t) = \text{var}\{X_i(t)\} = E\{[X_i(t) - E\{X_i(t)\}]^2\}$, where $E\{\cdots\} = \overline{\langle \cdots \rangle}$ denotes the expectation, $\langle \cdots \rangle$ is the average over the realizations of the Wiener process and over the initial distribution, and the overbar denotes the average over the realizations of the velocity field. For any specified distribution of the initial positions $\mathbf{X}(0) = \mathbf{X}_0$, the variance of the sum $X_i = X_{0i} + \tilde{X}_i$ is obviously given by the sum of the variances of the terms plus two times their covariance

$$\Sigma_{ii}(t) = \Sigma_{ii}(0) + \tilde{\Sigma}_{ii}(t) + m_{ii}(t), \tag{12}$$

where $\Sigma_{ii}(0) = \text{var}\{X_{0i}\} = E\{(X_{0i} - E\{X_{0i}\})^2\}$ is the variance of the initial positions, $\tilde{\Sigma}_{ii}(t) = \text{var}\{\tilde{X}_i(t)\} = E\{[\tilde{X}_i(t) - E\{\tilde{X}_i(t)\}]^2\}$, and

$$m_{ii}(t) = 2E\{(X_{0i} - E\{X_{0i}\})[\tilde{X}_i(t) - E\{\tilde{X}_i(t)\}]\}, \tag{13}$$

is the covariance term $m_{ii} = 2\text{cov}\{X_{0i}, \tilde{X}_i\}$, which carries the memory of the initial conditions and, therefore, has been called "memory term" [4,5].

To further analyze Eqs. (12) and (13), we note that $E\{\tilde{X}_i\}=E_{\mathbf{X}_0}\{E\{\tilde{X}_i|\mathbf{X}_0\}\}$, where $E_{\mathbf{X}_0}$ is the average over the initial positions and

$$E\{\tilde{X}_i|\mathbf{X}_0\}(t) = \int (x_i - x_{0i})\bar{g}(\mathbf{x},t|\mathbf{x}_0)d\mathbf{x}$$

is the conditional expectation. Were the mean Green's function invariant to spatial translations, $\bar{g}(\mathbf{x},t|\mathbf{x}_0)=\bar{g}(\mathbf{x}-\mathbf{x}_0,t|\mathbf{0})$, the averages $E\{\tilde{X}_i|\mathbf{X}_0\}$, $E\{\tilde{X}_i^2|\mathbf{X}_0\}$, and $\tilde{\Sigma}_{ii}$ would be independent of $\mathbf{X}_0$, $m_{ii}\equiv 0$ and, as follows from Eq. (12), $[\Sigma_{ii}(t)-\Sigma_{ii}(0)]$ would not depend on the initial conditions. The disagreement with the numerical results presented in Fig. 2 indicates that the transition probability $\bar{g}(\mathbf{x},t|\mathbf{x}_0)d\mathbf{x}$ depends on $\mathbf{x}_0$ and the displacements $\tilde{X}_i=X_i-X_{0i}$ are correlated with the deterministic initial positions X_{0i}; that is, $m_{ii}\neq 0$.

While the memory terms necessarily vanish if the transition probability is spatially homogeneous, the converse is not always true. For deterministic initial conditions $\mathbf{X}(0)=\mathbf{x}_0$ and singular initial distributions we have $x_{0i}=E\{x_{0i}\}$ and, according to Eq. (13), $m_{ii}=0$, but this does not imply the homogeneity of the transition probability. As shown in Sec. II, this is true if and only if the displacements and the Lagrangian velocity field are statistically homogeneous, properties that can be proved for deterministic initial conditions, if the existence of unique solutions of Itô or Fokker-Planck equations is ensured. If the process $\mathbf{X}$ has finite mean and variance at finite times, then it is equivalent in the wide sense, as far as second moments are concerned, with a Gaussian process [7,13]. Hence, the translation invariant Green's function $\bar{g}(\mathbf{x}-\mathbf{x}_0,t|\mathbf{0})$ for deterministic initial conditions can be given the explicit form of the space-homogeneous Gaussian transition probability with mean $\bar{\mathbf{u}}t$ and covariance $2\int_0^t D_{ij}(t)dt$, where, because the Lagrangian velocity is statistically homogeneous, the dispersion coefficients $D_{ij}(t)$ [Eq. (7)] do not depend on $\mathbf{x}_0$.

When considering transitions between states at arbitrary times $t_1<t_2<t_3$, we are no longer in the frame of the usual setup for the statistical homogeneity. Even if $\mathbf{X}(t_1)$ were a deterministic initial position, $\mathbf{X}(t_2,\omega)$ would depend on the velocity statistics, and when calculating the displacement $\mathbf{X}(t_3,\omega)-\mathbf{X}(t_2,\omega)$ we would have under the integral in Eq. (4) a velocity field with spatial argument translated by the random quantity $\mathbf{X}(t_2,\omega)$, which is no longer a measure preserving shift. Nevertheless, random initial conditions that result from the evolution of the process may often occur in applications, as for instance, in modeling contaminant transport in hydrogeophysical environments, where in most cases only states of the system after the beginning of the contaminant event are available from observations [20].

For $t_1<t_2<t_3$, similar to Eq. (12), we have

$$\tilde{\Sigma}_{ii}(t_1,t_3) = \tilde{\Sigma}_{ii}(t_1,t_2) + \tilde{\Sigma}_{ii}(t_2,t_3) + m_{ii}(t_1,t_2,t_3), \qquad (14)$$

where

$$\tilde{\Sigma}_{ii}(t_1,t_3) = \mathrm{var}\{X_i(t_3) - X_i(t_1)\},$$

$$\tilde{\Sigma}_{ii}(t_1,t_2) = \mathrm{var}\{X_i(t_2) - X_i(t_1)\},$$

$$\tilde{\Sigma}_{ii}(t_2,t_3) = \mathrm{var}\{X_i(t_3) - X_i(t_2)\},$$

$$m_{ii}(t_1,t_2,t_3) = 2\mathrm{cov}\{[X_i(t_2) - X_i(t_1)],[X_i(t_3) - X_i(t_2)]\}.$$

The memory term $m_{ii}(t_1,t_2,t_3)$ vanishes if and only if

$$E\{[X_i(t_2) - X_i(t_1)][X_i(t_3) - X_i(t_2)]\} = E\{X_i(t_2) - X_i(t_1)\}E\{X_i(t_3) - X_i(t_2)\}.$$

Hence, if $m_{ii}(t_1,t_2,t_3)\equiv 0$ the $\mathbf{X}(t)$ process has uncorrelated increments. This is necessarily the case if the transition probabilities are spatially homogeneous and the increments of the process are independent. Conversely, if the memory terms vanish for arbitrary successive times, the process has uncorrelated increments and, as shown by Doob ([7], Theorem II.3.2), there exists a Markov process with independent increments and spatially homogeneous transitions probabilities, a wide sense version of $\mathbf{X}(t)$, which is precisely the Gaussian process with the same first and second moments. Thus, if the memory terms vanish for arbitrary successive times, the transport processes are wide-sense versions of Gaussian processes with spatially homogeneous transition probabilities. As follows from Eq. (14), vanishing memory terms and spatially homogeneous transition probabilities are also equivalent with the additivity of the variance of the increments $\tilde{\Sigma}_{ii}$ with respect to nonoverlapping time intervals. This is obviously the case of the normal diffusion process with variance linear in time.

We note that the conclusions drawn from Eq. (14) are not limited to processes of diffusion in random fields but apply as well to discrete-time processes [5] or to the Itô process described by Eq. (2) for a fixed realization of the velocity field. In the latter case, as shown by Aït-Sahalia [9], we have, in addition, the stronger result saying that the only diffusion process [Eq. (2)] with space-homogeneous transition probabilities are Brownian motions with constant drift and diffusion coefficients. It follows that in the general case with space variable drift the Markovian Itô diffusion has memory and inhomogeneous transition probabilities. Memory effects associated with correlated Markov processes also occur in the study of recurrent and extreme events, where the absence of memory correspond to the limit of independent identically distributed random variables [21]. Non-Markovian processes described by generalized Langevin equation [22], by diffusion equations with memory [23], or by fractional diffusion equations [24] are necessarily affected by memory, because the transitions between states of the process depend explicitly on past history and the corresponding probability densities cannot be spatially homogeneous. In such cases, the normal Markovian diffusion process without memory can, however, be obtained asymptotically if the temporal memory kernel and the colored noise have finite time scales [22,23].

The memory term from Eq. (14) is explicitly given by the Itô equation [Eq. (2)] as a functional of the Lagrangian velocity covariance [5],

$$m_{ii}(t_1,t_2,t_3) = 2\int_{t_2}^{t_3}\int_{t_1}^{t_2} \text{cov}\{u_i[\mathbf{X}(t')],u_i[\mathbf{X}(t'')]\}dt'dt''. \tag{15}$$

As follows from Eq. (15), the memory terms cannot vanish for arbitrary successive times and, consequently, the transition probabilities cannot be spatially homogeneous during the entire evolution of the process whenever the Lagrangian velocity has nonvanishing covariances. In the case of diffusion in random velocity fields with short-range exponential correlation considered in the numerical example from Sec. III, the long-time limit of the memory terms [Eq. (15)] vanish [5] and, since there are finite limits of the dispersion coefficients [Eq. (10)], one expects that the process $\mathbf{X}(t)$ tends toward a Markovian Gaussian diffusion process [14,17] with linear variance, independent increments, and spatially homogeneous Gaussian transition probabilities. However, at finite times the process remembers its past itinerary (nonvanishing memory terms [Eq. (15)]), as well as the deterministic initial conditions (see Fig. 2), and the transition probabilities are spatially inhomogeneous.

Indefinitely persistent memory is a characteristic of the fractional Brownian motion introduced by Mandelbrot and van Ness, which has an infinite span of interdependence between the increments of the process [25]. Successive generalizations of this process to integrals with respect to fractional Lévy motions were recently used by Marquardt [26] to construct stationary processes with continuous paths and long memory, characterized by power-law correlations. Assuming that statistically homogeneous velocity fields with power law correlations and continuous samples can be similarly constructed, we investigate in the following their impact on the transition probabilities of the transport process by using the first-order approach presented in Sec. III.

Even if there is no physical length scale for power law correlations, a finite spatial scale of the observation, λ, can be used to define the Péclet number and the small parameter $\epsilon=\text{Pe}^{-1/2}=\mathcal{O}(\sigma/U)$ so that, for small velocity fluctuations and advection dominated problems, the first iteration of the Itô equation about the mean flow trajectory (Sec. III) approximates the leading terms of the dispersion coefficients. Since in this consistent approximation the Lagrangian velocity covariance is sampled along the deterministic mean trajectory $\mathbf{X}^{(0)}(t)=(t,0,0)$, the Kubo formula [Eq. (7)] can be evaluated exactly for a power law Eulerian covariance $\text{cov}\{u_i(\mathbf{x}_1),u_i(\mathbf{x}_2)\}=\sigma^2(1+|\mathbf{x}_1-\mathbf{x}_2|/\lambda)^{-\beta}$ with $0<\beta<2$. If $\beta\neq 1$ the time integration of Eq. (7) yields the dimensionless variance $\tilde{\Sigma}_{ii}(t_2-t_1)=2(t_2-t_1)+\gamma[1+(t_2-t_1)]^{2-\beta}$ of the increment $X_i(t_2)-X_i(t_1)$, where $\gamma=2(1-\beta)^{-1}(2-\beta)^{-1}$. The memory terms can be estimated to the first order either from Eq. (15) or by using Eq. (14) and they obviously behave for large times as $m_{ii}\sim t^{2-\beta}$.

Since the consistent first-order approximation of the second term in the Itô equation [Eq. (2)] does not depend on the Wiener process $W_i(t)$, the contribution of the velocity field can be analyzed separately by considering the centered process $Y_i(t)=\gamma^{-1/2}[X_i(t)-E\{X_i(t)\}-W_i(t)]$. For $0<t_1<t_2$ and $X_i(0)=0$, the two-time correlation can be expressed as $E\{Y_i(t_1)Y_i(t_2)\}=\frac{1}{2}m_{ii}(t_1,t_2)+E\{[Y_i(t_1)]^2\}$ and from Eq. (14) one obtains the long time behavior

$$E\{Y_i(t_1)Y_i(t_2)\} \sim \frac{1}{2}[t_1^{2-\beta}+t_2^{2-\beta}-(t_2-t_1)^{2-\beta}],$$

which corresponds to a fractional Brownian motion [25–28] with Hurst coefficient $H=1-\beta/2$, $0<H<1$, $H\neq 1/2$, and variance $E\{[Y_i(t)]^2\}\sim t^{2H}$. Long-range correlations of the velocity field with exponent $0<\beta<1$ [26] induce a superdiffusive behavior of Y_i, with $1/2<H<1$, which dominates the linear term $2t$ of the Wiener process at all times; hence $X_i(t)$ is also a superdiffusion process. For $1<\beta<2$, $Y_i(t)$ is subdiffusive with Hurst coefficient $0<H<1/2$ and the linear term $2t$ dominates the long time behavior of $X_i(t)$. For the value $\beta=1$, which separates the super- and subdiffusive regimes induced by power law correlations, the integral in Eq. (7) can again be evaluated exactly and one obtains $E\{[Y_i(t)]^2\}\sim(t\ln t-t)$, which is no longer a power-law dependence. As follows from Eq. (14), the memory terms of the process $X_i(t)$ coincide with those of the process $Y_i(t)$ with nonlinear variance and are nonvanishing at all times and for the whole range of exponents, $0<\beta<2$. Consequently, the transition probabilities for diffusion in random velocity fields with power law correlations are spatially inhomogeneous. The subdiffusive case $(0<H<1/2)$ with linear long-time behavior of the variance and non-vanishing memory terms shows that using only the criterion of linear variance may be misleading in discriminating between normal and anomalous diffusion (see also [24], where a similar behavior is produced by the competition between subdiffusion and Lévy flights). These theoretical considerations are also supported by a recently reported numerical evidence of non-Markovian effects for one-dimensional fractional Brownian motions with $0.1<H<0.9$ [28].

It is useful to remark that the fractional Brownian motion behavior of the transport process described by the Itô equation [Eq. (2)] is not necessarily a consequence of power law correlations of the Eulerian velocity field. A simple example is the two-dimensional perfectly stratified aquifer model of Matheron and de Marsily, consisting of particles driven by a normal Brownian motion and a random longitudinal velocity, function of only the transverse coordinate and with a short-range correlation [29]. Multiple visits of diffusing particles across different layers with constant velocities produce a Lagrangian velocity field with long-range correlation of exponent $\beta=1/2$ [5,29]. Thus, the longitudinal dispersion coefficient [Eq. (7)] can be computed exactly and one obtains a fractional Brownian motion with Hurst coefficient $H=3/4$. For a $(d+1)$ dimensional generalization of the model, one retrieves the subdiffusive regime of Y_1 if $d>2$, and the $t\ln t$ behavior if $d=2$ [27].

V. CONCLUSIONS

We have shown that statistical homogeneity of Green's function, displacement field, and Lagrangian velocity are equivalent properties which hold for homogeneous Eulerian velocity fields and unique classical solutions of the transport equations under deterministic initial conditions. When the uniqueness condition fails, as for instance in case of exponentially correlated velocity fields with nonsmooth samples, formal expansions of the dispersion coefficients based on postulated homogeneity of the Green's function cannot be used. However, dispersion coefficients can still be estimated from the first iteration of the Itô equation about a solution independent of velocity statistics. Numerical simulations show that after a transitory regime characterized by dependence on initial conditions, indicative of statistical inhomogeneity of the mean Green's function, the dispersion coefficients reach the predicted asymptotic values.

The usual homogeneity setup no longer applies to random initial states which are the result of the past evolution of the process so that in this case we should refer to transition probabilities rather than to Green's functions, which are usually associated with deterministic initial conditions. We have shown that, as far as second moments are concerned, spatial homogeneity of the transition probability for successive transition times is equivalent to vanishing correlations of the increments of the transport process. With the possible exceptions of transitions from deterministic initial states, in case of sufficiently smooth velocity samples, the increments are correlated and the transition probabilities are spatially inhomogeneous as long as the Lagrangian velocities of the diffusing particles are correlated. For power-law correlations, the memory effects manifested by spatially inhomogeneous transition probabilities persist indefinitely. For short-range correlations instead, the memory of the initial positions is lost and the transition probabilities tend in the long-time limit to spatially homogeneous Gaussian distributions.

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