Kinetic equation for spatially averaged molecular dynamics*

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\textbf{ABSTRACT}

We obtain a kinetic description of spatially averaged dynamics of particle systems. Spatial averaging is one of the three types of averaging relevant within the Irving–Kirkwood procedure (IKP), a general method for deriving macroscopic equations from molecular models. The other two types, ensemble averaging and time averaging, have been extensively studied, while spatial averaging is relatively less understood. We show that spatially averaged density, linear momentum and kinetic energy can be obtained from a single generating function. A kinetic equation for the generating function is obtained. The deconvolution closure used in the derivation becomes more accurate with increasing density. This yields a kinetic description suitable for fluids and amorphous solids. In addition, we consider extensions of the theory which include time averaging and coarsening in the velocity space. Since averaging is essentially a low-pass filter which damps high frequency oscillations, adding time- and velocity-variable averaging allows us to filter out all oscillations below a specified threshold. Unlike traditional kinetic theory, the proposed equation applies to single realizations of atomistic dynamics.

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1. Introduction

In 1950, Irving and Kirkwood [1] proposed an averaging method for deriving macroscopic theories from molecular description. Three types of averages are relevant within the Irving–Kirkwood procedure (IKP): ensemble averages [1, 2], time averages [3] and space averages [4–6]. All three types can be used either separately or in combination. Ensemble averaging and time averaging are well understood, while spatial averaging is relatively less explored. Studying space averaging is useful because (i) space–time averages represent the most realistic model of macroscopic measurements in a single experiment; (ii) the number of repetitions in engineering experiments can be too small for accurate sampling of the underlying probability distribution and (iii) spatial averages are easy to compute in molecular dynamics (MD) simulations whereas ensemble averaging requires costly integrations in phase space, and long-time averages may be inaccessible because of time-scale limitations of MD algorithms.

Thus it makes sense to ask the following question. What information can be obtained from spatial averages of a single realization MD? Assume that the particle dynamics is governed by Newton’s ordinary differential equations (ODEs), and the size of the system is so large that direct MD simulation is infeasible. Even though we do not have access to particle trajectories, it may be still possible to formulate a self-contained set of equations for mesoscopic functionals of interest (e.g. average
density, velocity and kinetic energy). Exact evolution equations for these quantities were derived in [3, 4], but these equations are not closed, and thus cannot be used without knowing positions and velocities of all particles. A possible solution is to find closed-form approximations (also called constitutive equations) for stress and heat flux. Another possibility is to develop a kinetic description that can be used instead of continuum balance equations. The purpose of this article is to develop such a description. We are particularly interested in dense fluids and soft matter because of the difficulties [7–10] with extending classical kinetic theory [11] to these media. The closure problem still remains, and we address it by adapting deconvolution techniques for momentum balance equations from [5, 6, 12]. The role of closure is to enable complexity reduction: a mesoscopic model should have fewer independent variables and we should be able to use this model without solving particle ODEs. A related work on problem reduction, renormalization and memory is [13].

Regardless of the chosen scales, the averages in IKP satisfy exact mesoscopic equations derived from the underlying ODEs [3, 4]. In addition, the averages can be made slowly varying by increasing the associated scales. The exact dynamics of these slowly varying averages can be viewed as the best available model of reality. It is, therefore, natural to require that a closure approximation reproduce this reality as closely as possible. It is of interest to mention that the Irving–Kirkwood averaging procedure also extends to the passage from quantum dynamics to macroscopic conservation laws [14].

Other objectives, such as obtaining a model that increases entropy, are secondary. Any model that reproduces the exact mesoscopic dynamics cannot be dissipative unless the exact dynamics itself is dissipative, at least to a good approximation. In this work, the issue of dissipativity is not addressed because fluctuation theorems [15, 16] suggest that the second law does not always hold in finite size systems considered on finite time intervals. This means that spatial averaging alone may be insufficient to make the exact dynamics dissipative. One option is to enforce dissipativity by working only with models that combine spatial averaging with statistical and (or) time averaging. Another option, which we adopt in this work, is to find an accurate but not necessarily dissipative closure for spatially averaged dynamics.

The first option represents the conventional approach employed in projection operator methods [17–20]. By projecting the exact dynamics onto the subspace spanned by slow dynamical functions, one obtains generalized Langevin equations, and then derives from them dissipative continuum models. But the method is not without drawbacks. Generalized Langevin equations are not closed because it is still necessary to run MD simulations to evaluate material parameters such as viscosity. Projection operator models are difficult to validate by direct simulation because computed ensemble and time averages may carry large error. Theoretical error estimates for these methods are currently unavailable. This makes the second option worthy of investigation. There are two possible difficulties here: (i) oscillations with respect to time may be poorly controlled and (ii) lack of dissipation may lead to numerical instability. One can manage the first difficulty by adding time averaging, and the second problem can be dealt with by using limiters and stability-preserving numerical schemes. Time averaging is considered in Section 6, and (ii) is left to future work.

The paper is organized as follows. Section 2 is devoted to definitions of various spatial averages. In this section, we introduce the generating function that plays a role similar to that of the one-particle distribution function of conventional kinetic theories. Closure is described in Section 3, where we also derive the kinetic equation for the generating function. Section 4 describes the results of some numerical experiments demonstrating accuracy of closure. Section 5 contains the discussion of obtained results and a brief description of possible extensions of the theory. The generating function varies slowly with respect to the spatial variable, but may be oscillatory with respect to time and velocity variables. Therefore, in Section 6 we obtain a kinetic description that incorporates velocity-variable coarsening and time averaging. Conclusion is given in Section 7.
2. Spatial averages and generating function

Consider a system of $N$ classical point particles of equal mass $m = M/N$ confined to the domain $\Omega$ and interacting with short-range forces generated by a pair potential $U$. Microscopic state variables are positions $q_j(t)$ and momenta $p_j(t)$. Mesoscopic behaviour of the system can be characterized using spatially averaged density $\bar{\rho}$, linear momentum $\bar{p}$ and kinetic energy $K$ defined as follows:

$$\bar{\rho}(t, x) = \sum_{j=1}^{N} m \psi(\eta)(x - q_j(t)), $$
$$\bar{p}(t, x) = \sum_{j=1}^{N} p_j(t) \psi(\eta)(x - q_j(t)), $$
$$K(t, x) = \frac{1}{2m} \sum_{j=1}^{N} p_j(t) \cdot p_j(t) \psi(\eta)(x - q_j(t)).$$  (1)

The mesoscopic length scale $\eta$ is much larger than the characteristic interparticle distance, but may be much smaller than the extent of the whole system. For each $\eta$, the averages in (1) satisfy exact continuum-style balance equations of mass, momentum and energy [3, 4].

The window function $\psi$ is normalized by requiring $\int \psi(x) \, dx = 1$, and then scaled by $\eta$ so that $\psi(\eta)(x - q_j) = \eta^{-d} \psi(x/\eta)$, where $d$ is the physical space dimension. Scaling ensures that $\psi(\eta)(x - q_j)$ converges to $\delta(x - q_j)$ as $\eta \to 0$. In this limit, one recovers the phase-space densities similar to the densities used in IKP [1] and statistical hydrodynamics [7]. Increasing $\eta$ increases the number of particles within the averaging volume (the support of $\psi(\eta)$). This has the effect of damping high frequency oscillations. For example, the Fourier transform $F_{x \to k} \bar{\rho}$ is equal to $\hat{\psi}(\eta k) \sum_{j=1}^{N} m e^{ikq_j}$, where $\hat{\psi}$ is the Fourier transform of $\psi$ and $\sum_{j=1}^{N} m e^{ikq_j}$ is the Fourier transform of $\sum_{j=1}^{N} m \delta(x - q_j)$. For larger $\eta$, the filter function $\hat{\psi}(\eta k)$ is more localized near $k = 0$ [21].

To derive a kinetic equation, we introduce another spatial average, called the generating function

$$Q(t, x, z) = \sum_{j=1}^{N} m e^{im^{-1}p \cdot z} \psi(\eta)(x - q_j).$$  (2)

As noted above, $Q$ is expected to be slowly varying for sufficiently large $\eta$ because its Fourier transform $F_{x \to k}$ will be more localized near $k = 0$. The continuum quantities $\bar{\rho}, \bar{p}$ and $K$ can be obtained from $Q$ as follows: $\bar{\rho} = Q(t, x, 0), \bar{p} = -i \nabla_z Q(t, x, 0), K = -\frac{1}{2} \nabla_z \cdot \nabla_z Q(t, x, 0)$. To relate this to conventional moment expressions, note that the Fourier transform $F_{x \to \xi}$ of $Q$ is the spatially coarsened phase space density

$$f(x, \xi) = \sum_{j=1}^{N} m \delta(\xi - m^{-1}p_j) \psi(\eta)(x - q_j)$$  (3)

that is similar to the density used by Mori [22]. Therefore, $z$-differentiation of $Q$ corresponds to multiplication of $f$ by $i\xi$, and evaluation at $z = 0$ corresponds to integration of moments of $f$ with respect to $\xi$. Using $Q$ instead of $f$ may be more convenient because it permits replacing integrations over the whole $\xi$-space with evaluation of derivatives at $z = 0$. To do this, it is enough to know $Q$ only for $z$ close to zero.
Taking time derivative in Equation (2) and using Newton’s equations \( \dot{p}_j = f_j \) yields the exact evolution equation

\[
\partial_t Q - i \nabla_x \cdot \nabla_z Q = iz \cdot F,
\]

where

\[
F(t; x, z) = \sum_{j=1}^{N} f_j e^{im\cdot q_j(t)} \psi_\eta(x - q_j(t))
\]

and

\[
f_j = - \sum_{k \neq j} U'(q_j - q_k) \frac{q_j - q_k}{|q_j - q_k|}
\]

is the total force acting on a particle \( j \). Equation (4) is exact but not closed, since one must know all \( q_j(t) \) and \( p_j(t) \) to evaluate \( F \).

3. Closure

Next we derive a closed-form coarse-grained equation for \( Q \). The main difficulty here is to find a closure approximation of the exact \( F \) in (5) by an operator acting on \( Q \). A self-contained kinetic equation is obtained by inserting this approximation into Equation (4). The first step is to approximate \( Q \) and \( F \) by integrals. In doing so, we deviate from the standard phase space description of dynamics and think instead of a physical domain containing moving particles. In physical space–time, the dynamics can be described by the micro-scale continuum deformation \( \tilde{q}(t, X) \) and velocity \( \tilde{v}(t, \tilde{q}) \) [5, 23]. Since these fields interpolate particle positions and velocities, one can use them to approximate sums by integrals:

\[
Q \approx \frac{M}{V_\Omega} \int_{\Omega} e^{i\tilde{v}(t, \tilde{q}(t,X)) \cdot z} \psi_\eta(x - \tilde{q}(t, X)) \, dX
\]

\[
= \frac{M}{V_\Omega} \int_{\Omega} e^{i\tilde{v}(t,y) \cdot z} \psi_\eta(x - y)J(t, y) \, dy,
\]

where \( V_\Omega \) is the volume of \( \Omega \) and \( J = |\text{det} \nabla \tilde{q}^{-1}| \).

Similarly,

\[
F \approx \frac{N}{V_\Omega} \int_{\Omega} e^{iz \tilde{v}(t, \tilde{q}(t,X))} \psi_\eta(x - \tilde{q}(t, X)) \sum_{k=1}^{N} -U' \left( \tilde{q}(t, X) - q_k(t) \right) \frac{\tilde{q}(t, X) - q_k(t)}{|\tilde{q}(t, X) - q_k(t)|} \, dX
\]

\[
= \frac{N}{V_\Omega} \int_{\Omega} e^{iz \tilde{v}(t,y)} \psi_\eta(x - y) \sum_{k=1}^{N} -U'(y - q_k(t)) \frac{y - q_k(t)}{|y - q_k(t)|} J(t, y) \, dy.
\]

The fine scale quantities in (8) are \( \tilde{v}, J, \) and positions \( q_k \). They have to be approximated in terms of the averaged quantity \( Q \). Since \( \psi_\eta \) is close to the delta function for small \( \eta \), (7) yields

\[
e^{i\tilde{v}(t,y) \cdot z} J(t, y) \approx \frac{V_\Omega}{M} Q(t, y, z).
\]

To approximate \( q_k \), we use the average density \( \bar{Q}(t, x) = Q(t, x, 0) \) and replace \( q_k(t) \) with the sampled average deformation \( \chi_k(t) \). To generate \( \chi_k \), split the physical domain into mesoscopic cells and then place fictitious particles inside each cell so that the average density of the fictitious array is equal to the known average density \( Q(t, x, 0) \) at the center of the cell. The details of the placement may vary. One may use locally periodic or random placements. The point is that only the knowledge of the
average(s) is required for the generation of the fictitious particle array. In that sense, the resulting equation is closed and can be evaluated by evolving averages without the need to resolve the actual particle trajectories $q_k$. A similar but more sophisticated procedure is described in [24]. In that work, we first find the average deformation $\chi(t,X)$ by solving the kinematic ODE $\dot{\chi} = m^{-1}\overline{p}(t,\chi)$ and then use locally periodic placement consistent with the average deformation and average concentration. This procedure generalizes a well-known Cauchy–Born rule.

Combining equations, we obtain the closed-form approximation

$$F(t,x,z) \approx F^{(cl)}(t,x,z) = \frac{N}{M} \int Q(t,y,z) \psi_h(x-y) \sum_{k=1}^{N} -U'(y - \chi_k(t)) \frac{y - \chi_k(t)}{|y - \chi_k(t)|} \, dy \quad (10)$$

and the corresponding kinetic equation

$$\partial_t Q - i \text{div}_x \nabla_z Q = i z \cdot F^{(cl)}.$$  

(11)

4. Numerical experiments

The function $Q_e$ given by Equation (2) and the approximate $Q_a$ that solves Equation (11) are both coarse scale quantities. If there is no need for additional coarsening, the purpose of closure should be accurate and efficient reproduction of the exact dynamics. Comparison of the two equations shows that a good closure should satisfy $F \approx F^{(cl)}$. To check this for the approximate $F^{(cl)}$ given by Equation (10), we ran an MD simulation of an 1-D system with $N = 10,000$ particles interacting with the Lennard–Jones potential $U$. The potential was truncated at the distance $20/\Omega_1$. We took $V_\Omega = 1$ and $M = 1$. Periodic boundary conditions were imposed, and the equations of motion were solved using the Verlet algorithm until $t = 0.01$ with the step size $2 \cdot 10^{-4}$. We considered two sets of initial conditions. In the first set, the initial positions were uniformly spaced, and the initial velocities were prescribed using a centred, piecewise polynomial, approximate Gaussian pulse for the middle third of the particles. In the second set, the initial velocities were set to zero, and the initial positions vary sinusoidally.

The averages were generated using the window function $\psi(x)$ equal to $\frac{15}{16}(1 + x)^2(1 - x)^2$ when $|x| < 1$, and equal to zero otherwise. We computed $Q$ and $F$ using $q_j, v_j$ from the MD simulation and then used the obtained $Q$ to evaluate $F^{(cl)}$ from (10). The integral quadrature in (10) was implemented by first generating a uniform grid with step size $h$, and then scaling the grid points by the density, so that

$$\sum_{k=1}^{N} -U'(y_i - \hat{\chi}_k(t)) \approx \sum_{k=1}^{N} -U'\left(\frac{h(i-k)}{Q(t,y_i,0)}\right).$$

The approximation of the flux $F$ by the closed-form $F^{(cl)}$ is quite accurate over the range of parameters we used. Figure 1 shows the real part of $F$ computed from Equation (5), compared with the real part of $F^{(cl)}$ computed from Equation (10). Increasing $\eta$ has only a slight effect on the quality of the approximation, as shown in the right panel of Figure 1. Approximately the same accuracy was obtained for all $z$ with $|x| \leq 0.2$. An example with $z = 0$ is shown in Figure 2. In this range of $z$, and for $\eta$ values between 0.01 and 0.03 (that is between 1% and 3% of the size of the computational domain) the largest relative error was 3%.

5. Discussion and possible extensions

Simulation results demonstrate good computational fidelity of the proposed closure during the simulation time. Therefore, the exact dynamics of $Q$ (Equation (4)) can be, in principle, well reproduced by the coarse-scale model given by Equation (11). The accuracy can be further improved by using a more sophisticated deconvolution closure [5, 21, 23]. This amounts to replacing $F^{(cl)}$ in Equation (9) with a more accurate approximate solution of the integral equation (7). One such solution is given by
Figure 1. Computed flux and closure approximation for $N = 10^4$, $t = 0.002$, $z = 0.2$, $\eta = 0.01$ (left panel) and $\eta = 0.03$ (right panel).

Figure 2. Computed flux and closure approximation for $N = 10^4$, $t = 0.002$, $\eta = 0.03$ and $z = 0$.

the classical Landweber (or Van Cittert) iteration [25]:

$$e^{i\psi z}J \approx \frac{\nu_{12}}{M} \sum_{s=0}^{n} (I - R)^s Q,$$

(12)

where $I$ is the identity operator and $R$ is the convolution operator with the kernel $\psi_{\eta}$. Taking a larger $n$ may improve the approximation. Equation (9) corresponds to $n = 0$. Error estimates for this and similar closures can be found in [21].

Recall that the Fourier transform of $Q$ with respect to $z$ is a spatially averaged one-particle phase space density $f$. The kinetic equation

$$\partial_t f + \xi \cdot \nabla_x f = \int \psi_{\eta}(x - y) \sum_{k=1}^{N} -U'(y - \chi_k(t)) \frac{y - \chi_k(t)}{|y - \chi_k(t)|} f(t, y, \xi) \, dy$$

(13)

is obtained from (11) by taking the Fourier transform with respect to $z$. The closure operator in the right-hand side is non-linear since $\chi_k$ depend on $\bar{\rho}(t, y) = \int f(t, y, \xi) \, d\xi$.

An accurate approximation of $Q$ leads to an accurate approximation of the dynamics of $f$. However, this dynamics is not necessarily dissipative. The fluctuation theorem of Evans et al. [15] implies that the second law of thermodynamics may fail for some initial conditions. Probability of failure increases with decreasing system size. A related theorem of Galavotti and Cohen [16] guarantees dissipativity of single realization averaged over infinite time intervals under additional condition of strong chaoticity. These results allow for the possibility that a single MD trajectory has decreasing entropy production on a finite time interval. Therefore, if kinetic theory involves only spatial averaging, then it cannot be expected to be dissipative in general.

To obtain dissipation, one may have to incorporate time- and (or) ensemble averaging. In this regard, note that all spatial averages depend on the initial positions $q_{0j}$ and momenta $p_{0j}$, since $q_j(t) = q_j(t, q_{01}, q_{02}, \ldots, q_{0N}, p_{01}, \ldots, p_{0N})$, and similarly for momenta. Choosing a probability density
\( p(t, q_1^0, q_2^0, \ldots, q_N^0, p_1^0, \ldots, p_N^0) \), one can define ensemble average of \( Q \) and then use projection operator method to separate the dissipative component of the ensemble-averaged dynamics, following, for example [22]. We do not attempt such an extension here. The goal of this work is to produce a coarse-scale theory that neither requires nor implies dissipativity. Unlike conventional kinetic equations, this theory can be useful even when the exact dynamics in Equation (4) does not admit a good dissipative approximation.

Finally, it seems possible to extend the theory to crystalline solids using the approach from [26]. Such an extension requires replacing pairwise forces with more general multi-body forces.

6. Time averaging and velocity coarsening

6.1. Coarsening in the velocity space

The function \( f \) in Equation (3) is slowly varying in \( x \) but may be still oscillatory with respect to \( \xi \) because of the presence of delta functions. If required, one can smooth out the velocity dependence by replacing \( f \) with

\[
\begin{align*}
    f_\lambda &= \sum_j m \chi_\lambda(\xi - v_j) \psi_\eta(x - q_j),
\end{align*}
\]

where \( \chi \) is another window function depending on a mesoscopic parameter \( \lambda \). In the following, it is convenient to use the Gaussian

\[
    \chi_\lambda(\xi) = \frac{1}{\lambda^d \pi^{d/2}} \frac{e^{-|\xi|^2/\lambda^2}}{\lambda^d},
\]

where \( d \) is the physical space dimension. Differentiating Equation (14) in time and using Newton’s equations to eliminate \( \dot{v}_j \) we find

\[
    \partial_t f_\lambda = \nabla_\xi \cdot G + \nabla_x \cdot \left( -\sum_{j=1}^N m v_j \chi_\lambda(\xi - v_j) \psi_\eta(x - q_j) \right),
\]

where

\[
    G(t, x, \xi) = -\sum_{j=1}^N f_j \chi_\lambda(\xi - v_j) \psi_\eta(x - q_j).
\]

Using

\[
    \nabla_\xi \chi_\lambda(\xi - v_j) = \frac{2(\xi - v_j)}{\lambda^2} \chi_\lambda(\xi - v_j),
\]

the second term in the right-hand side of Equation (16) can be written as follows:

\[
    \nabla_x \cdot \left( -\sum_{j=1}^N m v_j \chi_\lambda(\xi - v_j) \psi_\eta(x - q_j) \right) = \frac{\lambda^2}{2} \nabla_x \cdot \nabla_\xi f_\lambda - \xi \cdot \nabla_x f_\lambda.
\]

Combining this with (16) yields

\[
    \partial_t f_\lambda + \xi \cdot \nabla_x f_\lambda = \nabla_\xi \cdot G + \frac{\lambda^2}{2} \nabla_x \cdot \nabla_\xi f_\lambda.
\]
To close the above equation, \( G \) should be approximated by an operator acting on \( f_\lambda \). Repeating the arguments leading to (10) we obtain the kinetic equation

\[ \partial_t f_\lambda + \xi \cdot \nabla_x f_\lambda = \nabla_\xi \cdot \left( \frac{N}{M} \int f_\lambda(t, y, \xi) \psi_\eta(x - y) \sum_{k=1}^{N} \frac{1}{\lambda} - U'(y - x(t)) \frac{y - x_k(t)}{|y - x_k(t)|} \, dy + \frac{\lambda^2}{2} \nabla_x f_\lambda \right). \] (20)

Generating fictitious positions \( x_k \) can be computation-intensive. To bypass this difficulty, a different closure construction can be obtained. First, the antisymmetric property of the forces \( f_{jk} = -f_{kj} \) implies that

\[ G = \frac{1}{2} \sum_{j=1}^{N} \sum_{k=1}^{N} U'(q_j - q_k) \frac{q_j - q_k}{|q_j - q_k|} \left( \chi_\lambda(\xi - v_j) \psi_\eta(x - q_j) - \chi_\lambda(\xi - v_k) \psi_\eta(x - q_k) \right). \]

This sum can be approximated by the integral

\[ G \approx \frac{1}{2} \int U'(y' - y'') \frac{y' - y''}{|y' - y''|} \left( \psi_\eta(x - y') g_1(t, y', y'', \xi) - \psi_\eta(x - y'') \right) \times g_2(t, y', y'', \xi) \, dy' \, dy'', \] (21)

where

\[ g_1(t, y', y'', \xi) = \sum_{j=1}^{N} \sum_{k=1}^{N} \chi_\lambda(\xi - v_j) \psi_\eta(y' - q_j) \psi_\eta(y'' - q_k), \] (22)

and \( \tilde{\eta} \leq \eta \), so that \( \psi_{\tilde{\eta}} \) can be considered as an approximation of the delta function. Multiplying the first of these equations by \( 1 = \int d\xi' \chi_\lambda(\xi' - v_j) \), and the second by \( 1 = \int d\xi' \chi_\lambda(\xi' - v_k) \), and replacing \( \psi_{\tilde{\eta}} \) with \( \psi_\eta \) yields the approximations

\[ g_1(t, y', y'', \xi) \approx \int \left( \sum_{j=1}^{N} \chi_\lambda(\xi - v_j) \psi_\eta(y' - q_j) \right) \times \left( \sum_{k=1}^{N} \chi_\lambda(\xi - v_k) \psi_\eta(y'' - q_k) \right) \, d\xi', \] (23)

\[ g_2(t, y', y'', \xi) \approx \int \left( \sum_{j=1}^{N} \chi_\lambda(\xi' - v_j) \psi_\eta(y' - q_j) \right) \times \left( \sum_{k=1}^{N} \chi_\lambda(\xi' - v_k) \psi_\eta(y'' - q_k) \right) \, d\xi'. \] (24)
Combining Equations (21)–(23), we find the closure approximation
\[ \mathbf{G} \approx \mathbf{G}^{(c)} \]
\[ \approx \frac{N^2}{2M^2} \int \mathbf{U}(\mathbf{y}' - \mathbf{y}'') \left( \psi_{\eta}(\mathbf{x} - \mathbf{y}') f_{\lambda}(t, \mathbf{y}', \mathbf{u}, \xi) f_{\lambda}(t, \mathbf{y}'', \mathbf{u'}) \right) \]
\[ - \psi_{\eta}(\mathbf{x} - \mathbf{y}'') f_{\lambda}(t, \mathbf{y}'', \mathbf{u}) f_{\lambda}(t, \mathbf{y}', \mathbf{u'}) \, d\xi' \, d\mathbf{y}' \, d\mathbf{y}'', \]  
(25)

where \( \mathbf{U}(\mathbf{y}' - \mathbf{y}'') = \mathbf{U}'(\mathbf{y}' - \mathbf{y}'') \frac{\mathbf{y}' - \mathbf{y}''}{|\mathbf{y}' - \mathbf{y}''|} \). Similarly to Equation (10), the closures in Equations (20) and (25) can be improved by using deconvolution.

### 6.2. Time averaging

A physically meaningful time average should be backward. In practice, averaging would be carried out on a finite interval of length \( \tau \):
\[ \bar{f}(t, \mathbf{x}, \mathbf{u}) = \frac{1}{\tau} \int_{t-\tau}^{t} f(s, \mathbf{x}, \mathbf{u}) \, ds. \]  
(26)

More general backward time averages can be defined similarly [3].

It is easy to check that time averaging commutes with differentiation with respect to \( t, \mathbf{x} \) and \( \mathbf{u} \). Using this to average the exact kinetic Equation (19) we find
\[ (\partial_t + \mathbf{u} \cdot \nabla_{\mathbf{x}}) \bar{f}_{\lambda} = \nabla_{\mathbf{x}} \cdot \overline{\mathbf{G}} + \frac{\lambda^2}{2} \nabla_{\mathbf{x}} \cdot \nabla_{\mathbf{u}} \bar{f}_{\lambda}. \]  
(27)

To close this equation, \( \mathbf{G} \) should be approximated by an operator acting on available average quantities. Equation (20) shows that the required quantity is \( f_{\lambda}(t) \) (dependence on other variables is suppressed), while the available quantity is its time average \( \bar{f}_{\lambda}(t) \). So, to apply the already developed closure, we need to approximate the local in time value \( f_{\lambda}(t) \) by an operator acting on its time average. Since the time averaging operator \( T \) defined by
\[ T_{\tau} f(t) = \frac{1}{\tau} \int_{t-\tau}^{t} f(s) \, ds \]
is a convolution, we can use Landweber iteration to generate approximations
\[ f_{\lambda} \approx f_{\lambda}^{(n)} = \sum_{k=0}^{n} (I - T_{\tau})^k \bar{f}_{\lambda} \]  
(28)

of increasing complexity. The simplest approximation corresponding to \( n = 0 \) is simply \( f_{\lambda}(t) \approx \bar{f}_{\lambda}(t) \).

The next approximation \( f_{\lambda}^{(1)} = 2\bar{f}_{\lambda} - T_{\tau} \bar{f}_{\lambda} \), and so on. Combining Equations (20) and (28) yields the closure approximation
\[ \mathbf{G} \approx \mathbf{G}^{(n)} = \frac{N}{\tau M} \int_{t-\tau}^{t} \int f_{\lambda}^{(n)}(t, \mathbf{y}, \mathbf{u}) \psi_{\eta}(\mathbf{x} - \mathbf{y}) \sum_{k=1}^{N} -U' \left( \mathbf{y} - \chi_{k}(t) \right) \frac{\mathbf{y} - \chi_{k}(t)}{|\mathbf{y} - q_k(t)|} \, d\mathbf{y}, \]  
(29)

where \( n = 0, 1, 2 \ldots \), and \( \hat{q}_k \) are generated using \( \int f_{\lambda}^{(n)}(t, \mathbf{y}, \mathbf{u}) \, d\xi \). The closed-form kinetic equation is obtained from Equation (27) by replacing \( \mathbf{G} \) with \( \mathbf{G}^{(n)} \). A similar procedure can be applied to Equation (25) to produce its time-averaged version.
7. Conclusion

The kinetic description developed here meets the following criteria: (i) it is completely closed and thus can be used independently of solving MD equations; (ii) it accurately reproduces the exact dynamics of the functions \( Q, f_\lambda \) and \( \tilde{f}_\lambda \); (iii) it applies to single realizations, and thus impractical ensemble averaging can be avoided, if needed; (iv) it works for dense materials such as fluids and amorphous solids.

The crucial property (iv) is due to the nature of closure. The closure is constructed without assuming that dynamics is dominated by collisions. Instead, we use spatial interpolation, integral approximations and deconvolution closure. This permits long-range correlations and produces approximations that become more accurate with increasing particle density.

Disclosure statement

No potential conflict of interest was reported by the authors.

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