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Classical non-Markovian Boltzmann equation

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The modeling of particle transport involves anomalous diffusion, \( \langle x^2(t) \rangle \propto t^\alpha \) with \( \alpha \neq 1 \), with subdiffusive transport corresponding to \( 0 < \alpha < 1 \) and superdiffusive transport to \( \alpha > 1 \). These anomalies give rise to fractional advection-dispersion equations with memory in space and time. The usual Boltzmann equation, with only isolated binary collisions, is Markovian and, in particular, the contributions of the three-particle distribution function are neglected. We show that the inclusion of higher-order distribution functions give rise to an exact, non-Markovian Boltzmann equation with resulting transport equations for mass, momentum, and kinetic energy with memory in both time and space. The two- and the three-particle distribution functions are considered under the assumption that the two- and the three-particle correlation functions are translationally invariant that allows us to obtain advection-dispersion equations for modeling transport in terms of spatial and temporal fractional derivatives. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4886475]

I. INTRODUCTION

The Boltzmann equation forms the foundation of irreversible processes and leads to a successful description of dilute gases and the approach to equilibrium via Boltzmann’s H-theorem. The approach to equilibrium may be described by transport phenomena and the consequent three conservation theorems, viz., conservation of mass, conservation of momentum, and conservation of energy. It is important to remark that the Boltzmann equation considers only two-body collisions between initially uncorrelated particles—“Stosszahlansatz” or “molecular chaos assumption.” Therefore, the inclusion of higher-order collisions gives rise to transport equations where the conservation of mass equation does not change but both the conservations of momentum and energy will involve additional particles and so the treatment of two-body collisions that lead to local momentum and energy conservation is no longer valid.

More importantly, the usual Boltzmann equation contains one-time only and so describes a Markov process. Important physical processes, for instance, the seminal molecular dynamical simulations for hard spheres\(^1\) that showed that the two-time, velocity autocorrelation function decayed not exponentially in time but rather as a power of the time, viz., \( t^{-3/2} \). This result completely overthrew the existing kinetic theory and it requires the introduction of memory in the dynamical equations. Similarly, probability density functions show a heavy power law tail in space suggesting superdiffusive nonlocality in transport of tracers at the Earth surface.\(^2\) These important physical cases call for dynamical equations with memory, that is, non-Markovian processes, both for spatial and temporal variables. Now the case for time correlation functions has been fully addressed\(^3\) whereas that for spatial single-particle distribution functions has been addressed heuristically.\(^2\) Anomalous diffusion has been studied in the context of the continuous time random walk resulting in equations that contain fractional-order space and/or time derivatives\(^4-6\) and with the aid of random walk models, albeit with emphasis on subdiffusion.\(^7\)
It would be interesting to develop a fully non-Markovian Boltzmann equation and consequent transport equations. A non-Markovian kinetic equation has been used to determine correlation time in a non-ideal, non-equilibrium many-particle systems with binary collisions. The dynamics of a dense electron gas has been used, with the aid of a quantum non-Markovian kinetic equation, to study the relevance of retardation (memory) effects, energy broadening, and correlation build-up for femtosecond relaxation processes. Molecular dynamics simulations of quasi two-dimensional Yukawa liquids have shown the emergence and vanishing of superdiffusion.

The treatment of kinetic equation for strongly interacting particles has been derived in the framework of the density operator technique with the aid of a quantum non-Markovian equation. The latter is based on a generalized binary collision approximation, ladder or T-matrix approximation in order to retain full time dependencies, especially also on short time scales, thus including retardation and memory effects resulting from the dynamics of binary correlations and initial correlations. The method applies also to strongly correlated many-particle system given rise to a general quantum kinetic equation. The generalized T-matrix approximation incorporates initial binary correlations into the Kadanoff-Baym equations.

In what follows, the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy is used to derive a classical non-Markovian Boltzmann equation and the consequent transport equations for the conservation of mass, linear momentum, and energy. It is shown that higher order distribution functions do not modify the form of the transport equation associated with the conservation of mass but do lead to explicit memory terms in time and space in the transport equations for both the linear momentum and the kinetic energy. In the spirit of deriving fractional advection-dispersion equations for modeling transport, we suppose that the two- and three-particle correlation functions are translationally invariant and so express memory effects in terms of fractional derivatives.

II. THE BBGKY HIERARCHY

In a classical system of $N$ particles interacting via two-body forces and in the presence of external fields, the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy relates successive orders of the distribution function $f_s$, where $f_s$ gives the probability of finding $s$ particles at a given time with specific positions $\mathbf{r}_i$ and momenta $\mathbf{p}_i$ with $i = 1, 2, \ldots, s$.

Liouville’s theorem, viz., $d\rho/dt = 0$, together with Hamilton’s equations of the motion, can be cast into the form

$$\left[\frac{\partial}{\partial t} + \hat{h}_s\right]\rho(\mathbf{z}_1, \ldots, \mathbf{z}_N, t) = 0,$$

where the operators

$$\hat{h}_s = \sum_{i=1}^{s} \hat{S}_i + \frac{1}{2} \sum_{i,j=1, i \neq j}^{s} \hat{P}_{ij},$$

and

$$\hat{S}_i \equiv \frac{\mathbf{p}_i}{m} \cdot \nabla_{\mathbf{r}_i} + \mathbf{F}_i \cdot \nabla_{\mathbf{p}_i},$$

$$\hat{P}_{ij} \equiv \mathbf{K}_{ij} \cdot \nabla_{\mathbf{p}_i} + \mathbf{K}_{ji} \cdot \nabla_{\mathbf{p}_j} = \hat{P}_{ji},$$

where $i, j, s = 1, \ldots, N$.

In particular,

$$\left(\frac{\partial}{\partial t} + \hat{h}_s\right)f_s(\mathbf{z}_1, \ldots, \mathbf{z}_s, t) =$$

$$-\sum_{i=1}^{s} \int d\mathbf{z}_{s+1} \mathbf{K}_{i,s+1} \cdot \nabla_{\mathbf{p}_i} f_{s+1}(\mathbf{z}_1, \ldots, \mathbf{z}_{s+1}, t),$$

$$f_s = f_s(\mathbf{z}_1, \ldots, \mathbf{z}_s, t),$$

$$s = 1, 2, \ldots, N,$$
where \( z_i = (p_i, r_i) \), \( \int dz_i = \int d^3p_i d^3r_i \) and \( K_{ij} = -\nabla_{r_i} v(|r_i - r_j|) = -K_{ji} \), where \( v(r) \) is the two-particle potential and \( F_i \) is the external force acting on the \( i \)th particle.

The general \( s \)-particle distribution function is defined by

\[
\hat{f}_s(z_1, \ldots, z_s, t) \equiv \frac{N!}{(N - s)!} \int dz_{s+1} \ldots dz_N \rho(z_1, \ldots, z_N, t),
\]

where \( \rho(z_1, \ldots, z_N, t) \) is the density function characterizing the ensemble and \( \int dz_1 \ldots dz_N \rho(z_1, \ldots, z_N, t) = 1 \).

### III. NON-MARKOVIAN BOLTZMANN EQUATION

The single-particle \( f_1(z_1, t) \) and the two-particle \( f_2(z_1, z_2, t) \) distribution functions satisfy equations that follow from Eq. (4),

\[
\left( \frac{\partial}{\partial t} + \hat{D}_1 \right) f_1(z_1, t) = \hat{I}_1 f_2(z_1, z_2, t)
\]

and

\[
\left( \frac{\partial}{\partial t} + \hat{D}_2 \right) f_2(z_1, z_2, t) = \hat{I}_2 f_3(z_1, z_2, z_3, t),
\]

with the differential operators

\[
\hat{D}_1 = \hat{S}_1,
\]

\[
\hat{D}_2 = \hat{S}_1 + \hat{S}_2 + \hat{P}_{12},
\]

and the integral operators defined by

\[
\hat{I}_1 = - \int dz_3 K_{12} \cdot \nabla_{p_1},
\]

\[
\hat{I}_2 = - \int dz_3 (K_{13} \cdot \nabla_{p_1} + K_{23} \cdot \nabla_{p_2}).
\]

The solution of Eq. (7) is obtained via Laplace transforms and so Eq. (6) becomes

\[
\left( \frac{\partial}{\partial t} + \hat{D}_1 \right) f_1(z_1, t) = \hat{I}_1 \int_0^t d\tau e^{-\hat{D}_1(t-\tau)} \cdot [f_2(z_1, z_2, \tau) \delta(\tau) + \hat{I}_2 f_3(z_1, z_2, z_3, \tau)].
\]

The non-Markovian behavior for the single-particle distribution function \( f_1(z_1, t) \) follows from higher-order particle distribution functions in Eq. (10). If, however, the contribution of the three-particle distribution function is neglected in (7), then the Dirac \( \delta \)-function term in Eq. (10) gives rise to the usual Markovian Boltzmann equation,\(^8\) when \( \delta f_2(z_1, z_2, t)/\partial t = 0 \), since \( f_2(z_1, z_2, t) \) has a shorter time scale than \( f_1(z_1, t) \) thus reaching equilibrium earlier than \( f_1(z_1, t) \), and so \( \hat{D}_2 f_2(z_1, z_2, t) = 0 \) from Eq. (7). A more rigorous derivation of the usual Markovian Boltzmann equation follows by taking the classical limit of the quantum Boltzmann equation.\(^9\)

All three conservation laws for binary collisions, viz., mass, momentum, and thermal energy, where only two particles are involved in a collision, follow from the usual Markovian Boltzmann equation. The latter need not be so for the non-Markovian Boltzmann equation.\(^8\)\(^9\)
IV. TRANSPORT EQUATIONS

On integrating Eq. (6) over all momenta \( p_1 \), one obtains, owing to the divergence in \( I_1 \), the usual continuity equation

\[
\frac{\partial \rho(r, t)}{\partial t} + \nabla \cdot \{ \rho(r, t) u(r, t) \} = 0,
\]

(11)

where

\[
\rho(r, t) = m \int dp f(r, p, t),
\]

\[
\rho(r, t) u(r, t) = \int dp p f(r, p, t),
\]

(12)

\( f(r, p, t) \equiv f_1(z_1, t) \) and \( u(r, t) \equiv \langle v \rangle \).

Multiplying Eq. (10) by \( p_1 \) and integrating over all momenta, where we have set all external forces to zero, for simplicity, one obtains, after an integration by parts,

\[
\int_0^t d\tau e^{\frac{1}{m} \Delta \tau} \cdot [f_2(z_1, z_2, \tau) \delta(\tau) + \dot{I}_2 f_3(z_1, z_2, z_3, \tau)],
\]

\[
= \int dz_2 dp_1 K_{12}(r_2) \int_0^t d\tau \times \left\{ e^{-(t-\tau)(X+Y)} f_2(r, p_1; r + r_2, p_2; \tau) \delta(\tau) + \frac{(t-\tau)}{m} \int dz_3 K_{13}(r_3) \cdot \nabla r \times e^{-(t-\tau)(X+Y)} f_3(r, p_1; r + r_2, p_2; r + r_3, p_3; \tau) + \frac{(t-\tau)}{m} \int dz_3 K_{23}(r_3) \cdot \nabla r_2 \times e^{-(t-\tau)(X+Y)} f_3(r, p_1; r + r_2, p_2; r + r_2 + r_3, p_3; \tau) \right\},
\]

(13)

where the pressure tensor

\[
(\dot{\mathbf{P}})_{ij} = P_{ij} = \rho(r, t)(v_i - u_i)(v_j - u_j)
\]

\[= m \int dp (v_i - u_i)(v_j - u_j) f(r, p, t),
\]

(14)

\( K_{ij}(r) = -\nabla_i \langle v_j \rangle, \nabla_i \langle \dot{v}_j \rangle = \sum \frac{\partial P_{ij}}{\partial x_i} \) and the operators \( X \) and \( Y \) are defined by Eq. (A5) and Eq. (A6), respectively. The second equality in Eq. (13) follows from Eq. (A4) and after integrating over the momenta \( p_1 \) and \( p_2 \).

Note that if the contributions of \( f_3(z_1, z_2, z_3, t) \) are neglected on the right-hand side of (13) and \( \partial f_2(z_1, z_2, t)/\partial t = 0 \) from Eq. (7) and so the right-hand side of Eq. (13) becomes

\[
\int dz_2 dp_1 K_{12} f_2(z_1, z_2, t) = \int d\mathbf{r}_2 K_{12} g_2(\mathbf{r}_1, \mathbf{r}_2, t) = 0,
\]

(15)
K_{12}$ is a central force and for fluids the two-particle correlation function $g_2(r_1, r_2, t) = g_2(|r_1 - r_2|, t)$. Therefore, one has momentum conservation for binary collisions as in the usual Markovian Boltzmann equation.

Finally,

$$\frac{\partial [\rho(r, t)u_r(r, t)]}{\partial t} + \frac{1}{m} \nabla \cdot \mathbf{P} = \int_0^t \int d\tau \int d\zeta_2 d\mathbf{p}_1 \mathbf{K}_{12}(\mathbf{r}_2) \times \\left\{ e^{-(t-\tau)X} f_2(\mathbf{r} + \mathbf{D}(t - \tau), \mathbf{p}_1 + \mathbf{P}(t - \tau); \mathbf{r} + \mathbf{r}_2 + \mathbf{D}(t - \tau), \mathbf{p}_2; \right\}$$

with the aid of Eq. (A17), where the displacement $\mathbf{D}(t)$ is given by Eq. (A18) and the momentum $\mathbf{P}(t)$ by Eq. (A19) and $e^{B(t)}_{\nu_1} e^{P(t)}_{\nu_2} g(\mathbf{r}_1, \mathbf{p}_1) = g(\mathbf{r}_1 + \mathbf{D}(t), \mathbf{p}_1 + \mathbf{P}(t))$ for arbitrary function $g(\mathbf{r}_1, \mathbf{p}_1)$.

In a similar fashion, one can derive the transport equation for the temperature (kinetic energy) and one obtains,

$$\rho(r, t) \left( \frac{\partial}{\partial t} + \mathbf{u}(r, t) \cdot \nabla \right) \theta(r, t) = \frac{2}{3} \nabla \cdot \mathbf{q}(r, t) + \frac{2}{3} \mathbf{P} \cdot \dddot{\mathbf{r}}$$

where the content inside the braces in Eq. (17) is precisely the same as that appears in Eq. (16). $\mathbf{P} \cdot \dddot{\mathbf{r}} = \sum_{i,j} P_{ij} \Lambda_{ij}$ with the temperature $k_B \theta(\mathbf{r}, t) = \frac{1}{2} m \left( |\mathbf{v} - \mathbf{u}|^2 \right)$, heat flux vector $\mathbf{q}(\mathbf{r}, t) = \frac{1}{2} m \rho \mathbf{u}((\mathbf{v} - \mathbf{u})^2 - |\mathbf{u}|^2)$, and $\Lambda_{ij} = \frac{1}{2} m (\partial u_i / \partial x_j + \partial u_j / \partial x_i)$.

The right-hand side of Eq. (17) vanishes in the usual Boltzmann equation and so one obtains the conservation of thermal energy. However, in general, such is not the case for the non-Markovian Boltzmann equation. Note, however, that the conservation of total (the sum of kinetic and potential) energy holds true for the non-Markovian Boltzmann equation while the usual Boltzmann equation conserves only the kinetic energy. The ensemble average of the Hamiltonian is

$$\langle H \rangle = \int d\zeta_1 \frac{P_1^2}{2m} f_1(z_1, t) + \frac{1}{2} \int d\zeta_1 d\zeta_2 v_{12} f_2(z_1, z_2, t),$$

where $v_{12}$ is the interparticle potential. It can be shown, with the aid of the (6) and (7) that $dH/dt = 0$ after coordinate and momenta integrations and so one has total energy conservation as expected.

The non-Markovian nature of the integro-differential equations (16) and (17) shows up both in the temporal as well as in the spatial behavior of the left-hand side of (16) and (17). Note that the two-particle distribution function $f_2(z_1, z_2, t)$ and three-particle distribution functions $f_3(z_1, z_2, z_3, t)$, where $z_i = (\mathbf{p}_i, \mathbf{r}_i)$, must be known for earlier times $\tau \leq t$ and spatial coordinates $\mathbf{r} + \mathbf{D}(t - \tau)$ also for $\tau \leq t$. In the succeeding section, the two- and the three-particle distribution functions are expressed in terms of the two- and the three-particle correlation functions so that the nonlocality
in both the time and the spatial coordinates may be transferred to the single-particle distribution function.

V. CORRELATION FUNCTIONS

The transport equations (11), (16), and (17) are exact. The memory effects in the conservation of mass equation (11) are contained in its coupling to Eq. (16). The momentum conservation equation (16) depends on the two- and the three-particle distribution functions, where the latter is related, in turn, to the four- and higher-particle distribution functions up to the full N-body correlation function $f_N$ via the hierarchy given by Eq. (4). Note that the effect of memory comes in Eqs. (16) and (17) from both $f_2(z_1, z_2, t)$ and $f_3(z_1, z_2, z_3, t)$.

Attempts to express memory effects in terms of fractional derivatives or integrals of the single-particle distribution function must be based on introducing the correlation functions $g_2(z_1, z_2, t)$ and $g_3(z_1, z_2, z_3, t)$ defined by

$$f_2(z_1, z_2, t) = f_1(z_1, t)f_1(z_2, t) + g_2(z_1, z_2, t)$$

and

$$f_3(z_1, z_2, z_3, t) = f_1(z_1, t)f_1(z_2, t)f_1(z_3, t) + f_1(z_1, t)g_2(z_2, z_3, t) + f_1(z_2, t)g_2(z_1, z_3, t) + f_1(z_3, t)g_2(z_1, z_2, t) + g_3(z_1, z_2, z_3, t).$$

In the limit when the particles are far apart, (19) reduces to the product of single-particle distribution functions in accordance to the principle of correlation damping. One may consider, for simplicity, that the force $K$ vanishes outside a certain range $r_0$ and so the spatial integrals over the variables $r_2$ and $r_3$ of the second term in Eq. (10), which gives rise to memory effects, are subject to $|r_1 - r_2| < r_0$ and $|r_1 - r_3| < r_0$. One expects no correlations between particles $i$ and $j$ when $|r_i - r_j| \gg r_0$. Therefore, in the integral over $r_2$, for instance, one needs $g_2(z_1, z_2, t)$ not in the uncorrelated region $|r_1 - r_2| \gg r_0$, but rather in the region where the two particles are actually colliding and thus correlated, viz. $|r_1 - r_2| < r_0$.

Consider the first term inside the braces of the right-hand side of Eq. (16) that contains the two-particle distribution function, which is integrated over $p_1$, $r_2$, and $p_2$. Therefore, one has from (19) that

$$f_2(r + D(t), p_1 + P(t); r + r_2 + D(t), p_2; 0) = f_1(r + D(t), p_1 + P(t); 0)f_1(r + r_2 + D(t), p_2; 0) + g_2(r + D(t), p_1 + P(t); r + r_2 + D(t), p_2; 0).$$

Consider the second term inside the braces of the right-hand side of Eq. (16) that contains the three-particle distribution function, which is integrated over $p_1$, $r_2$, $p_2$, $r_3$, and $r$. Therefore, one has the following five terms corresponding to the five terms given in Eq. (19) with the first term given by

$$f_1(r + D(t - \tau), p_1 + P(t - \tau); r) \times f_1(r + r_2 + D(t - \tau), p_2; \tau)f_1(r + r_3 + D(t - \tau), p_3; \tau).$$

The second term in (19) given by

$$f_1(r + D(t - \tau), p_1 + P(t - \tau); r) \times g_2(r + r_2 + D(t - \tau), p_2; r + r_3 + D(t - \tau), p_3; \tau).$$
The third term in (19) given by

\[ f_1(r + r_2 + D(t - \tau), p_2; \tau) \]

\[ \times g_2(r + D(t - \tau), p_1 + P(t - \tau); r + r_3 + D(t - \tau), p_3; \tau). \]

The fourth term in (19) given by

\[ f_1(r + r_3 + D(t - \tau), p_3; \tau) \]

\[ \times g_2(r + D(t - \tau), p_1 + P(t - \tau); r + r_2 + D(t - \tau), p_2; \tau). \]

Finally, the fifth term in (19) given by

\[ g_3(r + D(t - \tau), p_1 + P(t - \tau); r + r_2 + D(t - \tau), p_2; \tau; \tau) \]

\[ r + r_3 + D(t - \tau), p_3; \tau). \]

Similar expressions also result for the third term inside the braces of the right-hand side of Eq. (16) that contains the three-particle distribution function, which is integrated also over \( p_1, r_2, p_2, r_3, p_3, \) and \( \tau \).

The transport equations (16) and (17), together with expressions (19)–(25), are exact.

VI. FRACTIONAL ADVECTION-DISPERSION EQUATIONS

The appearance of memory in Eq. (11) occurs via the function \( \rho(r, t)u(r, t) \), which is determined by Eq. (16). In order to obtain fractional-in-space and fractional-in-time advection-dispersion differential equations for \( \rho(r, t) \), one has to expand it in a Taylor-Riemann series for fractional derivatives the single-particle distribution function in both the two- and three-particle distribution functions appearing in Eqs. (20)–(24). Note, however, that all such terms involve the product of a single-particle distribution function multiplied by a function of \( r \). On expanding the single-particle distribution function one would obtain, for instance, diffusion coefficients that would depend on \( r \). Therefore, in order to obtain equations for fractional advection-dispersion equations modeling transport, the two-particle correlation function \( g_2 \) is assumed to be translationally invariant. Accordingly, all the two-particle correlation functions \( g_2 \) appearing in Eq. (20) and Eqs. (22)–(24) are actually independent of \( r \).

Closure relations are introduced to truncate the hierarchy (4) and are based on finding appropriate approximations for the three-particle correlation function \( g_3 \). A commonly used approximation is to set \( g_3 = 0 \). That is, the many-particle system is governed solely by the one- and two-particle distribution functions. However, the latter closure relation does not account for the interaction of particle 1 and particle 2 with the surrounding media. In an effort of take \( g_3 \) partially into account in the transport equation (16), one may suppose that the three-particle correlation function \( g_3 \) is translationally invariant. Hence, \( g_3 \) in (25) is independent of \( r \) and so contributes a function of time in the transport equation (16) and, owing to the gradient in the transport equation (11), makes no contribution to the transport equation (11). However, it does contribute to nonlocal effect in time (see Eq. (35) below).

The contributions to Eq. (16) of the two-particle distribution function of Eq. (20) and the three-particle distribution function indicated in Eq. (19) and Eqs. (21)–(25) are quite different owing to the integration over the variables \( p_1, z_2 \) for the term given by Eq. (20) and integration over the variables \( p_1, z_2, z_3, \) and \( \tau \) for the terms given by Eqs. (21)–(25). The single-particle distribution function in Eq. (23) depends on \( r_2 \) and so is acted on by the exponential operator \( e^{-t-t/3X} \), where the operator \( X \) is given by (A5). In addition, one has an integration over \( r_2 \). This is also the case for the term given by Eq. (24), which is integrated over \( r_3 \). This is not the case for the single-particle distribution function in Eq. (22), which is a function of \( r \) and is integrated only over the momentum \( p_1 \). Both terms associated with Eqs. (20) and (21) always give rise to a single-particle distribution function in the variable \( r \) multiplied by a function of \( r \). In addition, the correlation function in
Eq. (20) gives rise to only a function of time owing to the translational invariance of $g_2$. Therefore in what follows, we will consider only the term given by Eq. (22) in deriving the fractional in time and space, advection-dispersion equations.

If the mass density $\rho(\mathbf{r}, t)$ depends only on one spatial dimension, then the mass conservation Eq. (11) can be written as

$$\frac{\partial C(x, t)}{\partial t} = -\frac{\partial F(x, t)}{\partial x}, \quad \text{(26)}$$

where $C(x, t)$ is the particle concentration, mass per volume, and $F(x, t)$ is the flux, mass per area per time. Equation (26) is a Markovian equation which remains so when the flux $F(x, t)$ is assumed to be given by an expansion of the form

$$F(x, t) = a(t)C(x, t) + b(t)\frac{\partial C(x, t)}{\partial x} + \cdots. \quad \text{(27)}$$

Keeping the first two leading terms in expansion (27), Eq. (26) becomes

$$\frac{\partial C(x, t)}{\partial t} = -v\frac{\partial C(x, t)}{\partial x} + D\frac{\partial^2 C(x, t)}{\partial x^2}, \quad \text{(28)}$$

where we have supposed $a(t)$ and $b(t)$ to be actually independent of time and to correspond to the constants $a = v$, the average particle velocity $v$, and $b = -D$, with $D$ the dispersion coefficient. Equation (28) is known as the Fokker-Planck equation with both drift and Fick diffusion terms. Note that the partial differential equation (28) is local in both $x$ and $t$ since it involves partial derivatives in the respective variables, which require only knowledge of $C(x, t)$ in an arbitrarily small neighborhood of the point $(x, t)$ in question.

Let us cast the assumptions that gave rise to the differential equation (28) for $C(x, t)$ in terms of the exact expression (16) for the time rate of change of the flux $\frac{\partial F(x, t)}{\partial t}$ and find what constraints are placed on the terms appearing in Eq. (16). Clearly it is difficult, if not impossible, to obtain a differential equation for the particle concentration $C(x, t)$ from the exact equations (11) and (16) without assuming the translational invariance of the two-particle correlation function $g_2$ as done in Eq. (20) and Eqs. (22)–(24) as explained above. In addition, certain terms must be equated in order to reproduce the form (27) for the flux $F(x, t)$.

The quantity to be expanded, as explained above, is the single-particle distribution function that appears in Eq. (22) and so

$$f_1(\mathbf{r} + D(t - \tau), \mathbf{p}_1 + \mathbf{P}(t - \tau); \tau) = f_1(\mathbf{r}, \mathbf{p}_1; t)$$

$$+ (D(t - \tau) \cdot \nabla_r) f_1(\mathbf{r}, \mathbf{p}_1; t) + (\mathbf{P}(t - \tau) \cdot \nabla_{p_1}) f_1(\mathbf{r}, \mathbf{p}_1; t)$$

$$+ (\tau - t) \frac{\partial f_1(\mathbf{r}, \mathbf{p}_1; t)}{\partial t} + (\tau - t) (D(t - \tau) \cdot \nabla_r) \frac{\partial f_1(\mathbf{r}, \mathbf{p}_1; t)}{\partial t}$$

$$+ (D(t - \tau) \cdot \nabla_r) \xi \frac{\partial f_1(\mathbf{r}, \mathbf{p}_1; t)}{\partial t} + (\tau - t)\xi \frac{\partial^\gamma f_1(\mathbf{r}, \mathbf{p}_1; t)}{\partial t^\gamma}$$

$$+ (\tau - t) (D(t - \tau) \cdot \nabla_r) \xi \frac{\partial f_1(\mathbf{r}, \mathbf{p}_1; t)}{\partial t} \cdots,$$

where the last three terms in (29) follow from the fractional Taylor series (B9)–(B10) with $1 < \alpha \leq 2$ and $1 < \gamma \leq 2$.

On substituting Eq. (29) in Eq. (22), where $g_2$ is assumed to be translationally invariant, and the resulting equation into Eq. (16) one obtains, by equating terms that do not contain a partial derivative with respect to time and on integrating over $\mathbf{p}_1$, that

$$\rho(\mathbf{r}, t) \mathbf{u}(\mathbf{r}, t) = \rho(\mathbf{r}, t) \mathbf{a} + (\mathbf{b} \cdot \nabla_r \rho(\mathbf{r}, t)) \mathbf{c}$$

$$+ \nabla_r \cdot (\rho(\mathbf{r}, t) \mathbf{u}(\mathbf{r}, t)) \mathbf{d} + \cdots,$$
where \( a, b, c, \) and \( d \) are constant vectors. The terms corresponding to fractional derivatives in (29) will be considered in Subsections VI A and VI B. The third term in the right-hand side of Eq. (30) follows from the linear term in \( p_1 \) that appears in expression (A18) for \( D(t) \). In the case of one spatial direction, the first two terms in Eq. (30) coincide with the first two terms of Eq. (27).

A. Fractional-in-space, advection-dispersion equation

The nonlocality in space and time contained in Eqs. (11) and (16) has been replaced by local operations in Eq. (30). It is clear that one can retain the nonlocal properties of Eq. (11) in terms of derivatives provided the ordinary Taylor expansion used in obtaining Eq. (30) be replaced by the generalization of the Taylor-Riemann series for fractional derivatives as indicated in Appendix B. Note that whereas ordinary derivatives are local, fractional derivatives are nonlocal, that is, require knowledge of the function on a set of finite measure rather than a null set as is the case for ordinary derivatives.

It is important to remark that different fractional Taylor series give rise to differing fractional differential equations that represent spatial and/or temporal nonlocality. For instance, using the Taylor-Riemann series (B1), where for fractional differential equations that represent spatial and/or temporal nonlocality. For instance, using the derivatives. Knowledge of the function on a set of finite measure rather than a null set as is the case for ordinary derivatives.

The nonlocality in space and time contained in Eqs. (11) and (16) has been replaced by local operations in Eq. (30). It is clear that one can retain the nonlocal properties of Eq. (11) in terms of fractional derivatives provided the ordinary Taylor expansion used in obtaining Eq. (30) be replaced by the generalization of the Taylor-Riemann series for fractional derivatives as indicated in Appendix B. Note that whereas ordinary derivatives are local, fractional derivatives are nonlocal, that is, require knowledge of the function on a set of finite measure rather than a null set as is the case for ordinary derivatives.

B. Fractional-in-time, advection-dispersion equation

Nonlocal effects in time are used, for instance, to describe subdiffusion, slow stochastic particle transport, by equations of the form

\[
\frac{\partial C(x,t)}{\partial t} = -v \frac{\partial C(x,t)}{\partial x} + D \frac{\partial^{\alpha} C(x,t)}{\partial x^{\alpha}},
\]

where \( \alpha > 1 \). Expression (31) represents the contribution coming essentially from the third term of the right-hand side of Eq. (16). The second term in Eq. (16) contains the gradient \( \nabla_r \) which would result in additional terms with spatial derivatives of order higher than two.

The spatial Fourier transform of (31) gives

\[
\frac{\partial \hat{C}(k,t)}{\partial t} = -v(k)^{\alpha-1} \hat{C}(k,t) + D(k)^{\alpha} \hat{C}(k,t),
\]

where \( \partial^{\alpha} \exp(vx)/\partial x^{\alpha} = v^\alpha \exp(vx) \) and so the characteristic function is given by

\[
\hat{C}(k,t) = \exp[-v(k)^{\alpha-1} +Dt(k)^{\alpha}].
\]

Comparison with the characteristic function of the normal (or Gaussian) distribution, viz., \( q_{\mu,\sigma}(k) = \exp[-i\mu k + (i\sigma k/\sqrt{2})^2] \) with mean \( \mu \) and variance \( \sigma^2 \), one has from (33) a mean \( \mu, vt = \mu^{\alpha-1} \), and a deviation \( \sigma, Dt = (\sigma/\sqrt{2})^{\alpha} \), where one has \( \alpha > 2 \) for subdiffusion and \( 1 < \alpha < 2 \) for superdiffusion.

On the other hand, the Jumarie Taylor series, owing to the ordinary derivative(s) that appear in Eqs. (B9)–(B10), can give rise to spatial nonlocality of a fractional Fick’s law with \( D = (\sigma/\sqrt{2})^{\alpha} \) with \( \alpha > 0 \) but to nonfractional advection of the form

\[
\frac{\partial C(x,t)}{\partial t} = -v \frac{\partial C(x,t)}{\partial x} + D \frac{\partial^{\alpha} C(x,t)}{\partial x^{\alpha}},
\]

where the two terms on the right-hand side of Eq. (34) follow from the fourth and eighth terms in Eq. (29). Subdiffusion occurs for \( \alpha > 2 \) and superdiffusion for \( 0 < \alpha < 2 \).

As discussed after Eq. (31), the contributions in (34) come from the third term of the right-hand side of Eq. (16). The fractional derivatives in Eq. (29) that ought to appear in Eq. (30) will be considered in Subsection VI B when considering both spatial and temporal fractional derivatives in the transport equations.

B. Fractional-in-time, advection-dispersion equation

Nonlocal effects in time are used, for instance, to describe subdiffusion, slow stochastic particle transport, by equations of the form

\[
\frac{\partial^\gamma C(x,t)}{\partial t^\gamma} = -v \frac{\partial C(x,t)}{\partial x} + D \frac{\partial^2 C(x,t)}{\partial x^2},
\]
where $0 < \gamma < 1$. However, such equations cannot follow from the continuity equation (11) and the flux transport equation (16) with the aid of (29) since any Taylor expansion of the flux will be accompanied by a spatial derivative in Eq. (11) and so there is no way of getting rid of such mixed space and time derivatives in order to give rise to Eq. (35).

The derivation of equations of the form (35), which may also include fractional spatial derivatives, must rely solely on the momentum conservation equation (16), where the term $\partial \rho(\mathbf{r}, \mathbf{u}(\mathbf{r}, t))/\partial t$ can be canceled on both sides of Eq. (16). Accordingly, one obtains

$$\frac{\partial^\gamma C(x, t)}{\partial t^\gamma} = -\frac{\partial C(x, t)}{\partial x} + D \frac{\partial^\alpha C(x, t)}{\partial x^\alpha}, \quad (36)$$

where $\gamma > 0$ and $\alpha > 1$. Note that if the $\partial \rho(\mathbf{r}, \mathbf{u}(\mathbf{r}, t))/\partial t$ term does not cancel on both sides of Eq. (16), then one obtains a limit of waiting time that converges to a Lévy motion$^{18}$ with index $1 < \gamma < 2$ and jump sizes symmetric and heavy tailed with index $1 < \alpha \leq 2$, that is, with a term $\partial C(x, t)/\partial t$ added to Eq. (36).

VII. SUMMARY AND CONCLUSION

The integro-differential transport equation for the conservation of momentum, Eq. (16), explicitly depends on the temporal and spatial history of the two- and three-particle distribution function and so gives rise to memory in time and space. The usual Boltzmann equation is obtained by neglecting the contribution of the three-particle distribution function and if the Markov limit is taken for the solution of the two-particle correlation function.$^{12}$

In the spirit of obtaining memory effects in the form of fractional derivatives that lead to fractional advection-dispersion equations for modeling transport, one considers translationally invariant correlation functions. The single-particle distribution function is expanded in a fractional Taylor series whereby the integro-differential equation becomes a fractional transport equation with possible nonlocality in both time and space.

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APPENDIX A: “MOMENTUM INTEGRAL”

The integral $I(\mathbf{r}, t)$ on the right-hand side of Eq. (13) can be considerably simplified by changing the integrated spatial variables $\mathbf{r}_2$ and $\mathbf{r}_3$. We will, for simplicity, set all external forces on particles equal to zero, that is, $F_i = 0$ for all $i$'s. Thus,

$$I(\mathbf{r}, t) = \int dz_2 dp_1 K_{12}(\mathbf{r}_2) \int_0^t d\tau \exp \left[ - (t - \tau)(v_1 \cdot \nabla_r + v_2 \cdot \nabla_{r_2} + K_{12}(\mathbf{r}_2) \cdot \nabla_{p_1} + K_{21}(\mathbf{r}_2) \cdot \nabla_{p_2}) \right]$$

$$\times \left\{ f_2(\mathbf{r}, p_1; \mathbf{r} + \mathbf{r}_2, p_2; \tau) \delta(\tau) \right\}$$

$$\left\{ - \int dz_3 K_{13}(\mathbf{r}_3) \cdot \nabla_{p_1} f_3(\mathbf{r}, p_1; \mathbf{r} + \mathbf{r}_2, p_2; \mathbf{r} + \mathbf{r}_3, p_3; \tau) \right\}.$$

Now for any two operators $\hat{A}$ and $\hat{B}$, one has that

$$\exp(A) \hat{B} \exp(\hat{A}) = \hat{B} + [\hat{A}, \hat{B}] + \frac{1}{2!} [\hat{A}, [\hat{A}, \hat{B}]] + \cdots . \quad (A2)$$
Hence,
\[
\exp \left[ -(t - \tau) \left( v_1 \cdot \nabla r_1 + v_2 \cdot \nabla r_2 + K_{12} (r_2) \cdot \nabla p_i + K_{21} (r_2) \cdot \nabla p_i \right) \right] 
\times (K_{i3} (r_3) \cdot \nabla p_i) \exp \left[ (t - \tau) \left( v_1 \cdot \nabla r_1 + v_2 \cdot \nabla r_2 + K_{12} (r_2) \cdot \nabla p_i \right) \right] 
+ K_{21} (r_2) \cdot \nabla p_i \right] 
= K_{i3} (r_3) \cdot \nabla r_i ,
\]
where \( v_i = p_i / m \) and \( i = 1, 2, \ldots \).

Therefore, Eq. (A1) becomes
\[
I(r, t) = \int d\mathbf{z} \, dp_1 \, K_{12} (r_2) \int_0^t d\tau 
\times \left\{ e^{\xi (\tau)} f_2 (r, p_1; r + r_2, p_2; \tau) \delta (\tau) \right\} 
\right. 
- \int d\mathbf{z} \left( K_{i3} (r_3) \cdot \nabla p_i + \frac{(t - \tau)}{m} K_{i3} (r_3) \cdot \nabla r_i \right) 
\times e^{\xi (\tau)} f_3 (r, p_1; r + r_2, p_2; \tau) 
\int d\mathbf{z} \left( K_{23} (r_2) \cdot \nabla p_i + \frac{(t - \tau)}{m} K_{23} (r_2) \cdot \nabla r_i \right) 
\times e^{\xi (\tau)} f_3 (r, p_1; r + r_2, p_2; \tau) \right\},
\]
where
\[
X = \frac{p_2}{m} \cdot \nabla r_2 + K_{21} (r_2) \cdot \nabla p_2 
\]
and
\[
Y = \frac{p_1}{m} \cdot \nabla r_1 + K_{12} (r_2) \cdot \nabla p_1 .
\]

The Zassenhaus formula gives,\(^1^9\) for any two operators \( X \) and \( Y \), that
\[
e^{iX} e^{-(t + \tau)} X = e^{-iY} e^{-\frac{\tau}{m} [X, X] + \frac{1}{4} [X, [X, Y]]} 
\times e^{-\frac{\tau}{m} ![([X, X], X) + 3([Y, X], X)] + \frac{1}{4} [[X, [X, Y]], Y]} \times \ldots .
\]

Now for the operators given by Eqs. (A5)–(A6), one has that
\[
[X, Y] = \left( \frac{p_2}{m} \cdot \nabla r_2 \right) (K_{12} (r_2) \cdot \nabla p_i) ,
\]
\[
[X, [X, Y]] = \left( \frac{p_2}{m} \cdot \nabla r_2 \right) ^2 (K_{12} (r_2) \cdot \nabla p_i) ,
\]
\[
+ \frac{1}{m} (K_{21} (r_2) \cdot \nabla r_i) (K_{12} (r_2) \cdot \nabla p_i) ,
\]
\[
[Y, [X, Y]] = - \left( \frac{p_2}{m^2} \cdot \nabla r_2 \right) (K_{12} (r_2) \cdot \nabla r_i) ,
\]
\[
[[[X, Y], X], X] = \frac{1}{m^3} \left( \frac{p_2}{m^2} \cdot \nabla r_2 \right) ^3 (K_{12} (r_2) \cdot \nabla p_i) .
\]
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+ \frac{1}{m^2} (p_2 \cdot \nabla_{r_2}) (K_{21}(r_2) \cdot \nabla_{r_2}) (K_{12}(r_2) \cdot \nabla_{p_2})

+ \frac{2}{m^2} (K_{21}(r_2) \cdot \nabla_{r_2}) (p_2 \cdot \nabla_{r_2}) (K_{12}(r_2) \cdot \nabla_{p_2}).

\text{([[X, Y], X], Y) = ([[X, Y], Y], Y) = 0.}

Note that all the non-zero commutators in Eq. (A8) are given by the scalar product of a vector with either \( \nabla_{p_i} \) or \( \nabla_{r_i} \). In fact, this is so for all the non-zero commutators that appear in the exponential functions on the right-hand side of Eq. (A7). Therefore, Eq. (A7) is of the form

\[ e^{tY} e^{-t(X+Y)} = e^{d(t) \cdot \nabla_{r_1} \cdot \nabla_{p_1}}, \]

where the operators \( X \) and \( Y \) are given by Eqs. (A5) and (A6), respectively, and the vectors \( d(t) \) and \( p(t) \) are actually independent of \( r_1 \) and \( p_1 \).

\[ d(t) = \frac{1}{3} \frac{t^3}{m^2} (p_2 \cdot \nabla_{r_2}) K_{12}(r_2) + \cdots, \quad (A10) \]

and

\[ p(t) = -\frac{1}{2} \frac{t^2}{m} (p_2 \cdot \nabla_{r_2}) K_{12}(r_2) - \frac{1}{3} \frac{t^3}{m^2} (p_2 \cdot \nabla_{r_2})^2 K_{12}(r_2)

- \frac{1}{2} \frac{t^2}{m} (K_{21}(r_2) \cdot \nabla_{r_2}) K_{12}(r_2) + \cdots. \quad (A11) \]

The displacement \( d(t) \) and momentum \( p(t) \) given by Eqs. (A10) and (A11), respectively, follow more readily by integrating the series

\[ \dot{d}(t) \cdot \nabla_{r} + \dot{p}(t) \cdot \nabla_{p_1} + \frac{t}{m} \ddot{p}(t) \cdot \nabla_{r} = Y - e^{tX} Y e^{-tX} \]

\[ = -t[X, Y] \frac{t^2}{2!} [X, [X, Y]] - \frac{t^3}{3!} [X, [X, [X, Y]]] + \cdots. \quad (A12) \]

The differential equation in Eq. (A12) follows by differentiating Eq. (A9) with respect to time. Now the right-hand side of Eq. (A12) is given by the scalar product of a vector with the operator \( \nabla_{p_1} \) (see Eq. (A8)) and so

\[ \dot{d}(t) = -\frac{t}{m} \ddot{p}(t) \quad (A13) \]

and

\[ \ddot{p}(t) \cdot \nabla_{p_1} = -t[X, Y] \frac{t^2}{2!} [X, [X, Y]] - \frac{t^3}{3!} [X, [X, [X, Y]]] + \cdots. \quad (A14) \]

Therefore,

\[ \ddot{p}(t) = -\frac{t}{m} (p_2 \cdot \nabla_{r_2}) K_{12}(r_2) \]

\[ - \frac{1}{2} \frac{t^2}{m^2} (p_2 \cdot \nabla_{r_2})^2 K_{12}(r_2) \quad (A15) \]

\[ - \frac{1}{2} \frac{t^2}{m} (K_{21}(r_2) \cdot \nabla_{r_2}) K_{12}(r_2) + \cdots. \]

Results (A10) and (A11) follow on integrating Eq. (A13) and Eq. (A15).

Now

\[ \exp tY = \exp \left[ \frac{t}{m} (p_1 + \frac{t}{2} K_{12}(r_2)) \cdot \nabla_{r_1} \right] \times \exp \left[ t K_{12}(r_2) \cdot \nabla_{p_1} \right]. \quad (A16) \]
Therefore, Eq. (A9) becomes
\[ e^{-t(X+Y)} = e^{-tX} e^{D(t)Y} e^{P(t)X}, \]  
(A17)
where
\[ D(t) = -\frac{t}{m} p_1 + \frac{1}{2} \frac{t^2}{m} K_{12}(r_2) + d(t) \]  
(A18)
and
\[ P(t) = -t K_{12}(r_2) + p(t), \]
(A19)
which are independent of \( r \) and \( p_1 \) except for the explicit appearance of \( p_1 \) in the first term of the right-hand side of (A18).

**APPENDIX B: FRACTIONAL TAYLOR SERIES**

The generalization of the Taylor-Riemann series for fractional derivatives\(^{20,21}\) gives that
\[ f(z) = \sum_{n=-\infty}^{\infty} D_{\alpha}^n \frac{f(a)}{\Gamma(\alpha + n + 1)} (z - a)^{\alpha + n}, \]  
(B1)
where the Riemann-Liouville fractional derivative of order \( \alpha \) is given by
\[ D_{\alpha}^n f(x) = \frac{1}{\Gamma(1 - \alpha)} \frac{d}{dx} \int_a^x \frac{f(t)dt}{(x - t)^{1 - \alpha}}, \]  
(B2)
for \( 0 < \alpha < 1 \). The Riemann-Liouville fractional integral of order \( \alpha \) is given by
\[ I_{\alpha}^n f(x) = \frac{1}{\Gamma(\alpha)} \int_a^x \frac{f(t)dt}{(x - t)^{1 - \alpha}}, \]  
(B3)
for \( 0 < \alpha < \infty \).
If \( \alpha > 0 \) is not an integer, then
\[ D_{\alpha}^n f(x) = \frac{1}{\Gamma(n - \alpha)} \frac{d^n}{dx^n} \int_a^x \frac{f(t)dt}{(x - t)^{n - \alpha + 1}}, \]  
(B4)
where \( n = \lfloor \alpha \rfloor + 1 \) and \( \alpha = \lfloor \alpha \rfloor + \{ \alpha \} \), where \( \lfloor \alpha \rfloor \) denotes the integer part of \( \alpha \) and \( \{ \alpha \} \) denotes the remainder. If, however, \( \alpha < 0 \), then
\[ D_{\alpha}^n f(x) = I_{-\alpha}^n f(x). \]  
(B5)

There are other fractional Taylor series that avoid the problem of a non-zero fractional derivative of a constant, which occurs for the Riemann-Liouville fractional derivative (see Eq. (B2)). Consider, instead, the modified Riemann-Liouville fractional derivative\(^{22}\)
\[ f^{(\alpha)}(x) = \frac{1}{\Gamma(-\alpha)} \int_0^x (x - \xi)^{-\alpha - 1} (f(\xi) - f(0))d\xi, \quad (\alpha < 0), \]  
(B6)
\[ f^{(\alpha)}(x) = (f^{(\alpha - 1)}(x))' \quad (0 < \alpha < 1) \]
\[ = \frac{1}{\Gamma(1 - \alpha)} \frac{d}{dx} \int_0^x (x - \xi)^{-\alpha} (f(\xi) - f(0))d\xi, \]  
(B7)
and
\[ f^{(\alpha)}(x) = (f^{(\alpha - n)}(x))^{(n)} \quad (n \leq \alpha < n + 1, n \geq 1). \]  
(B8)
If \( f(0) = 0 \), then the fractional derivatives (B6)–(B7) are equal to the Riemann-Liouville fractional derivatives (B2)–(B5). The Taylor’s series of fractional order is\(^{22}\)
\[ f(x + h) = \sum_{k=0}^{\infty} \frac{h^k}{\Gamma(1 + ak)} f^{(\alpha k)}(x), \quad (0 < \alpha \leq 1). \]  
(B9)
If \( f(x) \) has derivatives of order \( k \) (integer), \( 1 \leq k \leq m \), then
\[
 f(x + h) = \sum_{k=0}^{m} \frac{h^k}{k!} f^{(k)}(x) + \sum_{k=1}^{\infty} \frac{h^{\alpha k+m}}{\Gamma(1+\alpha k+m)} f^{(\alpha k+m)}(x) \quad (0 < \alpha \leq 1).
\] (B10)

A generalization of Taylor’s formula, similar to (B9) but involving fractional derivatives in the Caputo-Djrbashian sense instead,\(^{23,24}\) has also been obtained.\(^{25}\) An advantage of the Jumarie definition of fractional derivative is that it applies to nondifferentiable function, which is not the case in the Caputo-Djrbashian sense. In addition, in order to obtain the first order derivative of a function, one must know beforehand the second order derivative.

The generalized Taylor-Riemann series (B1) is reminiscent of the Laurent expansion whereas the Jumarie series (B9) resembles somewhat the ordinary Taylor expansion. Note that (B1) becomes the ordinary Taylor series for \( \alpha = 0 \) and so does (B9) for \( \alpha = 1 \).

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