THE OVERDAMPED LIMIT OF DYNAMIC DENSITY FUNCTIONAL THEORY: RIGOROUS RESULTS[*]

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Abstract. Consider the overdamped limit for a system of interacting particles in the presence of hydrodynamic interactions. For two-body hydrodynamic interactions and one- and two-body potentials, a Smoluchowski-type evolution equation is rigorously derived for the one-particle distribution function. This new equation includes a novel definition of the diffusion tensor. A comparison with existing formulations of dynamic density functional theory is also made.

1. Introduction.

1.1. Review of existing dynamic density functional theory. Several problems in condensed matter physics such as colloidal suspensions and polymers can often be described as systems of interacting Brownian particles, either in the presence or absence of hydrodynamic interactions [20, 49]. Hydrodynamic interactions are due to forces on the colloid particles caused by flows in the suspending fluid (referred to as the bath), which are generated by the motion of the colloidal particles, and can be thought of as generalized friction forces. Such systems of interacting Brownian particles can be described either in phase space, when both positions and momenta of the particles are taken into account, or in configuration space, when inertial effects are neglected and only the position of the Brownian particles is taken into account. The evolution of the phase space distribution function is described by the Kramers equation [34, 35]. On the other hand, the evolution of the distribution function in configuration space is governed by the Smoluchowski equation [73, 24].

The Smoluchowski equation can be derived from the Kramers equation in the overdamped, i.e., high friction, limit. In this limit the velocity of the particles thermalizes quickly, i.e., the velocity distribution converges quickly to a Maxwellian, and the momentum variables can be eliminated through an appropriate adiabatic elimination procedure. This procedure is now well understood, both for a single Brownian particle as well as for systems of interacting particles, and both in finite as well as infinite dimensions [12, 13, 47, 59, 30].

Whilst the derivation of the Smoluchowski equation was already discussed by both Klein and Kramers, their approach was largely heuristic and a rigorous theory was not introduced until later [47]. Systematic adiabatic elimination techniques were introduced in the 1970s and applied to the problem of the rigorous derivation of

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the Smoluchowski equation for the Kramers equation, e.g., by Wilemski [75] and Titulaer [70, 71]. In particular, Titulaer considered the fully interacting N-body linear Kramers equation and used a multiple-time-scale or Chapman–Enskog expansion in the friction constant to systematically derive the N-body Smoluchowski equation and its corrections. These systematic adiabatic elimination procedures can be understood in the context of singular perturbation theory for Markov processes [50] or, more generally, in the framework of multiscale methods (see, e.g., [52]). A pedagogical discussion of the multiple-time-scale technique was given by Bocquet [8] for the case of noninteracting particles and for the one-body reduced distribution function. See also [52, Chap. 11] and [51, Chap. 8]. It is worth noting that in all these derivations, the equations for the one-body distribution function are linear.

Both the Kramers and Smoluchowski equations describe the full N-body dynamics, and, although they are linear, they are not well suited to computation. This is due to the large number of variables, and whilst the derivation of the Smoluchowski equation from the Kramers equation is of fundamental interest, the reduction from $6N$ to $3N$ variables by eliminating the momentum variables is insignificant in terms of computational complexity. However, a further simplification arises by integrating out over the positions (and momenta) of all but one particle, which then allows one to obtain the dynamics of the reduced distribution functions [5, 55]; see also (2.2). One of the main goals of statistical mechanics and kinetic theory is the derivation of closed equations for these reduced distribution functions, and in particular for the one-body distribution function. If there are no interparticle interactions, i.e., the only force comes from an external potential, and hydrodynamic interactions between particles are neglected, this reduction procedure results in one-particle versions of the Kramers and Smoluchowski equations. In the more general case where such interparticle effects may not be neglected, the equations must be closed by choosing a suitable approximation of the higher-body densities in terms of the one-body distribution function, e.g., a mean field approximation. Such a description is the ultimate aim of dynamic density functional theory (DDFT).

Consider a system of N interacting particles with N-body distribution function $f^{(N)}(\mathbf{r}_1, \mathbf{p}_1, \ldots, \mathbf{r}_N, \mathbf{p}_N, \tau)$, which gives the probability of finding particles at $\mathbf{r}_1, \ldots, \mathbf{r}_N$ with momenta $\mathbf{p}_1, \ldots, \mathbf{p}_N$ at time τ. The derivation of a self-consistent DDFT requires expressing the full N-body distribution function $f^{(N)}$ in terms of the one-body reduced distribution $f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, \tau)$ (see (2.2)) or, if starting from the Smoluchowski equation, in terms of $\rho(\mathbf{r}_1, \tau) := \int d\mathbf{p}_1 f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, \tau)$. Whilst it is known that $f^{(N)}$ (and thus all properties of the system including all lower n-body distributions) is given by a unique functional of ρ, both in [33, 45] and out [14] of equilibrium, in the general case this functional is unknown. However, much work has been done for the equilibrium case (density functional theory, or DFT, which allows for an accurate description of the microscopic properties of a fluid in terms of its density distribution; see [28, 60] for early work and, e.g., [76, 77] for recent overviews), and there exist accurate functionals, e.g., Rosenfeld's fundamental measure theory for hard spheres [61, 62, 63] and the mean field approximation [38, 1], mentioned earlier, which becomes exact for soft interactions at high densities. DFT represents one of the most widely used methods in condensed matter physics for the study of the microscopic structure of nonhomogeneous fluids within the framework of equilibrium statistical mechanics. It offers an increasingly popular compromise between computationally costly molecular dynamics simulations and various phenomenological approaches. It has been used to describe a wide variety of physical settings, ranging from polymers [37], liquid crystals [16], and molecular self-assembly [69, 29] to interfacial phenomena including

wetting transitions on substrates [67, 7, 48].

DDFT is also a popular approach in condensed matter physics and has been applied to a wide range of problems including spherical colloids without hydrodynamic interactions in both configuration [21, 40, 41, 26] and phase space [39, 3], dense atomic liquids [2], anisotropic colloids [58], and inhomogeneous granular fluids [43]. The effects of inertia [42, 44] and hydrodynamic interactions [56, 54] have also been studied. However, none of the formalisms derived so far is rigorous. The relationships between different approaches are summarized in Figure 1.1. We note, in particular, that the two routes for obtaining the one-body Smoluchowski equation give, in general, different formulations. These derivations can be divided into four cases, first by whether they start from the Kramers or Smoluchowski equation, and second by whether or not they include hydrodynamic interactions.

1.2. Starting from the Kramers or the Smoluchowski equation. As mentioned earlier, the Smoluchowski equation is expected to be valid in the overdamped limit, whereas for intermediate and small values of the friction coefficient the Kramers equation should be used. Perhaps the most common additional approximation is to ignore the effects of the hydrodynamic interactions between the particles (for exceptions, see [56, 54]). Whilst this may be acceptable in a very dilute system, such interactions decay only polynomially slowly with interparticle distance and are thus long-range and important in many applications [20].

When starting from the Smoluchowski equation and neglecting hydrodynamic interactions, it suffices to employ the adiabatic approximation, first introduced by Marconi and Tarazona [40, 41]. At equilibrium, Mermin's proof [45] shows that there exists a unique functional of ρ, $\mathcal{F}_{\mathrm{ex}}[\rho]$, called the excess free energy functional, which exactly determines the contributions from the many-body potentials (which a priori involve higher-order reduced distributions). It then remains to determine accurate, generally empirical approximations to the unknown functional $\mathcal{F}_{\mathrm{ex}}[\rho]$. The adiabatic approximation assumes that the same relationship holds away from equilibrium. This is equivalent to assuming that the nonequilibrium n-body distributions are identical to those in an equilibrium system with the same instantaneous density ρ. This approximation has proven accurate in a range of systems [1, 4, 57, 64].

If hydrodynamic interactions are included, this approximation is insufficient. This is because there are no hydrodynamic effects at equilibrium. Instead, at least for two-body interactions, one uses the identity $\rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) = \rho(\mathbf{r}, t)\rho(\mathbf{r}', \tau)g(\mathbf{r}, \mathbf{r}'; [\rho])$, where g is a pair-distribution function, whilst the function $g - 1$ is known as the pair correlation function (it provides a measure of the distance over which particles are correlated; for an ideal gas $g = 1$) and assumes that a good approximation to g is known [56] (often g can be approximated with different methodologies, such as the BBGKY hierarchy or the Ornstein–Zernike equation). Note in particular that g is a functional of ρ.

When starting from the Kramers equation an additional problem is encountered. In this case one obtains an infinite hierarchy of equations for the evolution of the momentum moments of $f^{(1)}(\mathbf{r}, \mathbf{p}, \tau)$, i.e., for $\int \mathrm{d}\mathbf{p}\, p_1^{a_1} p_2^{a_2} p_3^{a_3} f^{(1)}(\mathbf{r}, \mathbf{p}, \tau)$ with $a_j \geq 0$, $\sum a_j = n$. To obtain closure, one must truncate this hierarchy at a given level, which requires the approximation of higher moments. For example, for the standard truncation at the velocity ($n = 1$) level (i.e., the same level of description as the Navier–Stokes equations), one must control terms (in appropriate units) of the form $\int \mathrm{d}\mathbf{p}\,(\mathbf{p} \otimes \mathbf{p} - \mathbf{1})f^{(1)}(\mathbf{r}, \mathbf{p}, \tau)$, where $\mathbf{1}$ is the 3×3 identity matrix.

At equilibrium, this term vanishes. However, it is analogous to the kinetic energy
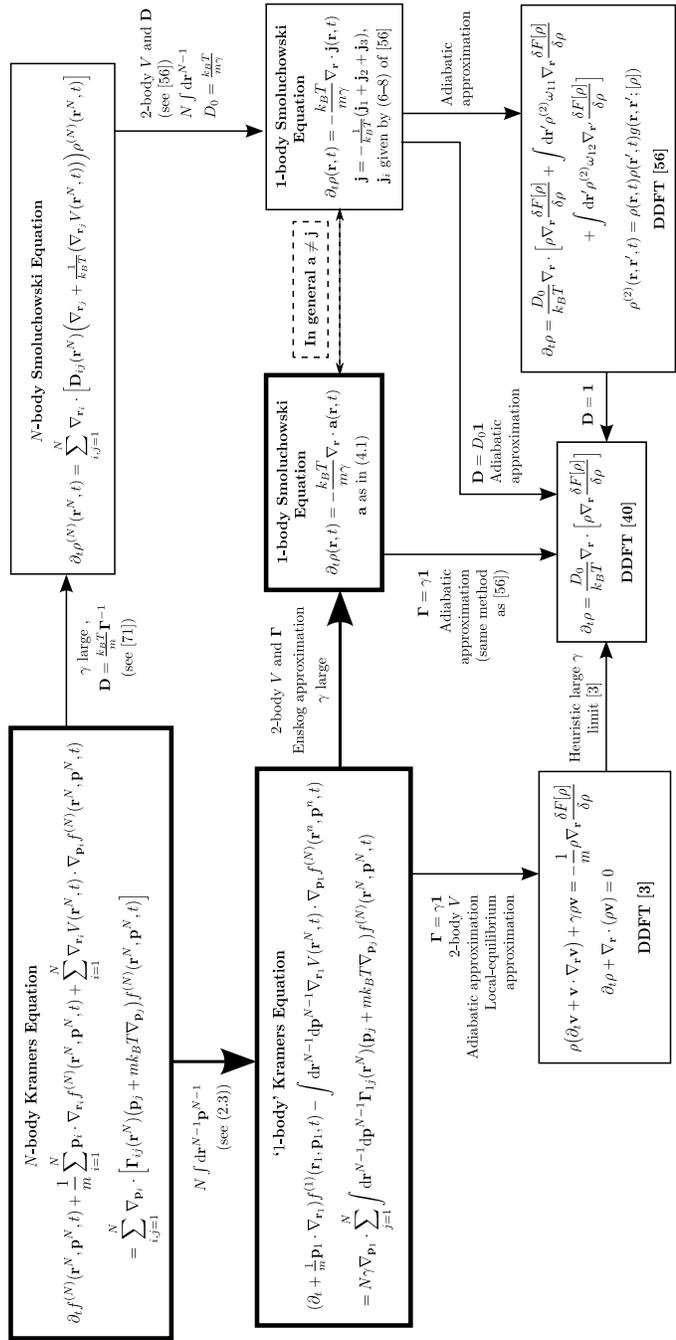
FIG. 1.1. *Flow diagram of the various approaches used to obtain one-body evolution equations and DDFTs from the full underlying dynamics. Arrows indicate the interconnectedness of the different approaches. Thick boxes/arrows: this work. Thin boxes/arrows: previous approaches. Dashed boxes/arrows: Note that the two routes produce different one-body Smoluchowski equations when hydrodynamic interactions are included, although both approaches are accurate to $\mathcal{O}(\epsilon^2)$. Text on arrows give brief descriptions of the approximations made; see the references for further details. Note in particular that the present formulation is a general one, and all existing formulations may be derived from it.*

tensor in nonequilibrium thermodynamics [36] and thus is not negligible in general. Hence, for atomic liquids [2], it has been assumed that it can be approximated by $\nu\partial_t\rho$, where ν is an arbitrary collision frequency. Although this resulted in a DDFT analogous to that previously derived for colloids in the high friction limit [40], it is not clear that this is the correct approximation in general. For colloids with no hydrodynamic interactions, this term can be dealt with using a local-equilibrium approximation, or a Taylor expansion close to equilibrium, or considering the high friction limit [3]. However, the first two approaches are unsatisfactory for general systems which may not lie close to (local) equilibrium, whilst the high friction limit was not analyzed rigorously. This high friction limit is the main objective of the present study. We will show that, in this limit, the term $\int d\mathbf{p}\,(\mathbf{p}\otimes\mathbf{p}-\mathbf{1})f^{(1)}(\mathbf{r},\mathbf{p},\tau)$ is indeed negligible compared to ρ and the momentum distribution.

It is worth noting that if hydrodynamic interactions are neglected, the heuristic high friction calculation made by Archer [3] produces the same DDFT as that derived by Marconi and Tarazona [40, 41]. We shall demonstrate that this still holds for the rigorous derivation. However, when hydrodynamic interactions are included, the two approaches do not lead to identical equations. Section 4 discusses these differences in detail.

1.3. Toward a rigorous derivation of dynamic density functional theory. Our main result is that, for a system of N identical, spherically symmetric colloid particles, up to errors of $\mathcal{O}(\epsilon^2)$, where $\epsilon\sim\gamma^{-1}$ with γ the friction constant for an infinitely dilute system (see section 2), the dynamics of the one-body position distribution ρ are given by

$$\partial_\tau\rho(\mathbf{r},\tau)=-\tfrac{k_BT}{m\gamma}\boldsymbol{\nabla}_\mathbf{r}\cdot\mathbf{a}(\mathbf{r},\tau),$$

where $\mathbf{a}$ is the solution to a particular Fredholm integral equation (Theorem 4.1), k_B is Boltzmann's constant, T is the absolute temperature, and m is the mass of the colloid particles. Furthermore, we show that if the one-body phase space distribution is written as a Hilbert [32] (or Chapman–Enskog [15]) expansion $f^{(1)}(\mathbf{r},\mathbf{p},t)=f_0(\mathbf{r},\mathbf{p},t)+\epsilon f_1(\mathbf{r},\mathbf{p},t)+\epsilon^2 f_2(\mathbf{r},\mathbf{p},t)+\cdots$, then (in appropriate units) the first two terms are of the forms $\rho_0(\mathbf{r},\tau)\,e^{-|\mathbf{p}|^2/2}$ and $\mathbf{a}(\mathbf{r},\tau)\cdot\mathbf{p}\,e^{-|\mathbf{p}|^2/2}$, respectively. In particular, nonzero terms in the integral $\int d\mathbf{p}(\mathbf{p}\otimes\mathbf{p}-\mathbf{1})f^{(1)}(\mathbf{r},\mathbf{p},t)$ are at most $\mathcal{O}(\epsilon^2)$.

We note that the evolution equation takes the form of a continuity equation. In the framework of standard fluid dynamics one would expect $\mathbf{a}(\mathbf{r},t)=\rho(\mathbf{r},t)\mathbf{v}(\mathbf{r},t)$, where ρ is the fluid density and $\mathbf{v}$ is the velocity field. Using the standard definition [3] $\mathbf{v}(\mathbf{r},t):=\rho^{-1}(\mathbf{r},t)\int d\mathbf{p}\,\mathbf{p}f^{(1)}(\mathbf{r},\mathbf{p},t)$, the Hilbert expansion (3.1), Corollary 3.3, and Lemma 3.13 show that, up to errors of $\mathcal{O}(\epsilon^2)$, this interpretation holds.

We now discuss the novelty of our approach and results. In previous work, the hydrodynamic interactions have been ignored. In this case, the leading-order term in the expansion in the inverse of the friction constant becomes linear (cf. (3.2a), where it is nonlinear), making the analysis significantly easier. As noted above, the full N-body equations are also linear. In this work, we will consider two-body hydrodynamic interactions, along with a two-body interparticle potential, which require the approximation of the two-body distribution. As will be seen, a standard approximation then leads to quadratic nonlinearities in the one-body equation, formally analogous to the quadratic nonlinearity of the collision operator in the Boltzmann equation [17, 11, 55]. For more general interactions, the nonlinearities will be of higher order; at least a priori, n-body interactions require n-body distributions. In section 4

we show that this heuristic argument does not actually hold when starting from the N-body Smoluchowski equation; higher distributions are required.

Let us now contrast the rigorous derivation of hydrodynamics from the Boltzmann equation (in the limit of small mean free path, or high collision frequency); see, e.g., the comprehensive review by Esposito, Lebowitz, and Marra [27]. The approach used therein, where time (and possibly space) are suitably rescaled and a Hilbert expansion is used to derive an infinite hierarchy of equations, which may then be solved to arbitrary order, is very similar in spirit to ours. The collision term is replaced by a term involving the hydrodynamic interactions, which has been much less widely studied than the Boltzmann collision operator. Determining the leading-order term in the Hilbert expansion requires finding the null space of the collision term (see (3.2a)). The full friction operator is a complicated integral operator, and determining its null space is nontrivial (see Lemma 3.2). In contrast, for the Boltzmann collision and self-friction operators it is straightforward to show that the null space contains only Maxwellians. Furthermore, in our situation, there are additional nonlinear terms due to the interparticle potentials. However, due to these terms being independent of $\mathbf{p}$, the momentum variable, and occurring with a higher power of the small parameter, they do not hinder the analysis in the same way as the hydrodynamic interaction terms. In addition, these nonlinearities affect our ability to control the evolution of the parts of the higher-order corrections which lie in the null space of the operator we need to invert. Sections 3.4 and 5 highlight these difficulties in detail.

The structure of the paper is summarized as follows. In section 2 we give a description of the model, in both the original and rescaled timescales, state our assumptions, and give an overview of the main result. In section 3 we develop the solvability condition for the Hilbert expansion of the one-body distribution $f^{(1)}$, which forms the basis for the proof of the main result stated in section 4, where we also discuss its relationship with existing formulations of the one-body Smoluchowski equation. In section 5 we discuss the impact of our main result, including its application to the derivation of DDFT, and also describe a number of associated open problems. Appendix A contains proofs of the more technical lemmas of section 3.

2. Description of the model and statement of main results. We begin by considering the full equations of motion, in both position and momentum, for a large number N of spherically symmetric colloid particles of mass m in a bath of a much larger number of much lighter particles. The interaction between the colloidal particles and the bath is modeled on the level of stochastic noise, and the interaction between colloidal particles mediated by the bath is modeled by friction terms. The magnitude of these two effects is correlated due to a generalized fluctuation-dissipation theorem [24, 46, 75, 25].

The evolution equations are

$$m\ddot{\mathbf{r}}_i = -\gamma m \sum_{j=1}^{N} \mathbf{\Gamma}_{ij}\dot{\mathbf{r}}_j + \mathbf{X}_i(\mathbf{r}^N) + \sum_{j=1}^{N} \sqrt{2\gamma m k_B T}\mathbf{A}_{ij}\dot{\mathbf{w}}_j,$$

where, for $\mathbf{r}^N = \mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N$, $\mathbf{\Gamma}_{ij} \in \mathbb{R}^{3\times3}(\mathbf{r}^N)$ and $\mathbf{\Gamma} = (\mathbf{\Gamma}_{ij}) \in \mathbb{R}^{3N\times3N}(\mathbf{r}^N)$ is the friction tensor, which is positive definite, and in particular has a square root. γ is the friction constant for a single isolated particle, and we are interested in the regime where $\gamma \gg 1$. The $\dot{\mathbf{w}}_j(t) = (\dot{w}_j^1(t), \dot{w}_j^2(t), \dot{w}_j^3(t))^T$ are mean zero, uncorrelated stochastic white noise terms and satisfy $\langle \dot{w}_j^n(t) \rangle = 0$ and $\langle \dot{w}_j^n(t)\dot{w}_k^m(t') \rangle = \delta_{jk}\delta_{nm}\delta(t - t')$. $\mathbf{\Gamma}$ is related to the strength of the stochastic white noise terms $\dot{\mathbf{w}}_j$ via a generalized

fluctuation-dissipation theorem, namely $\mathbf{A} = \sqrt{\boldsymbol{\Gamma}}$. $\mathbf{X}_i$ represents the force on particle i exerted by an external field and interactions with the other colloid particles. T is the absolute temperature, and k_B is Boltzmann's constant.

The motivation for this analysis is that in the high friction (overdamped, large γ) limit, the momenta should reach equilibrium on a much faster timescale than the positions. In particular, we are interested in times of $\mathcal{O}(\gamma^{-1})$ and so begin by rescaling the time variable as $t = k_B T/(m\gamma)\tau$ (where t and τ are the new and old times, respectively); then we set $\mathbf{X}_i = -\boldsymbol{\nabla}_{\mathbf{r}_i} U$ and define $\tilde{\mathbf{X}}_i := \mathbf{X}_i/(k_B T) = -\boldsymbol{\nabla}_{\mathbf{r}_i} V = -\boldsymbol{\nabla}_{\mathbf{r}_i} U/k_B T$ and $\epsilon = \sqrt{k_B T/m}\gamma^{-1}$. This rescaling leads to the following system of equations:

$$\dot{\mathbf{r}}_i = \frac{1}{\epsilon}\mathbf{p}_i,$$

$$\dot{\mathbf{p}}_i = -\frac{1}{\epsilon^2}\sum_{j=1}^N \boldsymbol{\Gamma}_{ij}(\mathbf{r}^N)\mathbf{p}_j + \frac{1}{\epsilon}\tilde{\mathbf{X}}_i(\mathbf{r}^N) + \sum_{j=1}^N \sqrt{2\epsilon^{-2}}\mathbf{A}_{ij}\dot{\mathbf{w}}_j.$$

The constant in the time rescaling corresponds physically to D_0, the diffusion constant for a single isolated particle. The rescaling of $\mathbf{X}$ corresponds to measuring potential energy in units of the temperature. For ϵ, we note that $\sqrt{k_B T/m}$ is the average thermal equilibrium speed of a particle at temperature T, whilst γ^{-1} is approximately the time required for the velocity distribution of the colloids to equilibrate. Hence ϵ has units of length, and in order to produce a nondimensional constant, it would be necessary to introduce another length scale. Such a scale is highly problem-dependent and could, for example, be the typical length over which the external potential varies, the length of a finite box in which the particles are contained, a typical separation of colloid particles, or the size of the colloids. As such, we retain the dimensional parameter ϵ and remark that the existence of a small parameter for applications must be checked on a case-by-case basis.

The corresponding Fokker–Planck equation for the N-body distribution function is

$$\partial_t f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t) + \frac{1}{\epsilon}\sum_{i=1}^N \mathbf{p}_i \cdot \boldsymbol{\nabla}_{\mathbf{r}_i} f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)$$

$$-\frac{1}{\epsilon}\sum_{i=1}^N \boldsymbol{\nabla}_{\mathbf{r}_i} V(\mathbf{r}^N) \cdot \boldsymbol{\nabla}_{\mathbf{p}_i} f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)$$

$$(2.1) \qquad = \frac{1}{\epsilon^2}\sum_{i,j=1}^N \boldsymbol{\nabla}_{\mathbf{p}_i} \cdot \left[\boldsymbol{\Gamma}_{ij}(\mathbf{r}^N)(\mathbf{p}_j + \boldsymbol{\nabla}_{\mathbf{p}_j}) f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)\right].$$

Here we have used the notation $\mathbf{r}^n = (\mathbf{r}_1, \ldots, \mathbf{r}_n)$ and the analogue for $\mathbf{p}^n$. We also find it convenient to write $\mathrm{d}\mathbf{r}^{N-n} = \mathrm{d}\mathbf{r}_{n+1}\ldots\mathrm{d}\mathbf{r}_N$ and the analogue for $\mathbf{p}$. In the above equation, $f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)$ is the probability of finding each particle i at position $\mathbf{r}_i$ with momentum $\mathbf{p}_i$ at time t. We note that the above equation is precisely the N-body Kramers equation; see, e.g., [19, 46].

It is clear that $f^{(N)}$ encodes a huge amount of information and, as such, is very computationally demanding. We are not interested in the distributions of the positions and momenta of all the identical particles but in the distribution of the average values of these quantities. For this reason we introduce the reduced probability distributions

$$(2.2) \qquad f^{(n)}(\mathbf{r}^n, \mathbf{p}^n, t) := \frac{N!}{(N-n)!}\int \mathrm{d}\mathbf{r}^{N-n}\mathrm{d}\mathbf{p}^{N-n} f^{(N)}(\mathbf{r}^N, \mathbf{p}^n, t).$$

Multiplying (2.1) by N and integrating over $\mathrm{d}\mathbf{r}^{N-1}\mathrm{d}\mathbf{p}^{N-1}$, all terms in the sums with $i \neq 1$ vanish and the evolution of the one-body distribution is given by

$$\left(\partial_t + \frac{1}{\epsilon}\mathbf{p}_1 \cdot \boldsymbol{\nabla}_{\mathbf{r}_1}\right)f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t) - \frac{N}{\epsilon}\int \mathrm{d}\mathbf{r}^{N-1}\mathrm{d}\mathbf{p}^{N-1}\boldsymbol{\nabla}_{\mathbf{r}_1}V(\mathbf{r}^N) \cdot \boldsymbol{\nabla}_{\mathbf{p}_1}f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)$$

$$(2.3) \qquad = \frac{N}{\epsilon^2}\boldsymbol{\nabla}_{\mathbf{p}_1} \cdot \sum_{j=1}^{N}\int \mathrm{d}\mathbf{r}^{N-1}\mathrm{d}\mathbf{p}^{N-1}\boldsymbol{\Gamma}_{1j}(\mathbf{r}^N)(\mathbf{p}_j + \boldsymbol{\nabla}_{\mathbf{p}_j})f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t).$$

The difficulty in solving this equation lies primarily in the fact that the last two terms still involve $f^{(N)}$, the full N-body distribution. In order to remove this dependence and obtain a closed equation, it is necessary to make some assumptions. First we assume that the potential and friction tensor contain at most two-body interactions. We will show that this is equivalent to requiring knowledge of only $f^{(2)}$. From previous studies on the derivation of DDFT (see, e.g., [14]), it is known that the full N-body distribution function $f^{(N)}$ can be written as a functional of the one-body spatial distribution, and therefore so can $f^{(2)}$. We make the Enskog approximation to the two-body distribution, in particular assuming that $f^{(2)}(\mathbf{r}_1, \mathbf{p}_1, \mathbf{r}_2, \mathbf{p}_2, t) = f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t)f^{(1)}(\mathbf{r}_2, \mathbf{p}_2, t)g(\mathbf{r}_1, \mathbf{r}_2)$, where the pair-distribution function g is assumed to be independent of $\mathbf{p}$ and ϵ.

With this assumption, (2.2) shows that $\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}f^{(1)}(\mathbf{r}, \mathbf{p}, t)g(\mathbf{r}, \mathbf{r}') = N - 1$. The role of g is to describe positional correlations of the particles, such as finite-size exclusion effects. It is an intermediate level of approximation between the mean field approximation ($g \equiv 1$) and the full two-body distribution function $f^{(2)}$. We note that if $\boldsymbol{\Gamma}_{ij} = \delta_{ij}\mathbf{1}$, the method outlined below allows a Smoluchowski equation to be derived even if g depends on $\mathbf{p}_1$ and $\mathbf{p}_2$. It seems unlikely that an analogous result holds in general, as nontrivial momentum correlations on the two-particle level would prevent the system from thermalizing to a Maxwellian momentum distribution.

The assumption that g is independent of ϵ is known not to be valid in general; we have only that $g = g(\mathbf{r}, \mathbf{r}'; [\rho])$ (g is a functional of ρ), where $\rho(\mathbf{r}, t) = \int \mathrm{d}\mathbf{p}f^{(1)}(\mathbf{r}, \mathbf{p}, t)$. This assumption does hold if the only contribution to ρ comes from the zeroth-order term in $f^{(1)}$. Even so, as we will discuss in section 5, if we expand g in a power series in ϵ, $g = g_0 + \epsilon g_1 + \cdots$, then the derivation changes only at the ϵ^0 level, and we recover an analogous Smoluchowski equation. Such an approximation is standard in the physics literature and, as suggested by the name, was first proposed by Enskog (see [10]) and revised by Van Beijeren and Ernst [72] to ensure consistency with irreversible thermodynamics. See also [55].

To summarize, we make the following assumptions:

- *Assumption* 1 (pairwise additive potential).

$$V(\mathbf{r}^N, t) = \sum_{i=1}^{N}V_1(\mathbf{r}_i, t) + \frac{1}{2}\sum_{i \neq j}V_2(\mathbf{r}_i, \mathbf{r}_j).$$

- *Assumption* 2 (pairwise additive friction).

$$\boldsymbol{\Gamma}_{ij} = \delta_{ij}\mathbf{1} + \tilde{\boldsymbol{\Gamma}}_{ij} = \delta_{ij}\left(\mathbf{1} + \sum_{\ell \neq i}\mathbf{Z}_1(\mathbf{r}_i, \mathbf{r}_\ell)\right) + (1 - \delta_{ij})\mathbf{Z}_2(\mathbf{r}_i, \mathbf{r}_j)$$

$$= \delta_{ij}\sum_{\ell \neq i}\left(\tfrac{1}{N-1}\mathbf{1} + \mathbf{Z}_1(\mathbf{r}_i, \mathbf{r}_\ell)\right) + (1 - \delta_{ij})\mathbf{Z}_2(\mathbf{r}_i, \mathbf{r}_j),$$

with the $\mathbf{Z}_j$ symmetric 3×3 matrices.

- *Assumption* 3 (Enskog approximation).

$$(2.4) \qquad f^{(2)}(\mathbf{r}_1, \mathbf{p}_1, \mathbf{r}_2, \mathbf{p}_2, t) = f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t) f^{(1)}(\mathbf{r}_2, \mathbf{p}_2, t) g(\mathbf{r}_1, \mathbf{r}_2).$$

Making these assumptions, we now calculate the two remaining terms in (2.3). Using standard symmetry arguments gives

$$N \int d\mathbf{r}^{N-1} d\mathbf{p}^{N-1} \boldsymbol{\nabla}_{\mathbf{r}_1} V(\mathbf{r}^N) \cdot \boldsymbol{\nabla}_{\mathbf{p}_1} f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)$$

$$= \left[\boldsymbol{\nabla}_{\mathbf{r}_1} V_1(\mathbf{r}_1, t) + \int d\mathbf{r}_2 d\mathbf{p}_2 g(\mathbf{r}_1, \mathbf{r}_2) f^{(1)}(\mathbf{r}_2, \mathbf{p}_2, t) \boldsymbol{\nabla}_{\mathbf{r}_1} V_2(\mathbf{r}_1, \mathbf{r}_2) \right] \cdot \boldsymbol{\nabla}_{\mathbf{p}_1} f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t)$$

and

$$N \boldsymbol{\nabla}_{\mathbf{p}_1} \cdot \sum_{j=1}^{N} \int d\mathbf{r}^{N-1} d\mathbf{p}^{N-1} \boldsymbol{\Gamma}_{1j}(\mathbf{r}^N)(\mathbf{p}_j + \boldsymbol{\nabla}_{\mathbf{p}_j}) f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)$$

$$= \boldsymbol{\nabla}_{\mathbf{p}_1} \cdot \Bigg[(\mathbf{p}_1 + \boldsymbol{\nabla}_{\mathbf{p}_1}) f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t)$$

$$+ \int d\mathbf{r}_2 d\mathbf{p}_2 g(\mathbf{r}_1, \mathbf{r}_2) f^{(1)}(\mathbf{r}_2, \mathbf{p}_2, t) \mathbf{Z}_1(\mathbf{r}_1, \mathbf{r}_2) \times (\mathbf{p}_1 + \boldsymbol{\nabla}_{\mathbf{p}_1}) f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t)$$

$$+ \int d\mathbf{r}_2 d\mathbf{p}_2 g(\mathbf{r}_1, \mathbf{r}_2) \mathbf{Z}_2(\mathbf{r}_1, \mathbf{r}_2)(\mathbf{p}_2 + \boldsymbol{\nabla}_{\mathbf{p}_2}) f^{(1)}(\mathbf{r}_2, \mathbf{p}_2, t) \times f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t) \Bigg].$$

Hence, we have the following proposition.

PROPOSITION 2.1. *Under Assumptions* 1, 2, *and* 3, *the evolution of the one-body reduced distribution satisfies*

$$\partial_t f^{(1)}(\mathbf{r}, \mathbf{p}, t) = \frac{1}{\epsilon} \Bigg[-\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} + \boldsymbol{\nabla}_{\mathbf{r}} V_1(\mathbf{r}, t) \cdot \boldsymbol{\nabla}_{\mathbf{p}}$$

$$+ \int d\mathbf{r}' d\mathbf{p}' f^{(1)}(\mathbf{r}', \mathbf{p}', t) g(\mathbf{r}, \mathbf{r}') \boldsymbol{\nabla}_{\mathbf{r}} V_2(\mathbf{r}, \mathbf{r}') \cdot \boldsymbol{\nabla}_{\mathbf{p}} \Bigg] f^{(1)}(\mathbf{r}, \mathbf{p}, t)$$

$$+ \frac{1}{\epsilon^2} \boldsymbol{\nabla}_{\mathbf{p}} \cdot \Bigg[(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f^{(1)}(\mathbf{r}, \mathbf{p}, t)$$

$$+ \int d\mathbf{r}' d\mathbf{p}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') f^{(1)}(\mathbf{r}', \mathbf{p}', t) \times (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f^{(1)}(\mathbf{r}, \mathbf{p}, t)$$

$$+ \int d\mathbf{r}' d\mathbf{p}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'}) f^{(1)}(\mathbf{r}', \mathbf{p}', t) \times f^{(1)}(\mathbf{r}, \mathbf{p}, t) \Bigg]$$

$$(2.5) \qquad =: \frac{1}{\epsilon} \left[\mathcal{L}_1 f^{(1)} + \mathcal{N}_1(f^{(1)}, f^{(1)}) \right] + \frac{1}{\epsilon^2} \left[\mathcal{L}_0 f^{(1)} + \mathcal{N}_0(f^{(1)}, f^{(1)}) \right],$$

where

$$(2.6a) \quad \mathcal{L}_0 f = \boldsymbol{\nabla}_{\mathbf{p}} \cdot (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t),$$

$$(2.6b) \quad \mathcal{L}_1 f = [-\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} + \boldsymbol{\nabla}_{\mathbf{r}} V_1(\mathbf{r}, t) \cdot \boldsymbol{\nabla}_{\mathbf{p}}] f(\mathbf{r}, \mathbf{p}, t),$$

$(2.6c)$

$$\mathcal{N}_0(f, \tilde{f}) = \boldsymbol{\nabla}_{\mathbf{p}} \cdot \int d\mathbf{r}' d\mathbf{p}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') f(\mathbf{r}', \mathbf{p}', t) \times (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \tilde{f}(\mathbf{r}, \mathbf{p}, t)$$

$$+ \boldsymbol{\nabla}_{\mathbf{p}} \cdot \int d\mathbf{r}' d\mathbf{p}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'}) f(\mathbf{r}', \mathbf{p}', t) \times \tilde{f}(\mathbf{r}, \mathbf{p}, t),$$

$(2.6d)$

$$\mathcal{N}_1(f, \tilde{f}) = \int d\mathbf{r}' d\mathbf{p}' f(\mathbf{r}', \mathbf{p}', t) g(\mathbf{r}, \mathbf{r}') \boldsymbol{\nabla}_{\mathbf{r}} V_2(\mathbf{r}, \mathbf{r}') \cdot \boldsymbol{\nabla}_{\mathbf{p}} \tilde{f}(\mathbf{r}, \mathbf{p}, t),$$

and for ease of notation we have omitted the $(\mathbf{r}, \mathbf{p}, t)$ *dependence of the functions on the left-hand sides of* (2.6a)–(2.6d).

It is noteworthy that, although they are quadratic, the nonlinear terms are not symmetric in the two arguments. The first argument has been taken to be that inside the integral.

In the following we will assume that $f^{(1)}$ is bounded and positive[1] and that all functions are sufficiently regular and have sufficient decay at infinity for operators and integrals to be defined.

To state our main result, we recall that the position distribution, which is the object of interest in the one-particle Smoluchowski regime, is defined by $\rho(\mathbf{r}, t) = \int \mathrm{d}\mathbf{p} f^{(1)}(\mathbf{r}, \mathbf{p}, t)$. We will show that its evolution equation is given by the following theorem.

THEOREM 2.2 (Smoluchowski equation). *Under suitable assumptions on* $f^{(1)}$, U_j, $\mathbf{Z}_j$, $j = 1, 2$ *(see Theorem* 4.1*), up to errors of* $\mathcal{O}(\epsilon^2)$ *the dynamics of the one-body position distribution are given (in the original timescale) by*

$$\partial_\tau \rho(\mathbf{r}, \tau) = -\tfrac{k_B T}{m\gamma} \boldsymbol{\nabla}_\mathbf{r} \cdot \mathbf{a}(\mathbf{r}, \tau),$$

where $\mathbf{a}(\mathbf{r}, \tau)$ *is the solution to*

$$\mathbf{a}(\mathbf{r}, \tau) + \int \mathrm{d}\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \rho(\mathbf{r}', \tau) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \times \mathbf{a}(\mathbf{r}, \tau) + \rho(\mathbf{r}, \tau) \int \mathrm{d}\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}') \mathbf{a}(\mathbf{r}', \tau)$$

$$(2.7) \quad = -\left[\boldsymbol{\nabla}_\mathbf{r} + \tfrac{1}{k_B T} \left(\boldsymbol{\nabla}_\mathbf{r} U_1(\mathbf{r}, \tau) + \int \mathrm{d}\mathbf{r}' \rho(\mathbf{r}', \tau) g(\mathbf{r}, \mathbf{r}') \boldsymbol{\nabla}_\mathbf{r} U_2(\mathbf{r}, \mathbf{r}') \right) \right] \rho(\mathbf{r}, \tau).$$

3. The Hilbert expansion. We now expand $f^{(1)}$ in powers of ϵ as

$$(3.1) \qquad\qquad f^{(1)}(\mathbf{r}, \mathbf{p}, t) = \sum_{n=0}^{\infty} \epsilon^n f_n(\mathbf{r}, \mathbf{p}, t).$$

Due to the singular nature of the problem, we do not expect such a regular perturbation expansion to converge uniformly. The expansion should be valid only for times $t \gg \epsilon$ and not for shorter times; i.e., we expect there to be a boundary layer in time of size $\mathcal{O}(\epsilon)$. Since we are interested in times much larger than ϵ, interest lies in the leading-order terms; one would then hope to be able to truncate the series and prove suitable bounds on the remainder term, as in [27]. We also assume that such an expansion then converges, in particular, that the f_n are sufficiently well behaved in $\mathbf{r}$ and $\mathbf{p}$.

Inserting (3.1) into the evolution equation (2.5) and collecting powers of ϵ gives the hierarchy of equations

$$(3.2a) \qquad\qquad \mathcal{L}_0 f_0 + \mathcal{N}_0(f_0, f_0) = 0,$$

$$(3.2b) \qquad \mathcal{L}_0 f_1 + \mathcal{L}_1 f_0 + \mathcal{N}_0(f_0, f_1) + \mathcal{N}_0(f_1, f_0) + \mathcal{N}_1(f_0, f_0) = 0,$$

$$\mathcal{L}_0 f_2 + \mathcal{L}_1 f_1 + \mathcal{N}_0(f_2, f_0) + \mathcal{N}_0(f_0, f_2) + \mathcal{N}_0(f_1, f_1)$$
$$(3.2c) \qquad\qquad\qquad + \mathcal{N}_1(f_1, f_0) + \mathcal{N}_1(f_0, f_1) = \partial_t f_0,$$

$$\mathcal{L}_0 f_n + \mathcal{L}_1 f_{n-1} + \sum_{i+j=n} \mathcal{N}_0(f_i, f_j) + \sum_{i+j=n-1} \mathcal{N}_1(f_i, f_j) = \partial_t f_{n-2}, \quad n \geq 3.$$

---

[1] $f^{(1)}$ is nonnegative by definition, but it may be zero, e.g., if there are excluded areas of the phase space due to confining potentials. It is clear that it may be made positive with arbitrarily small errors.

We now solve these equations order by order. First, to solve (3.2a), we need to determine the null space of $\mathcal{L}_0 \cdot + \mathcal{N}_0(\cdot, \cdot)$. However, before we do so, the following lemma will be useful. It is essentially a result of the positive definiteness of $\boldsymbol{\Gamma}$.

LEMMA 3.1. *For $\mathbf{v}(\mathbf{r}, \mathbf{p}, t)$ an arbitrary vector such that the integrals below exist, and for f satisfying* (2.4)*, there exists $\delta > 0$ such that*

$$\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}\mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' f(\mathbf{r}, \mathbf{p}, t) f(\mathbf{r}', \mathbf{p}', t) g(\mathbf{r}, \mathbf{r}')$$
$$\times \left[\mathbf{v}(\mathbf{r}, \mathbf{p}, t) \cdot \left(\tfrac{1}{N-1}\mathbf{1} + \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \right) \mathbf{v}(\mathbf{r}, \mathbf{p}, t) + \mathbf{v}(\mathbf{r}, \mathbf{p}, t) \cdot \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')\mathbf{v}(\mathbf{r}', \mathbf{p}', t) \right]$$
$$\geq \delta \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f(\mathbf{r}, \mathbf{p}, t)|\mathbf{v}(\mathbf{r}, \mathbf{p}, t)|^2.$$

In particular, the result holds when f is chosen to be either f_0 or $f^{(1)}$.

Proof. See section A.1. $\quad\square$

We now proceed with the analysis of (3.2a)–(3.2c), beginning by determining the solution to (3.2a).

3.1. Solution of the ϵ^{-2} equation. In this section we find the solution f_0 of (3.2a).

LEMMA 3.2. *For f satisfying* (2.4)*, the null space of $\mathcal{L}_0 f + \mathcal{N}_0(f, f)$ consists of functions of the form $f(\mathbf{r}, \mathbf{p}, t) = \exp\left(-|\mathbf{p}|^2/2\right)\phi(\mathbf{r}, t)$.*

Proof. We begin by assuming that f satisfies (2.4) and $\mathcal{L}_0 f + \mathcal{N}_0(f, f) = 0$. We define ϕ by $f(\mathbf{r}, \mathbf{p}, t) =: \mathrm{e}^{-|\mathbf{p}|^2/2}\,\phi(\mathbf{r}, \mathbf{p}, t)$ and note that ϕ is positive. We therefore have $\ln\phi[\mathcal{L}_0 f + \mathcal{N}_0(f, f)] = 0$. Note that $f(\mathbf{r}, \mathbf{p}, t) = \frac{1}{N-1}\int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' f^{(2)}(\mathbf{r}, \mathbf{p}, \mathbf{r}', \mathbf{p}', t)$ and so

$$\mathcal{L}_0 f(\mathbf{r}, \mathbf{p}, t) = \tfrac{1}{N-1}\boldsymbol{\nabla}_{\mathbf{p}} \cdot \int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f(\mathbf{r}', \mathbf{p}', t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t).$$

Using this reformulation, along with the definition (2.6c) and the notation $\tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}') := \tfrac{1}{N-1}\mathbf{1} + \mathbf{Z}_1(\mathbf{r}, \mathbf{r}')$, and integrating over $\mathbf{r}$ and $\mathbf{p}$ gives

$$0 = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}\,\ln\phi(\mathbf{r}, \mathbf{p}, t)\boldsymbol{\nabla}_{\mathbf{p}} \cdot \int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}')\Big[f(\mathbf{r}', \mathbf{p}')\tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}')(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)$$
$$+ f(\mathbf{r}, \mathbf{p}, t)\mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'}) f(\mathbf{r}', \mathbf{p}', t) \Big]$$
$$= -\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}\mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}'\, \mathrm{e}^{-|\mathbf{p}|^2/2}\,\mathrm{e}^{-|\mathbf{p}'|^2/2}\, g(\mathbf{r}, \mathbf{r}')\frac{\boldsymbol{\nabla}_{\mathbf{p}}\phi(\mathbf{r}, \mathbf{p}, t)}{\phi(\mathbf{r}, \mathbf{p}, t)}$$
$$\times \Big[\phi(\mathbf{r}', \mathbf{p}', t)\tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}')\boldsymbol{\nabla}_{\mathbf{p}}\phi(\mathbf{r}, \mathbf{p}, t) + \phi(\mathbf{r}, \mathbf{p}, t)\mathbf{Z}_2(\mathbf{r}, \mathbf{r}')\boldsymbol{\nabla}_{\mathbf{p}'}\phi(\mathbf{r}', \mathbf{p}', t) \Big],$$

where we have used integration by parts and Fubini's theorem, along with the identity $(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}})[\,\mathrm{e}^{-|\mathbf{p}|^2/2}\,\phi(\mathbf{r}, \mathbf{p}, t)] = \mathrm{e}^{-|\mathbf{p}|^2/2}\,\boldsymbol{\nabla}_{\mathbf{p}}\phi(\mathbf{r}, \mathbf{p}, t)$.

Letting $\mathbf{v}(\mathbf{r}, \mathbf{p}, t) := \boldsymbol{\nabla}_{\mathbf{p}}\phi(\mathbf{r}, \mathbf{p}, t)/\phi(\mathbf{r}, \mathbf{p}, t)$ we have

$$0 = -\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}\mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}'\, \mathrm{e}^{-|\mathbf{p}|^2/2}\,\mathrm{e}^{-|\mathbf{p}'|^2/2}\,\phi(\mathbf{r}, \mathbf{p}, t)\phi(\mathbf{r}', \mathbf{p}', t) g(\mathbf{r}, \mathbf{r}')$$
$$\times \Big[\mathbf{v}(\mathbf{r}, \mathbf{p}, t) \cdot \tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}')\mathbf{v}(\mathbf{r}, \mathbf{p}, t) + \mathbf{v}(\mathbf{r}, \mathbf{p}, t) \cdot \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')\mathbf{v}(\mathbf{r}', \mathbf{p}', t) \Big].$$

Since $f(\mathbf{r}, \mathbf{p}, t) = \exp\left(-|\mathbf{p}|^2/2\right)\phi(\mathbf{r}, \mathbf{p}, t) > 0$, we may apply Lemma 3.1, which shows that

$$0 = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f(\mathbf{r}, \mathbf{p}, t)|\mathbf{v}(\mathbf{r}, \mathbf{p}, t)|^2 = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}\, \mathrm{e}^{-|\mathbf{p}|^2/2}\,\phi^{-1}(\mathbf{r}, \mathbf{p}, t)|\boldsymbol{\nabla}_{\mathbf{p}}\phi(\mathbf{r}, \mathbf{p}, t)|^2.$$

Hence, since ϕ is bounded, the integrand is zero if and only if $\boldsymbol{\nabla_p}\phi(\mathbf{r}, \mathbf{p}, t) \equiv 0$ and the result holds. $\square$

This result gives an explicit form for the $\mathbf{p}$-dependence of f_0.

COROLLARY 3.3. *The zeroth-order term in the ϵ-expansion of $f^{(1)}$ is given by*

$$f_0(\mathbf{r}, \mathbf{p}, t) = \frac{1}{(2\pi)^{3/2}} \, \mathrm{e}^{-\frac{|\mathbf{p}|^2}{2}} \, \rho_0(\mathbf{r}, t) =: Z^{-1} \, \mathrm{e}^{-\frac{|\mathbf{p}|^2}{2}} \, \rho_0(\mathbf{r}, t).$$

Proof. The proof follows immediately from (3.2a) and Lemma 3.2. $\square$

3.2. Solution of the ϵ^{-1} equation. In order to find f_1 from (3.2b), we rewrite it as

$$(3.3) \qquad \mathcal{L}_0 f_1 + \mathcal{N}_0(f_0, f_1) + \mathcal{N}_0(f_1, f_0) = -\mathcal{L}_1 f_0 - \mathcal{N}_1(f_0, f_0).$$

The first point to note is that for known f_0, this is a linear-operator equation for f_1. Although this can be seen in an abstract sense from the nonlinearities being of a quadratic nature, we will require the explicit form of the operator.

LEMMA 3.4. *For $\mathcal{N}_0$ as in (2.6c), $f_0(\mathbf{r}, \mathbf{p}, t)$ as given by Corollary 3.3, and arbitrary $\tilde{f}(\mathbf{r}, \mathbf{p}, t)$,*

$$\mathcal{N}_0(f_0, \tilde{f}) = \boldsymbol{\nabla_p} \cdot \left[\int \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \times (\mathbf{p} + \boldsymbol{\nabla_p}) \tilde{f}(\mathbf{r}, \mathbf{p}, t) \right],$$

$$\mathcal{N}_0(\tilde{f}, f_0) = -f_0(\mathbf{r}, \mathbf{p}, t) \mathbf{p} \cdot \int \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla_{p'}}) \tilde{f}(\mathbf{r}', \mathbf{p}', t).$$

Proof. For $\mathcal{N}_0(f_0, \tilde{f})$, note that $(\mathbf{p} + \boldsymbol{\nabla_p}) \mathrm{e}^{-|\mathbf{p}|^2/2} = 0$, and hence the second line in (2.6c) gives zero and the first result follows. For $\mathcal{N}_0(\tilde{f}, f_0)$, the same argument shows that the first line in (2.6c) is zero. For the remaining term, the result of the integral is a vector which depends only on $\mathbf{r}$, and for any such vector $\mathbf{z}$, $\boldsymbol{\nabla_p} \cdot [\mathbf{z}(\mathbf{r}) f(\mathbf{r}, \mathbf{p}, t)] = \mathbf{z}(\mathbf{r}) \cdot \boldsymbol{\nabla_p} f(\mathbf{r}, \mathbf{p}, t)$. The result then follows from the identity $\boldsymbol{\nabla_p} f_0(\mathbf{r}, \mathbf{p}, t) = -\mathbf{p} f_0(\mathbf{r}, \mathbf{p}, t)$. $\square$

COROLLARY 3.5. *For known f_0, and for arbitrary f, $\tilde{\mathcal{L}} f := \mathcal{L}_0 f + \mathcal{N}_0(f_0, f) + \mathcal{N}_0(f, f_0)$ is a linear operator on f.*

As is customary for problems of this form, it is convenient to work in the L^2 space weighted by the inverse of the invariant measure of $\tilde{\mathcal{L}}$. In this case, the inner product is defined as

$$\langle f(\mathbf{r}, \mathbf{p}, t), \tilde{f}(\mathbf{r}, \mathbf{p}, t) \rangle_{f_0^{-1}} := \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) f(\mathbf{r}, \mathbf{p}, t) \tilde{f}(\mathbf{r}, \mathbf{p}, t).$$

We denote this space by $L^2_{f_0^{-1}}$. In this weighted space, $\tilde{\mathcal{L}}$ is self-adjoint and has compact resolvent, allowing us to apply Fredholm's theory.

LEMMA 3.6. $\tilde{\mathcal{L}}$ *is self-adjoint in $L^2_{f_0^{-1}}$.*

Proof. See section A.2. $\square$

LEMMA 3.7. *The resolvent of $\tilde{\mathcal{L}}$ is compact in $L^2_{f_0^{-1}}$.*

Proof. See section A.3. $\square$

Since (3.3) may be rewritten as $\tilde{\mathcal{L}} f_1 = -\tilde{\mathcal{L}}_1 f_0 - \mathcal{N}_1(f_0, f_0)$, to determine the solvability condition we must determine the null space of $\tilde{\mathcal{L}}^*$.

LEMMA 3.8. *The null space of $\tilde{\mathcal{L}}$ (and of $\tilde{\mathcal{L}}^*$) contains only functions of the form* $f(\mathbf{r}, \mathbf{p}, t) = \exp\left(-|\mathbf{p}|^2/2\right)\phi(\mathbf{r}, t)$.

Proof. The proof is analogous to that of Lemma 3.2 but is more straightforward, as the operator is now linear and we are already working in the appropriate weighted space. As in the proof of Lemma 3.2, we have

$$\mathcal{L}_0 f(\mathbf{r}, \mathbf{p}, t) = \tfrac{1}{N-1} \boldsymbol{\nabla}_{\mathbf{p}} \cdot \int \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t),$$

and hence, setting $\tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}') := \tfrac{1}{N-1} \mathbf{1} + \mathbf{Z}_1(\mathbf{r}, \mathbf{r}')$, we have

$$\tilde{\mathcal{L}} f(\mathbf{r}, \mathbf{p}, t) = \boldsymbol{\nabla}_{\mathbf{p}} \cdot \int \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t) \tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}')(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)$$
$$+ g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}, \mathbf{p}, t) \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'}) f(\mathbf{r}', \mathbf{p}', t) = 0.$$

Taking the $L^2_{f_0^{-1}}$ inner product with f gives

$$0 = \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) f(\mathbf{r}, \mathbf{p}, t)$$
$$\times \boldsymbol{\nabla}_{\mathbf{p}} \cdot \int \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' \Big[g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t) \tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}')(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)$$
$$+ g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}, \mathbf{p}, t) \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'}) f(\mathbf{r}', \mathbf{p}', t) \Big].$$

Integrating by parts and using the identity $\boldsymbol{\nabla}_{\mathbf{p}} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) = \mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t)$ and Fubini's theorem gives

$$0 = \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' f_0^{-1}(\mathbf{r}, \mathbf{p}, t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)$$
$$\cdot \Big[g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t) \tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}')(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)$$
$$+ g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}, \mathbf{p}, t) \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'}) f(\mathbf{r}', \mathbf{p}', t) \Big].$$

Setting $\mathbf{v}(\mathbf{r}, \mathbf{p}, t) := f_0^{-1}(\mathbf{r}, \mathbf{p}, t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)$ then gives

$$0 = \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' f_0(\mathbf{r}, \mathbf{p}, t) f_0(\mathbf{r}', \mathbf{p}', t) g(\mathbf{r}, \mathbf{r}') \mathbf{v}(\mathbf{r}, \mathbf{p}, t)$$
$$\cdot \big[\tilde{\mathbf{Z}}_1(\mathbf{r}, \mathbf{r}') \mathbf{v}(\mathbf{r}, \mathbf{p}, t) + \mathbf{Z}_2(\mathbf{r}, \mathbf{r}') \mathbf{v}(\mathbf{r}', \mathbf{p}') \big].$$

Hence, by Lemma 3.1, we have

$$0 = \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} f_0(\mathbf{r}, \mathbf{p}, t) |\mathbf{v}(\mathbf{r}, \mathbf{p}, t)|^2 = \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) |(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)|^2.$$

Since f_0 is bounded and positive, we must have $(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t) \equiv 0$. Writing $f(\mathbf{r}, \mathbf{p}, t) = Z^{-1} \exp(-|\mathbf{p}|^2/2) \phi(\mathbf{r}, \mathbf{p}, t)$ then gives $\boldsymbol{\nabla}_{\mathbf{p}} \phi(\mathbf{r}, \mathbf{p}, t) \equiv 0$, and the result follows. $\square$

Determining the explicit solvability condition finally requires the explicit calculation of the right-hand side of (3.3).

LEMMA 3.9. *For $\mathcal{L}_1$ and $\mathcal{N}_1$ as in (2.6b) and (2.6d), and for f_0 as in Corollary 3.3, we have*

$$\mathcal{L}_1 f_0 = -Z^{-1} \mathrm{e}^{-\frac{|\mathbf{p}|^2}{2}} \mathbf{p} \cdot \big[\boldsymbol{\nabla}_{\mathbf{r}} + \boldsymbol{\nabla}_{\mathbf{r}} V_1(\mathbf{r}, t) \big] \rho_0(\mathbf{r}, t)$$

and

$$\mathcal{N}_1(f_0, f_0)(\mathbf{r}, \mathbf{p}, t) = -Z^{-1} \mathrm{e}^{-\frac{|\mathbf{p}|^2}{2}} \rho_0(\mathbf{r}, t)\mathbf{p} \cdot \int \mathrm{d}\mathbf{r}' \rho_0(\mathbf{r}', t)g(\mathbf{r}, \mathbf{r}')\boldsymbol{\nabla}_\mathbf{r} V_2(\mathbf{r}, \mathbf{r}').$$

Proof. This is a simple calculation: For $\mathcal{L}_1$ we have

$$\mathcal{L}_1 f_0(\mathbf{r}, \mathbf{p}, t) = \left[-\mathbf{p} \cdot \boldsymbol{\nabla}_\mathbf{r} + \boldsymbol{\nabla}_\mathbf{r} V_1(\mathbf{r}, t) \cdot \boldsymbol{\nabla}_\mathbf{p}\right] \left[Z^{-1} \mathrm{e}^{-\frac{|\mathbf{p}|^2}{2}} \rho_0(\mathbf{r}, t)\right]$$

$$= Z^{-1} \mathrm{e}^{-\frac{|\mathbf{p}|^2}{2}} \left[-\mathbf{p} \cdot \boldsymbol{\nabla}_\mathbf{r} \rho_0(\mathbf{r}, t)\right] + \boldsymbol{\nabla}_\mathbf{r} V_1(\mathbf{r}, t) \cdot \left[-\mathbf{p}\right] Z^{-1} \mathrm{e}^{-\frac{|\mathbf{p}|^2}{2}} \rho_0(\mathbf{r}, t),$$

and the result follows.

The result for $\mathcal{N}_1$ follows from the two identities $\int \mathrm{d}\mathbf{p} f_0(\mathbf{r}, \mathbf{p}, t) = \rho_0(\mathbf{r}, t)$ and $\boldsymbol{\nabla}_\mathbf{p} f_0(\mathbf{r}, \mathbf{p}, t) = -\mathbf{p}\, \mathrm{e}^{-|\mathbf{p}|^2/2} \rho_0(\mathbf{r}, t)$. $\quad\square$

Recall that we are trying to solve (3.3), and we have shown that in $L^2_{f_0^{-1}}$, $\tilde{\mathcal{L}}$ is self-adjoint with compact resolvent and has null space elements $\exp(-|\mathbf{p}|^2/2)\phi(\mathbf{r}, t)$. In order for (3.3) to be soluble, we therefore require, by the Fredholm alternative, that its inner product (in $L^2_{f_0^{-1}}$) with any element of the null space of $\tilde{\mathcal{L}}^*$ be zero.

Note that $\left\langle \mathrm{e}^{-|\mathbf{p}|^2/2} \phi(\mathbf{r}, t), f\right\rangle_{f_0^{-1}} = \langle \rho_0(\mathbf{r}, t)\phi(\mathbf{r}, t), f\rangle$, and we therefore require that the integral with respect to $\mathbf{p}$ of the right-hand side of (3.3) be zero. This is an easy corollary of Lemma 3.9 since the $\mathbf{p}$-dependence of both terms is of the form $\exp(-|\mathbf{p}|^2/2)\mathbf{p}$, which integrates to zero.

COROLLARY 3.10. $-\mathcal{L}_1 f_0 - \mathcal{N}_1(f_0, f_0)$ *is orthogonal to the null space of* $\tilde{\mathcal{L}}^*$, *and thus* (3.3) *always has a solution.*

Since we now know that (3.3) is soluble, we can invert its left-hand side. The standard approach would be to expand in a basis of the eigenfunctions of $\tilde{\mathcal{L}}$. However, since $\mathbf{Z}_1$ and $\mathbf{Z}_2$ (which enter $\tilde{\mathcal{L}}$ through $\mathcal{N}_0$) are unknown, we expand in a basis of products of generalized Hermite polynomials multiplied by a Maxwellian, which are eigenfunctions for the case $\mathbf{Z}_1 = \mathbf{Z}_2 = 0$. This turns out to be sufficient, as we do not need to explicitly invert $\tilde{\mathcal{L}}$.

DEFINITION 3.11. *We define the basis of* $L^2(\mathbb{R}^3, \mathrm{e}^{-|\mathbf{p}|^2/2})$, *for* $\mathbf{p} = (p_1, p_2, p_3)^\mathrm{T}$,

$$P_{n,\mathbf{a}}(\mathbf{p}) := H_{a_1}(p_1)H_{a_2}(p_2)H_{a_3}(p_3),$$

where $n \in \mathbb{N}$, $\mathbf{a} = (a_1, a_2, a_3)^\mathrm{T}$, $a_i \in \mathbb{N}$, $|\mathbf{a}| := \sum a_i = n$, *and* H_n *are the standard one-dimensional Hermite polynomials.*

Since $\mathbb{R}^3$ is a product space and the Hermite polynomials form an orthogonal basis of $L^2(\mathbb{R}, \mathrm{e}^{-p^2/2})$, it is clear that the $P_{n,\mathbf{a}}$ form an orthogonal basis of $L^2(\mathbb{R}^3, \mathrm{e}^{-|\mathbf{p}|^2/2})$ (see, e.g., [23]). We now show that $\tilde{\mathcal{L}}$ preserves n, the degree of the Hermite polynomial, when applied to $\mathrm{e}^{-|\mathbf{p}|^2/2} P_{n,\mathbf{a}}(\mathbf{p})$. Hence, with a slight abuse of notation, we find

$$\tilde{\mathcal{L}}^{-1}[\mathrm{e}^{-|\mathbf{p}|^2/2} P_{n,\mathbf{a}}(\mathbf{p})] \in \mathrm{Span}\big\{\{\mathrm{e}^{-|\mathbf{p}|^2/2} P_{n,\mathbf{b}}(\mathbf{p}) \,|\, |\mathbf{b}| = n\} \cup \mathrm{e}^{-|\mathbf{p}|^2/2}\big\},$$

where the span runs over coefficients in $\mathbf{r}$ and t, and we note that $\mathrm{e}^{-|\mathbf{p}|^2/2}$ is the $\mathbf{p}$-dependent part of the kernel of $\mathcal{L}$.

LEMMA 3.12. *Denote the expansions of $\tilde{\mathcal{L}}f$ and f by*

$$\tilde{\mathcal{L}}f =: \mathrm{e}^{-|\mathbf{p}|^2/2} \sum_{n=0}^{\infty} \sum_{|\mathbf{a}|=n} \tilde{\gamma}_{n,\mathbf{a}}(\mathbf{r},t) P_{n,\mathbf{a}}(\mathbf{p}),$$

$$f =: \mathrm{e}^{-|\mathbf{p}|^2/2} \sum_{n=0}^{\infty} \sum_{|\mathbf{a}|=n} \gamma_{n,\mathbf{a}}(\mathbf{r},t) P_{n,\mathbf{a}}(\mathbf{p}).$$

Then, for each fixed $n \geq 1$, $\gamma_{n,\mathbf{a}}(\mathbf{r},t) \equiv 0$ for all $\mathbf{a}$ if and only if $\tilde{\gamma}_{n,\mathbf{a}}(\mathbf{r},t) \equiv 0$ for all $\mathbf{a}$.

Proof. See section A.4. □

We are now in the position to determine the explicit form of f_1 and thus solve (3.2b).

LEMMA 3.13. $f_1(\mathbf{r},\mathbf{p},t) = [\mathbf{a}(\mathbf{r},t) \cdot \mathbf{p} + \psi(\mathbf{r},t)]Z^{-1}\exp(-|\mathbf{p}|^2/2)$, *where $\mathbf{a}(\mathbf{r},t)$ is given by the solution of*

$$\mathbf{a}(\mathbf{r},t) + \int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\rho_0(\mathbf{r}',t)\mathbf{Z}_1(\mathbf{r},\mathbf{r}') \times \mathbf{a}(\mathbf{r},t) + \rho_0(\mathbf{r},t)\int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\mathbf{Z}_2(\mathbf{r},\mathbf{r}')\mathbf{a}(\mathbf{r}',t)$$

$$= -\left[\boldsymbol{\nabla}_\mathbf{r} + \boldsymbol{\nabla}_\mathbf{r}V_1(\mathbf{r},t) + \int \mathrm{d}\mathbf{r}'\rho_0(\mathbf{r}',t)g(\mathbf{r},\mathbf{r}')\boldsymbol{\nabla}_\mathbf{r}V_2(\mathbf{r},\mathbf{r}')\right]\rho_0(\mathbf{r},t).$$

Proof. By (3.3) and Lemma 3.9 we have

$$\tilde{\mathcal{L}}f_1 = -\mathcal{L}f_0 - \mathcal{N}_1(f_0,f_0)$$

$$= \left[\boldsymbol{\nabla}_\mathbf{r} + \boldsymbol{\nabla}_\mathbf{r}V_1(\mathbf{r},t) + \int \mathrm{d}\mathbf{r}'\rho_0(\mathbf{r}',t)g(\mathbf{r},\mathbf{r}')\boldsymbol{\nabla}_\mathbf{r}V_2(\mathbf{r},\mathbf{r}')\right]\rho_0(\mathbf{r},t) \cdot \mathbf{p}Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}$$

$$(3.4) \quad =: \tilde{\mathbf{a}}(\mathbf{r},t) \cdot \mathbf{p}Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}.$$

Hence, by Lemma 3.12 and the definitions $P_{1,\mathbf{e}_j} = p_j$ for $\mathbf{e}_j$ the standard unit vectors, it follows that $f_1(\mathbf{r},\mathbf{p},t) = [\mathbf{a}(\mathbf{r},t) \cdot \mathbf{p} + \psi(\mathbf{r},t)]Z^{-1}\exp(-|\mathbf{p}|^2/2)$ for some $\mathbf{a}$. Evaluating each of the terms of $\tilde{\mathcal{L}}f_1$ then gives, first from (2.6a),

$$\mathcal{L}_0 f_1 = \boldsymbol{\nabla}_\mathbf{p} \cdot \left[(\mathbf{p} + \boldsymbol{\nabla}_\mathbf{p})[\mathbf{a}(\mathbf{r},t) \cdot \mathbf{p} + \psi(\mathbf{r},t)]Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\right]$$

$$= \boldsymbol{\nabla}_\mathbf{p} \cdot \left[Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\,\boldsymbol{\nabla}_\mathbf{p}[\mathbf{a}(\mathbf{r},t) \cdot \mathbf{p} + \psi(\mathbf{r},t)]\right]$$

$$= \boldsymbol{\nabla}_\mathbf{p} \cdot \left[Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\,\mathbf{a}(\mathbf{r},t)\right] = -Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\,\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t).$$

Using the explicit form of $\mathcal{N}_0(f_0,f_1)$, as given by Lemma 3.4, gives

$$\mathcal{N}_0(f_0,f_1) = \boldsymbol{\nabla}_\mathbf{p} \cdot \left[\int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\rho_0(\mathbf{r}',t)\mathbf{Z}_1(\mathbf{r},\mathbf{r}')\right.$$

$$\left. \times (\mathbf{p} + \boldsymbol{\nabla}_\mathbf{p})\big[[\mathbf{a}(\mathbf{r},t) \cdot \mathbf{p} + \psi(\mathbf{r},t)]Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\big]\right]$$

$$= \boldsymbol{\nabla}_\mathbf{p} \cdot \left[\int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\rho_0(\mathbf{r}',t)\mathbf{Z}_1(\mathbf{r},\mathbf{r}') \times \mathbf{a}(\mathbf{r},t)Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\right].$$

For a general matrix $\mathbf{Z}(\mathbf{r})$, we have

$$\boldsymbol{\nabla}_{\mathbf{p}} \cdot \left[\mathbf{Z}(\mathbf{r})\mathbf{a}(\mathbf{r},t)Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\right] = \sum_{i,j=1}^{3} \partial_{p_i}\left[Z_{ij}(\mathbf{r},t)a_j Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\right]$$

$$= -\sum_{i,j=1}^{3} Z_{ij}(\mathbf{r},t)p_i a_j Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2} = -\mathbf{p}\cdot\mathbf{Z}(\mathbf{r},t)\mathbf{a}(\mathbf{r},t)Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2},$$

and hence

$$\mathcal{N}_0(f_0,f_1) = -Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\,\mathbf{p}\cdot\int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\rho_0(\mathbf{r}',t)\mathbf{Z}_1(\mathbf{r},\mathbf{r}')\mathbf{a}(\mathbf{r},t).$$

For the third term, we take $\mathcal{N}(f_1,f_0)$ as given by Lemma 3.4 and note that $(\mathbf{p}'+\boldsymbol{\nabla}_{\mathbf{p}'})f_1(\mathbf{r}',\mathbf{p}',t) = \mathbf{a}(\mathbf{r}',t)\,\mathrm{e}^{-|\mathbf{p}'|^2/2}$, giving

$$\mathcal{N}_0(f_1,f_0) = -Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\,\rho_0(\mathbf{r},t)\mathbf{p}\cdot\int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}'g(\mathbf{r},\mathbf{r}')\mathbf{Z}_2(\mathbf{r},\mathbf{r}')\mathbf{a}(\mathbf{r}',t)Z^{-1}\,\mathrm{e}^{-|\mathbf{p}'|^2/2}$$

$$= -Z^{-1}\,\mathrm{e}^{-|\mathbf{p}|^2/2}\,\rho_0(\mathbf{r},t)\mathbf{p}\cdot\int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\mathbf{Z}_2(\mathbf{r},\mathbf{r}')\mathbf{a}(\mathbf{r}',t).$$

Collecting the three terms gives

$$\tilde{\mathbf{a}} = -\mathbf{a}(\mathbf{r},t) - \int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\rho_0(\mathbf{r}',t)\mathbf{Z}_1(\mathbf{r},\mathbf{r}')\times\mathbf{a}(\mathbf{r},t)$$

$$- \rho_0(\mathbf{r},t)\int \mathrm{d}\mathbf{r}'g(\mathbf{r},\mathbf{r}')\mathbf{Z}_2(\mathbf{r},\mathbf{r}')\mathbf{a}(\mathbf{r}',t),$$

and the result follows by (3.4). $\quad\square$

3.3. Solution of the ϵ^0 equation. We have, by Corollary 3.3 and Lemma 3.13, that

$$f_0(\mathbf{r},\mathbf{p},t) = Z^{-1}\exp(-|\mathbf{p}|^2/2)\rho_0(\mathbf{r},t),$$
$$f_1(\mathbf{r},\mathbf{p},t) = [\mathbf{a}(\mathbf{r},t)\cdot\mathbf{p}+\psi(\mathbf{r},t)]Z^{-1}\exp(-|\mathbf{p}|^2/2).$$

We now show that the evolution equation for ρ_0 is given by the solvability condition for the equation corresponding to ϵ^0, namely (3.2c). We begin by rewriting (3.2c) as

$$(3.5) \qquad -\tilde{\mathcal{L}}f_2 = \mathcal{L}_1 f_1 + \mathcal{N}_0(f_1,f_1) + \mathcal{N}_1(f_1,f_0) + \mathcal{N}_1(f_0,f_1) - \partial_t f_0.$$

We then require that the right-hand side be orthogonal to the null space of $\tilde{\mathcal{L}}^*$ in $L^2_{f_0^{-1}}$ which, by Lemma 3.8, is equivalent to it being orthogonal to constants in $\mathbf{p}$ in the unweighted space L^2. Hence, using the divergence theorem and the explicit forms of $\mathcal{N}_0$ and $\mathcal{N}_1$ as given by (2.6c) and (2.6d), the requirement reduces to $\int \mathrm{d}\mathbf{p}(\mathcal{L}_1 f_1 - \partial_t f_0) = 0$, and we need only calculate these two terms. However, for completeness and later use, we also calculate the $\mathcal{N}_0$ and $\mathcal{N}_1$ terms.

LEMMA 3.14. *For $\mathcal{L}_1$, $\mathcal{N}_0$, and $\mathcal{N}_1$ as in* (2.6b)–(2.6d)*, f_0 as in Corollary* 3.3*, and f_1 as in Lemma* 3.13*, we have*

$$\mathcal{L}_1 f_1 = Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \Big[-\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} \mathbf{a}(\mathbf{r},t)\mathbf{p} - \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} \psi(\mathbf{r},t)$$
$$+ \boldsymbol{\nabla}_{\mathbf{r}} V_1(\mathbf{r},t) \cdot \big[\mathbf{a}(\mathbf{r},t) - \mathbf{p}\big(\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t)\big) \big] \Big],$$

$$\mathcal{N}_1(f_0, f_1) = Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \int \mathrm{d}\mathbf{r}' \rho_0(\mathbf{r}',t) g(\mathbf{r},\mathbf{r}') \boldsymbol{\nabla}_{\mathbf{r}} V_2(\mathbf{r},\mathbf{r}')$$
$$\cdot \big[\mathbf{a}(\mathbf{r},t) - \mathbf{p}(\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t)) \big],$$

$$\mathcal{N}_1(f_1, f_0) = -Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \rho_0(\mathbf{r},t)\mathbf{p} \cdot \int \mathrm{d}\mathbf{r}' \psi(\mathbf{r}',t) g(\mathbf{r},\mathbf{r}') \boldsymbol{\nabla}_{\mathbf{r}} V_2(\mathbf{r},\mathbf{r}'),$$

$$\mathcal{N}_0(f_1, f_1) = -Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \mathbf{p} \cdot \left[\int \mathrm{d}\mathbf{r}' g(\mathbf{r},\mathbf{r}') \psi(\mathbf{r}',t) \mathbf{Z}_1(\mathbf{r},\mathbf{r}') \right] \mathbf{a}(\mathbf{r},t)$$
$$+ \mathrm{e}^{-|\mathbf{p}|^2/2} \left[\int \mathrm{d}\mathbf{r}' g(\mathbf{r},\mathbf{r}') \mathbf{Z}_2(\mathbf{r},\mathbf{r}') \mathbf{a}(\mathbf{r}',t) \right]$$
$$\cdot \big[\mathbf{a}(\mathbf{r},t) - \mathbf{p}(\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t)) \big].$$

Proof. We begin with $\mathcal{L}_1 f_1$, which is given by

$$\mathcal{L}_1 f_1(\mathbf{r},\mathbf{p},t) = [-\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} + \boldsymbol{\nabla}_{\mathbf{r}} V_1(\mathbf{r},t) \cdot \boldsymbol{\nabla}_{\mathbf{p}}] \big[Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \big(\mathbf{a}(\mathbf{r},t) \cdot \mathbf{p} + \psi(\mathbf{r},t) \big) \big].$$

Simple calculations show that

$$\boldsymbol{\nabla}_{\mathbf{p}}[Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \big(\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t) \big)] = Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} [-\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}](\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t))$$
$$(3.6) \qquad\qquad = Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \big[\mathbf{a}(\mathbf{r},t) - \mathbf{p}\big(\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t)\big) \big],$$
$$-\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}}(Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \psi(\mathbf{r},t)) = -Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} \psi(\mathbf{r},t).$$

This gives three of the terms. The calculation of the fourth is easier in coordinates. Using that $\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} = \sum_{i=1}^{3} p_i \partial_{r_i}$ and $\mathbf{p} \cdot \mathbf{a} = \sum_{j=1}^{3} p_j a_j$ gives

$$-\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}}[Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \mathbf{p} \cdot \mathbf{a}(\mathbf{r},t)] = -Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}}[\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t)]$$
$$= -Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \sum_{i,j=1}^{3} p_i \partial_{r_i}[p_j a_j(\mathbf{r},t)]$$
$$= -Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} \mathbf{a}(\mathbf{r},t)\mathbf{p},$$

which gives the result for $\mathcal{L}_1 f_1$.

The expressions for $\mathcal{N}_1(f_0, f_1)$ and $\mathcal{N}_1(f_1, f_0)$ result from the trivial identities $\int \mathrm{d}\mathbf{p}' f_0(\mathbf{r}',\mathbf{p}',t) = \rho_0(\mathbf{r},t)$, $\int \mathrm{d}\mathbf{p}' f_1(\mathbf{r}',\mathbf{p}',t) = \psi(\mathbf{r},t)$, $\boldsymbol{\nabla}_{\mathbf{p}} f_0(\mathbf{r},\mathbf{p},t) = -\mathbf{p} f_0(\mathbf{r},\mathbf{p},t)$, and (3.6).

Finally, for $\mathcal{N}_0(f_1, f_1)$ we use the trivial identity $\int \mathrm{d}\mathbf{p}' f_1(\mathbf{r}',\mathbf{p}',t) = \psi(\mathbf{r},t)$, and that $(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f_1(\mathbf{r},\mathbf{p},t) = Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \boldsymbol{\nabla}_{\mathbf{p}}[\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t)] = Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \mathbf{a}(\mathbf{r},t)$, and so $\int \mathrm{d}\mathbf{p}'(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'}) f_1(\mathbf{r},\mathbf{p}',t) = \mathbf{a}(\mathbf{r}',t)$. Then

$$\mathcal{N}_0(f_1, f_1) = \boldsymbol{\nabla}_{\mathbf{p}} \cdot \left[\int \mathrm{d}\mathbf{r}' g(\mathbf{r},\mathbf{r}') \psi(\mathbf{r}',t) \mathbf{Z}_1(\mathbf{r},\mathbf{r}') \times \mathbf{a}(\mathbf{r},t) Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \right]$$
$$+ \boldsymbol{\nabla}_{\mathbf{p}} \cdot \left[\int \mathrm{d}\mathbf{r}' g(\mathbf{r},\mathbf{r}') \mathbf{Z}_2(\mathbf{r},\mathbf{r}') \mathbf{a}(\mathbf{r}',t) \times Z^{-1} \mathrm{e}^{-|\mathbf{p}|^2/2} \big(\mathbf{p} \cdot \mathbf{a}(\mathbf{r},t) + \psi(\mathbf{r},t) \big) \right],$$

both terms of which are of the form $\boldsymbol{\nabla}_{\mathbf{p}} \cdot \mathbf{v}(\mathbf{r}, t) \phi(\mathbf{r}, \mathbf{p}, t)$, where $\mathbf{v}$ is a vector. Using $\boldsymbol{\nabla}_{\mathbf{p}} \cdot \mathbf{v}(\mathbf{r}, t) \phi(\mathbf{r}, \mathbf{p}, t) = \mathbf{v}(\mathbf{r}, t) \cdot \boldsymbol{\nabla}_{\mathbf{p}} \phi(\mathbf{r}, \mathbf{p}, t)$, $\boldsymbol{\nabla}_{\mathbf{p}} e^{-|\mathbf{p}|^2/2} = -\mathbf{p} e^{-|\mathbf{p}|^2/2}$, and (3.6) completes the proof. $\quad\square$

Recall that we wish to solve (3.5) and require that $\int d\mathbf{p}(\mathcal{L}_1 f_1 - \partial_t f_0) = 0$. The identities $\int d\mathbf{p} Z^{-1} e^{-|\mathbf{p}|^2/2} = 1$, $\int d\mathbf{p} Z^{-1} e^{-|\mathbf{p}|^2/2} p_i p_j = \delta_{ij}$ show that

$$\int d\mathbf{p} Z^{-1} e^{-|\mathbf{p}|^2/2} [\mathbf{a}(\mathbf{r}, t) - \mathbf{p}(\mathbf{p} \cdot \mathbf{a})] = \mathbf{a}(\mathbf{r}, t) - \int d\mathbf{p} Z^{-1} e^{-|\mathbf{p}|^2/2} \mathbf{p} \sum_{i=1}^{3} p_j a_j(\mathbf{r}, t)$$
$$= \mathbf{a}(\mathbf{r}, t) - \mathbf{a}(\mathbf{r}, t) = 0$$

(which also follows from these terms resulting from the $\boldsymbol{\nabla}_{\mathbf{r}} V_1 \cdot \boldsymbol{\nabla}_{\mathbf{p}}$ term in $\mathcal{L}_1$ and the divergence theorem) and

$$\int d\mathbf{p} Z^{-1} e^{-|\mathbf{p}|^2/2} \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} \mathbf{a}(\mathbf{r}, t) \mathbf{p} = \int d\mathbf{p} Z^{-1} e^{-|\mathbf{p}|^2/2} \sum_{i,j=1}^{3} p_i p_j \partial_{r_j} a_j(\mathbf{r}, t) = \boldsymbol{\nabla}_{\mathbf{r}} \cdot \mathbf{a}(\mathbf{r}, t),$$

and hence $\int d\mathbf{p} \mathcal{L}_1 f_1 = -\boldsymbol{\nabla}_{\mathbf{r}} \cdot \mathbf{a}(\mathbf{r}, t)$. Since $\int d\mathbf{p} \partial_t f_0(\mathbf{r}, \mathbf{p}, t) = \partial_t \rho_0(\mathbf{r}, t)$, the solvability condition becomes

(3.7) $$\partial_t \rho_0(\mathbf{r}, t) = -\boldsymbol{\nabla}_{\mathbf{r}} \cdot \mathbf{a}(\mathbf{r}, t),$$

which is precisely the equation describing the one-body position distribution evolution for the Smoluchowski equation, as given in Theorem 4.1.

3.4. Solution of the ϵ^1 equation. We now demonstrate that $\psi(\mathbf{r}, t) \equiv 0$ if $\psi(\mathbf{r}, 0) \equiv 0$. This should result from the solvability condition for the ϵ^1 equation, which has the form

$$-\tilde{\mathcal{L}} f_3 = \mathcal{L}_1 f_2 + \mathcal{N}_0(f_2, f_1) + \mathcal{N}_0(f_1, f_2) + \mathcal{N}_1(f_2, f_0) + \mathcal{N}_1(f_0, f_2) + \mathcal{N}_1(f_1, f_1) - \partial_t f_1.$$

Since once again the $\mathcal{N}_0$ and $\mathcal{N}_1$ terms do not contribute to the Fredholm alternative calculation, we have

$$\int d\mathbf{p}(\mathcal{L}_1 f_2 - \partial_t f_1) = 0.$$

From (3.5), we have that

(3.8) $$f_2 = (-\tilde{\mathcal{L}})^{-1} [\mathcal{L}_1 f_1 + \mathcal{N}_0(f_1, f_1) + \mathcal{N}_1(f_1, f_0) + \mathcal{N}_1(f_0, f_1) - \partial_t f_0],$$

with $\partial_t f_1 = Z^{-1} e^{-|\mathbf{p}|^2/2} [\mathbf{p} \cdot \partial_t \mathbf{a}(\mathbf{r}, t) + \partial_t \psi(\mathbf{r}, t)]$ and the remaining terms given by Lemma 3.14.

For the $\mathcal{L}_1 f_2$ term, we have $\mathcal{L}_1 f_2 = [-\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} + \boldsymbol{\nabla}_{\mathbf{r}} V_1(\mathbf{r}, t) \cdot \boldsymbol{\nabla}_{\mathbf{p}}] f_2(\mathbf{r}, \mathbf{p}, t)$, and, by the divergence theorem, the second term vanishes upon integration. Hence we are interested only in

$$\int d\mathbf{p} \big[Z^{-1} e^{-|\mathbf{p}|^2/2} \partial_t \psi(\mathbf{r}, t) + \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} f_2(\mathbf{r}, \mathbf{p}, t) \big] = \partial_t \psi(\mathbf{r}, t) + \int d\mathbf{p} \, \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} f_2(\mathbf{r}, \mathbf{p}, t).$$

Since $H_{1, \mathbf{e}_j}(p) = p_j$, the only terms from f_2 which contribute to the integral are of the form $\mathbf{p} \cdot \mathbf{a}_2(\mathbf{r}, t) Z^{-1} e^{-|\mathbf{p}|^2/2}$, i.e., $\mathbf{p} \cdot \mathcal{P}_1(f_2)$, where $\mathcal{P}_1$ is the projection onto $\mathbf{p}$,

i.e., $\mathcal{P}_1 f = \int d\mathbf{p}\,\mathbf{p} f(\mathbf{r}, \mathbf{p}, t)$. By (3.8) and Lemma 3.12, it therefore suffices to consider only terms of the form $-\mathbf{p} \cdot \tilde{\mathbf{a}}_2(\mathbf{r}, t) Z^{-1} e^{-|\mathbf{p}|^2/2}$ in $\mathcal{L}_1 f_1 + \mathcal{N}_0(f_1, f_1) + \mathcal{N}_1(f_1, f_0) + \mathcal{N}_1(f_0, f_1) - \partial_t f_0$, i.e., $\tilde{\mathbf{a}}_2(\mathbf{r}, t) = -\mathcal{P}_1\big(\mathcal{L}_1 f_1 + \mathcal{N}_0(f_1, f_1) + \mathcal{N}_1(f_1, f_0) + \mathcal{N}_1(f_0, f_1) - \partial_t f_0\big)$.

By Lemma 3.14 and $\partial_t f_0(\mathbf{r}, \mathbf{p}, t) = Z^{-1} e^{-|\mathbf{p}|^2/2} \partial_t \rho_0(\mathbf{r}, t)$, we have

$$\mathcal{P}_1(-\tilde{\mathcal{L}} f_2) = \mathcal{P}_1\big(\mathcal{L}_1 f_1 + \mathcal{N}_0(f_1, f_1) + \mathcal{N}_1(f_1, f_0) + \mathcal{N}_1(f_0, f_1) - \partial_t f_0\big) =: -\tilde{\mathbf{a}}_2(\mathbf{r}, t),$$

with

$$
\begin{aligned}
\tilde{\mathbf{a}}_2(\mathbf{r}, t) := \bigg[& \boldsymbol{\nabla}_{\mathbf{r}} \psi(\mathbf{r}, t) + \psi(\mathbf{r}, t) \boldsymbol{\nabla}_{\mathbf{r}} V_1(\mathbf{r}, t) + \int d\mathbf{r}' \rho_0(\mathbf{r}, t) g(\mathbf{r}, \mathbf{r}') \boldsymbol{\nabla}_{\mathbf{r}} V_2(\mathbf{r}, \mathbf{r}') \psi(\mathbf{r}, t) \\
& + \rho_0(\mathbf{r}, t) \bigg(\int d\mathbf{r}' \psi(\mathbf{r}', t) g(\mathbf{r}, \mathbf{r}') \boldsymbol{\nabla}_{\mathbf{r}} V_2(\mathbf{r}, \mathbf{r}') \bigg) \\
& + \bigg(\int d\mathbf{r}' \psi(\mathbf{r}', t) g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \bigg) \mathbf{a}(\mathbf{r}, t) \\
& + \bigg(\int d\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}') \mathbf{a}(\mathbf{r}', t) \bigg) \psi(\mathbf{r}, t) \bigg],
\end{aligned}
$$

where $\mathbf{a}(\mathbf{r}, t)$ is given by Lemma 3.13. Thus, by (the proof of) Lemma 3.13,

$$\mathcal{P}_1 f_2(\mathbf{r}, \mathbf{p}, t) = \mathbf{a}_2(\mathbf{r}, t),$$

with $\mathbf{a}_2$ the solution of

$$
\begin{aligned}
-\tilde{\mathbf{a}}_2(\mathbf{r}, t) = \mathbf{a}_2(\mathbf{r}, t) + \int d\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \rho_0(\mathbf{r}', t) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \times \mathbf{a}_2(\mathbf{r}, t) \\
+ \rho_0(\mathbf{r}, t) \int d\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}') \mathbf{a}_2(\mathbf{r}', t).
\end{aligned}
$$

Hence, for the ϵ^1 equation to be solvable,

$$
\begin{aligned}
0 &= \partial_t \psi(\mathbf{r}, t) + \int d\mathbf{p}\,\mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} \mathcal{P}_1 f_2(\mathbf{r}, \mathbf{p}, t) \\
&= \partial_t \psi(\mathbf{r}, t) + \int d\mathbf{p} Z^{-1} e^{-|\mathbf{p}|^2/2} \mathbf{p} \cdot \boldsymbol{\nabla}_{\mathbf{r}} [\mathbf{p} \cdot \mathbf{a}_2(\mathbf{r}, t)] = \partial_t \psi(\mathbf{r}, t) + \boldsymbol{\nabla}_{\mathbf{r}} \cdot \mathbf{a}_2(\mathbf{r}, t)
\end{aligned}
$$

or

(3.9)
$$\partial_t \psi(\mathbf{r}, t) = -\boldsymbol{\nabla}_{\mathbf{r}} \cdot \mathbf{a}_2(\mathbf{r}, t).$$

To ensure that $\psi(\mathbf{r}, t) \equiv 0$, we first note that $\psi(\mathbf{r}, 0) \equiv 0$ is equivalent to assuming that the initial condition $f^{(1)}(\mathbf{r}, \mathbf{p}, 0)$ is independent of ϵ. For this to hold for all t, it is necessary to show that (3.9) is dissipative (or, since $\partial_t \int d\mathbf{r} \psi(\mathbf{r}, t) = 0$, that (3.9) is nonnegativity preserving).

The proof in the linear (V_2, $\mathbf{Z}_1$, $\mathbf{Z}_2$ all zero) case is trivial, as it turns out that ψ and ρ satisfy the same equation. Thus, since the Smoluchowski equation for ρ must be nonnegativity preserving, so must the equation for ψ. The proof in the general case is complicated both by the equations for ρ and ψ not being identical (due to the nonlinear terms) and by needing to prove dissipativity results for the resulting nonlinear operators. In general (for $\mathbf{Z}_2 \neq 0$), the equations are not even explicit, as one needs to solve the Fredholm integral equations for $\mathbf{a}$ and $\mathbf{a}_2$. However, since the

full friction tensor is positive definite, one would expect (3.9) to be a parabolic PDE, and so, for potentials and hydrodynamic interaction terms with sufficient bounded derivatives, the result should follow from standard PDE theory; see, e.g., [74]. We therefore assume that V_1, V_2, $\mathbf{Z}_1$, and $\mathbf{Z}_2$ are such that if $\psi(\mathbf{r}, 0) \equiv 0$, then $\psi(\mathbf{r}, t) \equiv 0$ for all $t \geq 0$.

4. The Smoluchowski equation. We are now in a position to state our rigorously derived Smoluchowski equation. For ease of comparison with existing results, we return to the original scalings of time and potentials.

THEOREM 4.1 (Smoluchowski equation). *Suppose* $f^{(1)}(\mathbf{r}, \mathbf{p}, 0) = f_0(\mathbf{r}, \mathbf{p}, 0) = Z^{-1} e^{-|\mathbf{p}|^2/2} \rho_0(\mathbf{r}, 0)$ *is independent of* ϵ. *Suppose further that* $\rho_0(\mathbf{r}, 0)$, U_j, *and* $\mathbf{Z}_j$, $j = 1, 2$, *are such that the solutions of* (2.5), (3.7), *and* (3.9) *exist for times* $[0, t_0]$ *and that* (3.9) *is nonnegativity preserving. Then, up to errors of* $\mathcal{O}(\epsilon^2)$, *the dynamics of the one-body position distribution are given (in the original timescale) for* $\tau \in [0, m\gamma/(k_B T)t_0]$ *by*

$$\partial_\tau \rho(\mathbf{r}, \tau) = -\frac{k_B T}{m\gamma} \boldsymbol{\nabla}_\mathbf{r} \cdot \mathbf{a}(\mathbf{r}, \tau),$$

where $\mathbf{a}(\mathbf{r}, \tau)$ *is the solution to*

$$\mathbf{a}(\mathbf{r}, \tau) + \int d\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \rho(\mathbf{r}', \tau) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \times \mathbf{a}(\mathbf{r}, \tau) + \rho(\mathbf{r}, \tau) \int d\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}') \mathbf{a}(\mathbf{r}', \tau)$$

$$(4.1) \qquad = -\left[\boldsymbol{\nabla}_\mathbf{r} + \frac{1}{k_B T} \left(\boldsymbol{\nabla}_\mathbf{r} U_1(\mathbf{r}, \tau) + \int d\mathbf{r}' \rho(\mathbf{r}', \tau) g(\mathbf{r}, \mathbf{r}') \boldsymbol{\nabla}_\mathbf{r} U_2(\mathbf{r}, \mathbf{r}') \right) \right] \rho(\mathbf{r}, \tau).$$

Proof. The evolution equation is given by (3.7), which is the solvability condition for (3.2c), and $\mathbf{a}(\mathbf{r}, t)$ is given by Lemma 3.13. Returning to the original timescale introduces the factor of $\mu = k_B T/(m\gamma)$ in the right-hand side. We also replace V by its original value of $U/(k_B T)$, where $\mathbf{X}_i = -\boldsymbol{\nabla}_{\mathbf{r}_i} U(\mathbf{r}^N)$. The conditions on (3.9) and the initial condition ensure that, using the notation of Lemma 3.13, $\psi(\mathbf{r}, t) \equiv 0$ for all times. Hence $\rho(\mathbf{r}, \tau) = \rho_0(\mathbf{r}, \tau) + \mathcal{O}(\epsilon^2)$. $\quad\square$

We note here that the assumptions on the initial condition and on the existence of solutions are analogous to those made for the Boltzmann equation; see, e.g., [27]. We expect that proving that such assumptions hold for a physically interesting range of potentials and friction tensors would be a formidable problem in its own right, but this is beyond the scope of the present study. An analysis of the corresponding problem for the Boltzmann equation is given in [22].

To demonstrate the connection to existing formulations, we assume that $\mathbf{Z}_2 \equiv 0$, which allows us to find $\mathbf{a}$ explicitly. We then have the following corollary.

COROLLARY 4.2. *Under the same assumptions as in Theorem 4.1, if* $\mathbf{Z}_2 \equiv 0$, *the one-body position dynamics are, up to errors of* $\mathcal{O}(\epsilon^2)$, *governed by*

$$\partial_\tau \rho(\mathbf{r}, \tau) = \boldsymbol{\nabla}_\mathbf{r} \cdot \left(\mathbf{D}(\mathbf{r}, \tau) \left[\boldsymbol{\nabla}_\mathbf{r} \rho(\mathbf{r}, \tau) + \frac{1}{k_B T} \rho(\mathbf{r}, \tau) \boldsymbol{\nabla}_\mathbf{r} V_1(\mathbf{r}, \tau) \right.\right.$$

$$(4.2) \qquad \qquad \left.\left. + \frac{1}{k_B T} \int d\mathbf{r}' \rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) \boldsymbol{\nabla}_\mathbf{r} V_2(\mathbf{r}, \mathbf{r}') \right] \right),$$

where we have defined $\rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) := \rho(\mathbf{r}, \tau) \rho(\mathbf{r}', \tau) g(\mathbf{r}, \mathbf{r}', \tau)$, *as it would be for the Enskog approximation, and the* 3×3 *diffusion tensor* $\mathbf{D}$ *is given by*

$$\mathbf{D}(\mathbf{r}, \tau) = \frac{k_B T}{m\gamma} \left[\mathbf{1} + \int d\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \rho(\mathbf{r}', \tau) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \right]^{-1}.$$

Corollary 4.2 gives a one-body Smoluchowski equation with a novel form for the diffusion tensor $\mathbf{D}$. As is clear from the notation, $\mathbf{D}(\mathbf{r}, \tau)$ depends not only on the position but also on the time. This time dependence is present through the time dependence of ρ, against which the two-body terms must be averaged.

One obvious question is whether $\mathbf{D}$ is positive definite. A simple calculation shows that this is indeed the case. Note that $\mathbf{1} + \sum_{j \neq 1} \mathbf{Z}_1(\mathbf{r}_1, \mathbf{r}_j)$ is positive definite (since it is a principal minor of $\mathbf{\Gamma}$, which is positive definite). Hence, for any $\mathbf{v}(\mathbf{r}, \tau)$, we have, for some $\delta > 0$,

$$\mathbf{v}(\mathbf{r}_1, \tau) \cdot \left[\mathbf{1} + \sum_{j \neq 1} \mathbf{Z}_1(\mathbf{r}_1, \mathbf{r}_j) \right] \mathbf{v}(\mathbf{r}_1, \tau) \geq \delta |\mathbf{v}(\mathbf{r}_1, \tau)|^2$$

$$\Rightarrow \mathbf{v}(\mathbf{r}_1, \tau) \cdot \int d\mathbf{r}_2 \rho(\mathbf{r}_2, \tau) g(\mathbf{r}_1, \mathbf{r}_2) [\tfrac{1}{N-1} \mathbf{1} + \mathbf{Z}_1(\mathbf{r}_1, \mathbf{r}_2)] \mathbf{v}(\mathbf{r}_1, \tau) \geq \delta |\mathbf{v}(\mathbf{r}_1, \tau)|^2,$$

where the proof is virtually identical to that of Lemma 3.1, except we do not integrate over $\mathbf{r}_1$. Since $\int d\mathbf{r}_2 \rho(\mathbf{r}_2, \tau) g(\mathbf{r}, \mathbf{r}_2) = N - 1$, and $k_B T$, m, and γ are positive, this is equivalent to $\mathbf{D}^{-1}$, and hence $\mathbf{D}$, being positive definite.

We now compare our result with that derived by Rex and Löwen [56, equations (5)–(8)]. As demonstrated in Figure 1.1, their Smoluchowski equation is derived from the N-body Smoluchowski equation for pairwise additivity of both the potential (our Assumption 1) and the diffusion tensor. The second assumption is analogous to our Assumption 2, but not equivalent, as the inverse of a matrix (recall $\mathbf{\Gamma D} = k_B T/m\mathbf{1}$) with pairwise terms need not contain only pairwise terms. However, there are situations where the two assumptions are essentially equivalent, such as in a diffuse colloid system. The underlying assumption then is that there exists an additional small parameter, say λ, with $1 \gg \lambda \gg \epsilon$ and such that $\mathbf{Z}_1 = \mathcal{O}(\lambda)$. Then, up to errors of $\mathcal{O}(\lambda^2)$, $\mathbf{D}(\mathbf{r}, \tau) = \frac{k_B T}{m\gamma}[\mathbf{1} - \int d\mathbf{r}' g(\mathbf{r}, \mathbf{r}') \rho(\mathbf{r}', \tau) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}')]$, and thus the diffusion tensor is a two-body one. We note that the analogue of Assumption 3 is $\rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2, t) = \rho(\mathbf{r}_1, t) \rho(\mathbf{r}_2, t) g(\mathbf{r}_1, \mathbf{r}_2)$, which can be seen by integrating out the momentum dependence.

The simplest case is that in which both $\mathbf{\Gamma} := \gamma \mathbf{1}$ and $\mathbf{D} := D_0 \mathbf{1}$ are proportional to the identity matrix, when we have the standard definition $D_0 = k_B T/(m\gamma)$. In this case it is easy to check that the two formulations agree (see also [3]). This is unsurprising, as both the difficulties and interest in this analysis lie with the nonuniform terms in the friction tensor.

To demonstrate that the two formulations differ in general, we consider the simple example used in Corollary 4.2. In addition, we assume the existence of a parameter λ, as described above. Then, by the block diagonal form of $\mathbf{\Gamma}$, $\mathbf{D}$ is also block diagonal with blocks $D_0(\mathbf{1} - \sum_{\ell \neq i} \mathbf{Z}_1(\mathbf{r}_i, \mathbf{r}_\ell))$; i.e., in the notation of [56], $\mathbf{w}_{11} = -\mathbf{Z}_1$. The result to compare with (4.2) is (see [56])

$$\partial_\tau \rho(\mathbf{r}, \tau) = D_0 \mathbf{\nabla_r} \cdot \left[\mathbf{\nabla_r} \rho(\mathbf{r}, \tau) + \tfrac{1}{k_B T} \rho(\mathbf{r}, \tau) \mathbf{\nabla_r} V_1(\mathbf{r}, \tau) \right.$$

$$+ \tfrac{1}{k_B T} \int d\mathbf{r}' \rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) \mathbf{\nabla_r} V_2(\mathbf{r}, \mathbf{r}')$$

$$- \int d\mathbf{r}' \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \left(\mathbf{\nabla_r} \rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) + \tfrac{1}{k_B T} \mathbf{\nabla_r}[V_1(\mathbf{r}, \tau) + V_2(\mathbf{r}, \mathbf{r}')] \rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) \right.$$

$$\left. \left. + \tfrac{1}{k_B T} \int d\mathbf{r}'' \rho^{(3)}(\mathbf{r}, \mathbf{r}', \mathbf{r}'', \tau) \mathbf{\nabla_r} V_2(\mathbf{r}, \mathbf{r}'') \right) \right].$$

Using the approximate two-body form of $\mathbf{D}$ in (4.2) gives

$$
\begin{aligned}
\partial_\tau \rho(\mathbf{r}, \tau) = D_0 \boldsymbol{\nabla}_\mathbf{r} \cdot \Bigg[&\boldsymbol{\nabla}_\mathbf{r} \rho(\mathbf{r}, \tau) + \tfrac{1}{k_B T} \rho(\mathbf{r}, \tau) \boldsymbol{\nabla}_\mathbf{r} V_1(\mathbf{r}, \tau) \\
&+ \tfrac{1}{k_B T} \int \mathrm{d}\mathbf{r}' \rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) \boldsymbol{\nabla}_\mathbf{r} V_2(\mathbf{r}, \mathbf{r}') \\
&- \int \mathrm{d}\mathbf{r}' \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \Big(\rho(\mathbf{r}', \tau) g(\mathbf{r}, \mathbf{r}') \boldsymbol{\nabla}_\mathbf{r} \rho(\mathbf{r}, \tau) + \tfrac{1}{k_B T} \rho^{(2)}(\mathbf{r}, \mathbf{r}', \tau) \boldsymbol{\nabla}_\mathbf{r} V_1(\mathbf{r}, \tau) \\
&\qquad\qquad + \tfrac{1}{k_B T} \rho(\mathbf{r}', t) g(\mathbf{r}, \mathbf{r}') \int \mathrm{d}\mathbf{r}'' \rho^{(2)}(\mathbf{r}, \mathbf{r}'', \tau) \boldsymbol{\nabla}_\mathbf{r} V_2(\mathbf{r}, \mathbf{r}'') \Big) \Bigg],
\end{aligned}
$$

and it is clear that the two formulations are not, in general, equivalent. See Figure 1.1 for a diagrammatic representation of the difference in the formalisms.

Interestingly, despite their obvious differences, in the overdamped limit both formulations are accurate to $\mathcal{O}(\epsilon^2)$. There does not seem to be any mathematical or physical justification to say that one of them is "more correct" than the other. However, these differences make it clear that the two processes, (i) adiabatically eliminating the fast momentum variable and (ii) integrating over all but one particle's coordinates, do not commute. It is worth noting that the need for knowledge of $\rho^{(3)}$ in the first case stems from the explicit coupling of the two-body diffusion tensor and potential in the N-body Smoluchowski equation. In contrast, the potential and friction tensor are not explicitly coupled in the N-body Kramers equation, and thus only $\rho^{(2)}$ is required. This is a partial explanation of why the resulting equations must be different.

It would be interesting to perform numerical studies to see if one can quantify the differences, i.e., if one can determine the magnitude of the difference in the $\mathcal{O}(\epsilon^2)$ terms. The first form above has been implemented numerically as a DDFT by making the further approximation that the term involving the many-body potential is given by its value in an equilibrium system with the same one-body density [56]. This introduces additional, uncontrolled errors, and, as such, a direct comparison with the new formulation presented here, which requires no further approximations, would likely be uninformative. For further numerical studies, including comparison with the full underlying stochastic dynamics, demonstrations of the large qualitative and quantitative effects of hydrodynamic interactions, and a novel DDFT including inertial effects, see [31].

We close by stating a result which is most useful when reducing from a phase-space DDFT to one only in position space.

COROLLARY 4.3. *Terms proportional to Hermite polynomials of order* 2 *and higher in* $\mathbf{p}$ *enter* $f(\mathbf{r}, \mathbf{p}, t)$ *at most with order* ϵ^2.

5. Conclusions and open problems. Our main result is that, for suitable two-body potentials and friction tensors, and using the Enskog approximation in the limit of small ϵ, the leading-order solution to (2.5) is given by Theorem 4.1. This is a novel Smoluchowski-type equation with a new definition of the one-body diffusion tensor. In addition, the Hilbert expansion studied in section 3 allows us to show rigorously that a term typically neglected by heuristic arguments in the derivation of DDFT [2, 3] is indeed negligible in the overdamped limit; see Corollary 4.3. However, these results have only been shown to hold when the initial condition is independent of ϵ, along with assuming that g is independent of ϵ and $\mathbf{p}$. We now discuss how removing these assumptions should be tackled.

The assumption that the initial condition is independent of ϵ was made for con-

venience, as it allows for analytical progress. In general, however, the question of how the initial condition for Kramers equation is related to the correct corresponding initial condition for the Smoluchowski equation needs to be addressed. As with the Boltzmann equation (cf. [27]) and also noted in the discussion following (3.1), we would expect a boundary layer in time (much shorter than the macroscopic timescale discussed in this work), over which the given initial condition is attracted to one with Gaussian momentum distribution. However, this introduces additional complications that can be studied by modifying the Hilbert expansion appropriately, introducing terms that account for the boundary layer and decay exponentially in time; see, e.g., [6]. We note that even if the nonnegativity-preserving assumption of Theorem 4.1 did not hold, then the evolution equations given would be accurate to $\mathcal{O}(\epsilon)$. Whilst it would be ideal to have a proof that the evolutions in section 3.4 preserve the non-negativity of ψ, as mentioned previously, this leads to significant additional technical difficulties and may well require further assumptions on the potentials and hydrodynamic interactions.

In addition, as mentioned in section 1, according to DFT, g is also a functional of ρ. If ρ is independent of ϵ (i.e., depends only on f_0), then the analysis is unaffected. This is the case if the initial condition is independent of ϵ and the terms ψ_i (the part of f_i in the null space of $\tilde{\mathcal{L}}$) are uniformly zero for all times and all i. As mentioned earlier, such a result would rely on the dissipativity of the determining equations or, equivalently, on the equations being nonnegativity preserving.

If $\rho(\mathbf{r}, t)$ depends on ϵ, then the nonlinear operators are no longer quadratic in f, as ρ depends on higher-order, ϵ-dependent parts of f. Furthermore, we do not know the precise dependence of g on ρ. However, if we expand g as a power series in ϵ, $g = g_0 + \epsilon g_1 + \cdots$, then the equations in ϵ^{-2} and ϵ^{-1} change only by replacing g with g_0. This is because there are no extra terms in the ϵ^{-2} equation, and only the extra term $\mathcal{N}(f_0, f_0, g_1) = 0$ enters the ϵ^{-1} equation. The main point is that g_1 enters only through nonlinear terms, namely, by the addition of the terms $\mathcal{N}_0(f_1, f_0, g_1)$, $\mathcal{N}_0(f_0, f_1, g_1)$, and $\mathcal{N}_1(f_0, f_0, g_1)$ to the right-hand side of (3.5) (where we have now shown the explicit dependence of the nonlinear terms on the g_j). Thus the conclusion that the dynamics of f_0 are governed by the solvability condition $\int d\mathbf{p}(\mathcal{L}_1 f_1 - \partial_t f_0) = 0$ still holds.

The first difference comes when determining f_2, or, more precisely, $P_1 f_2$, which gains additional g_1-dependent terms. The evolution equation for ψ looks superficially similar, but it results in a new definition of $\tilde{\mathbf{a}}_2$ and hence also of $\mathbf{a}_2$.

We note here that a similar argument would apply if g were chosen to depend explicitly on time. In particular, there are no further difficulties if g depends only on the slowest timescale, i.e., if it is independent of ϵ. However, how one would choose this explicit dependence is unclear. The standard approach is to choose g either to be a functional of a suitably averaged distribution $\bar{\rho}(t)$ or to satisfy the generalized Ornstein–Zernike equation [56]. In both cases, the time dependence of g is due only to the time dependence of ρ and is not prescribed explicitly.

Allowing g to depend (symmetrically) on $\mathbf{p}_1$ and $\mathbf{p}_2$ introduces many additional complications in the analysis. If the $\mathbf{p}$-dependence is introduced at leading order, it significantly changes the analysis of the nonlinear terms. Whilst, by (2.2), it still holds that $\int d\mathbf{r}' d\mathbf{p}' f^{(1)}(\mathbf{r}, \mathbf{p}, \tau) g(\mathbf{r}, \mathbf{r}', \mathbf{p}, \mathbf{p}') = N - 1$, we actually require an expression for $\int d\mathbf{p}' f^{(1)}(\mathbf{r}, \mathbf{p}, \tau) g(\mathbf{r}, \mathbf{r}', \mathbf{p}, \mathbf{p}')$. For example, the $\mathcal{N}_1(f_0, f_0)$ term in Lemma 3.9 is significantly more complicated.

In this work we have solved the dynamics of the one-body distribution up to errors of $\mathcal{O}(\epsilon^2)$. However, an interesting question is whether the true solution and

the solution given by Theorem 4.1 are also close in some suitable norm. Ideally, one would like to prove a result analogous to Theorem 3.1 of [27], which states that, under suitable assumptions, in a suitable norm, and for a fixed macroscopic time period $(0, t_0]$, the solution to the Boltzmann equation is $\mathcal{O}(\epsilon)$ close to the local Maxwellian whose parameters vary according to the hydrodynamic equations. Note that in the case where $t_0 \to \infty$, the constant in the $\mathcal{O}(\epsilon)$ bound may diverge.

In order to prove such a result, one must truncate the Hilbert series at a finite order and add a remainder term. One then determines bounds on each of these terms, which require sufficiently good estimates on the collision term (the hydrodynamic interactions and V_2 in our case). This truncation is necessary, as the Hilbert expansion does not converge uniformly in the small parameter. Since such estimates on the collision operator depend on its precise form (in particular, it is assumed that the kernel of the Boltzmann collision operator has finite range; this is not true for hydrodynamic interactions) and a specific choice of norms, we have restricted our analysis to determining the leading-order terms in such an expansion.

We close by discussing some open problems. The first area concerns confined fluids and the effects of boundaries. Although the external potential V_1 may be used to model boundaries which are impermeable to the colloid particles but permeable to the fluid, a truly confined fluid cannot be modeled in this way. Extension to such systems would require a treatment of the hydrodynamic interactions caused by the boundaries. Such effects break the symmetry of the bath as well as change the mobility of the colloid particles near the boundaries. Additional complications would result from the presence of heterogeneities at boundaries, which is indeed the case in practice. Heterogeneous boundaries, either chemical or topographical, can have a significant effect on the behavior of fluids at both the microscale (e.g., they can influence the thickness of the wetting layer in the immediate vicinity of the boundary and corresponding wetting transitions) and the macroscale (they can affect the shape of the gas-liquid interface away from the boundaries) [68, 53, 9, 65, 66, 48]. It would also be of interest to study mixtures of colloid particles, e.g., a system with two types of particles which differ in their sizes, masses, or interparticle potentials V_2. As mentioned above, a full treatment of the problem would involve analysis of boundary layer effects, including how the initial condition for the Smoluchowski equation should be determined by that for the Kramers equation. These and related issues are currently under investigation.

Appendix A. Proofs of some lemmas of section 3.

A.1. Proof of Lemma 3.1. Let $\mathbf{w} = \big(\mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t), \ldots, \mathbf{v}(\mathbf{r}_N, \mathbf{p}_N, t)\big)^T$. Since $\boldsymbol{\Gamma}(\mathbf{r}^N)$ is positive definite, we have, for some $\delta > 0$, $\mathbf{w} \cdot \boldsymbol{\Gamma}\mathbf{w} \geq \delta \mathbf{w} \cdot \mathbf{w}$. Hence

$$
\begin{aligned}
\mathbf{w} \cdot \boldsymbol{\Gamma}\mathbf{w} &= \sum_{i,j=1}^{N} \mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t) \cdot \boldsymbol{\Gamma}_{ij}(\mathbf{r}^N)\mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t) \\
&= \sum_{i=1}^{N} \mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t) \cdot \left[\mathbf{1} + \sum_{j \neq i} \mathbf{Z}_1(\mathbf{r}_i, \mathbf{r}_j)\right]\mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t) \\
&\quad + \sum_{i \neq j} \mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t) \cdot \mathbf{Z}_2(\mathbf{r}_i, \mathbf{r}_j)\mathbf{v}(\mathbf{r}_j, \mathbf{p}_j, t) \geq \delta \sum_{i=1}^{N} |\mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t)|^2.
\end{aligned}
$$

Since $f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t)$ is nonnegative, and is positive on a set of nonzero measure (as by definition $f^{(N)} \geq 0$ and $\int \mathrm{d}\mathbf{r}^N \mathrm{d}\mathbf{p}^N f^{(N)} = N$), we have

$$\int \mathrm{d}\mathbf{r}^N \mathrm{d}\mathbf{p}^N f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t) \Bigg(\sum_{i=1}^{N} \mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t) \cdot \Big[\mathbf{1} + \sum_{j \neq i} \mathbf{Z}_1(\mathbf{r}_i, \mathbf{r}_j) \Big] \mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t)$$

$$+ \sum_{i \neq j} \mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t) \cdot \mathbf{Z}_2(\mathbf{r}_i, \mathbf{r}_j) \mathbf{v}(\mathbf{r}_j, \mathbf{p}_j, t) \Bigg)$$

$$\geq \delta \int \mathrm{d}\mathbf{r}^N \mathrm{d}\mathbf{p}^N f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t) \sum_{i=1}^{N} |\mathbf{v}(\mathbf{r}_i, \mathbf{p}_i, t)|^2.$$

By the symmetry of $f^{(N)}$, interchanging dummy variables of integration gives

$$\int \mathrm{d}\mathbf{r}^N \mathrm{d}\mathbf{p}^N f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t) \Big(N \mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t) \cdot \Big[\mathbf{1} + (N-1) \mathbf{Z}_1(\mathbf{r}_1, \mathbf{r}_2) \Big] \mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t)$$

$$+ N(N-1) \mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t) \cdot \mathbf{Z}_2(\mathbf{r}_1, \mathbf{r}_2) \mathbf{v}(\mathbf{r}_2, \mathbf{p}_2, t) \Big)$$

$$\geq \delta N \int \mathrm{d}\mathbf{r}^N \mathrm{d}\mathbf{p}^N f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t) |\mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t)|^2.$$

Using (2.2) for the cases with $n = 1$ and $n = 2$, i.e.,

$$f^{(2)}(\mathbf{r}_1, \mathbf{p}_1, \mathbf{r}_2, \mathbf{p}_2, t) = N(N-1) \int \mathrm{d}\mathbf{r}^{N-2} \mathrm{d}\mathbf{p}^{N-2} f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t) \text{ and}$$

$$f^{(1)}(\mathbf{r}_1, \mathbf{p}_1, t) = N \int \mathrm{d}\mathbf{r}^{N-1} \mathrm{d}\mathbf{p}^{N-1} f^{(N)}(\mathbf{r}^N, \mathbf{p}^N, t),$$

gives

$$\int \mathrm{d}\mathbf{r}_1 \mathrm{d}\mathbf{p}_1 \mathrm{d}\mathbf{r}_2 \mathrm{d}\mathbf{p}_2 f^{(2)}(\mathbf{r}_1, \mathbf{p}_1, \mathbf{r}_2, \mathbf{p}_2, t) \Big(\mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t) \cdot \Big[\tfrac{1}{N-1} \mathbf{1} + \mathbf{Z}_1(\mathbf{r}_1, \mathbf{r}_2) \Big] \mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t)$$

$$+ \mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t) \cdot \mathbf{Z}_2(\mathbf{r}_1, \mathbf{r}_2) \mathbf{v}(\mathbf{r}_2, \mathbf{p}_2, t) \Big)$$

$$\geq \delta \int \mathrm{d}\mathbf{r}_1 \mathrm{d}\mathbf{p}_1 f(\mathbf{r}_1, \mathbf{p}_1, t) |\mathbf{v}(\mathbf{r}_1, \mathbf{p}_1, t)|^2.$$

Inserting the definition $f^{(2)}(\mathbf{r}_1, \mathbf{p}_1, \mathbf{r}_2, \mathbf{p}_2, t) = g(\mathbf{r}_1, \mathbf{r}_2) f(\mathbf{r}_1, \mathbf{p}_1, t) f(\mathbf{r}_2, \mathbf{p}_2, t)$ and renaming the dummy variables gives the result.

The fact that f may be chosen as $f^{(1)}$ is trivial. To see that the result holds when f is replaced by f_0, we insert the expansion (3.1) and note $\int \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f^{(1)}(\mathbf{r}', \mathbf{p}', t) = N - 1$ holds for all ϵ, in particular for $\epsilon = 0$. $\quad\square$

A.2. Proof of Lemma 3.6. We consider each of the three operators in $\tilde{\mathcal{L}}$ individually, starting with $\mathcal{L}_0$. For arbitrary f, $\tilde{f}$, and using Corollary 3.3, in particular that $\boldsymbol{\nabla}_{\mathbf{p}}(f_0^{-1}) = \mathbf{p} f_0^{-1}$,

$$\langle f, \mathcal{L}_0 \tilde{f} \rangle_{f_0^{-1}} = \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) f(\mathbf{r}, \mathbf{p}, t) \boldsymbol{\nabla}_{\mathbf{p}} \cdot [(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \tilde{f}(\mathbf{r}, \mathbf{p}, t)]$$

$$= - \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t) \cdot [(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \tilde{f}(\mathbf{r}, \mathbf{p}, t)]$$

$$= \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) \tilde{f}(\mathbf{r}, \mathbf{p}, t) \boldsymbol{\nabla}_{\mathbf{p}} \cdot [(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)],$$

where the second and third lines both follow via integration by parts. Hence $\mathcal{L}_0$ is self-adjoint.

For $\mathcal{N}_0(f_0, \tilde{f})$, by Lemma 3.4 we have

$$\langle f, \mathcal{N}_0(f_0, \tilde{f})\rangle_{f_0^{-1}} = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) f(\mathbf{r}, \mathbf{p}, t)$$
$$\times \boldsymbol{\nabla}_{\mathbf{p}} \cdot \left[\int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \times (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}})\tilde{f}(\mathbf{r}, \mathbf{p}, t)\right]$$
$$= -\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t)$$
$$\cdot \left[\int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t) \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \times (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}})\tilde{f}(\mathbf{r}, \mathbf{p}, t)\right]$$
$$= -\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}\mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' f_0^{-1}(\mathbf{r}, \mathbf{p}, t) g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t)$$
$$\times (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t) \cdot \mathbf{Z}_1(\mathbf{r}, \mathbf{r}')(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}})\tilde{f}(\mathbf{r}, \mathbf{p}, t),$$

where we have used integration by parts and Fubini's theorem. Now, since $\mathbf{Z}_1$ is a symmetric matrix, f and $\tilde{f}$ can be interchanged and the argument reversed, showing that $\mathcal{N}_0(f_0, \tilde{f})$ is self-adjoint.

It remains to calculate the adjoint of $\mathcal{N}_0(\tilde{f}, f_0)$. Using Lemma 3.4 gives

$$\langle f, \mathcal{N}_0(\tilde{f}, f_0)\rangle_{f_0^{-1}} = -\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r}, \mathbf{p}, t) f(\mathbf{r}, \mathbf{p}, t) f_0(\mathbf{r}, \mathbf{p}, t)$$
$$\times \int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'})\tilde{f}(\mathbf{r}', \mathbf{p}', t) \cdot \mathbf{p}$$
$$= -\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p}\mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f(\mathbf{r}, \mathbf{p}, t)\tilde{f}(\mathbf{r}', \mathbf{p}', t)\mathbf{p} \cdot \mathbf{Z}_2(\mathbf{r}, \mathbf{r}')\mathbf{p}',$$

where we have used the divergence theorem, Fubini's theorem, and the identity (for symmetric matrices) $\mathbf{Z}_2\mathbf{p}' \cdot \mathbf{p} = \mathbf{p} \cdot \mathbf{Z}_2\mathbf{p}'$. Since this final term, along with the rest of the integral, is symmetric under interchanging the pairs of dummy variables $(\mathbf{r}, \mathbf{p}) \leftrightarrow (\mathbf{r}', \mathbf{p}')$, we see that $\mathcal{N}_0(\tilde{f}, f_0)$ is also self-adjoint. The overall result now follows from linearity of the integral and hence of the adjoint. $\quad\square$

A.3. Proof of Lemma 3.7. We prove the equivalent statement (for self-adjoint operators, as $\tilde{\mathcal{L}}$ is by Lemma 3.6) that there exists an orthonormal basis $(\xi_j)_{j=1}^\infty$ of $L^2_{f_0^{-1}}$ such that $\tilde{\mathcal{L}}\xi_j = \lambda_j\xi_j$, $\lambda_j \in \mathbb{R}$, such that $\lim_{j\to\infty} |\lambda_j| = \infty$ [18, Theorem 11.3.13]. We make use of Lemma 3.1, which allows us to compare the eigenvalues of $\tilde{L}$ to those where $\mathbf{Z}_i \equiv 0$, and the fact that the eigenfunctions and eigenvalues of the resulting operator can be constructed explicitly. We note that the $P_{n,\mathbf{a}}$ form a basis of $L^2(\mathbb{R}^3, \mathrm{e}^{-|\mathbf{p}|^2/2})$, and so the functions $\mathrm{e}^{-|\mathbf{p}|^2/2} P_{n,\mathbf{a}}$ form a basis of $L^2(\mathbb{R}^3, \mathrm{e}^{|\mathbf{p}|^2/2})$.

First note, by Lemma 3.12, that the spaces

$$\mathrm{Span}\{\,\mathrm{e}^{-|\mathbf{p}|^2/2} P_{n,\mathbf{a}}(\mathbf{p})|n \text{ fixed}, |\mathbf{a}| = n\}$$

(where the coefficients may be functions of $\mathbf{r}$, t) are invariant under $\tilde{\mathcal{L}}$. Thus all eigenfunctions may be written in the form

$$(A.1) \qquad \psi_{n,j}(\mathbf{r}, \mathbf{p}, t) = \sum_{\mathbf{a}} \beta_{\mathbf{a},j}(\mathbf{r}, t)\,\mathrm{e}^{-|\mathbf{p}|^2/2} P_{n,\mathbf{a}}(\mathbf{p}),$$

where $j = 1, \ldots, T(n+1)$, with $T(n)$ the nth triangular number (which corresponds to the number of solutions to $a_1 + a_2 + a_3 = n - 1$). Also, as noted in the proof of Lemma 3.12, $\tilde{\mathcal{N}}(f, f_0)$ contributes only for $n = 1$, and as such we may ignore it when calculating the eigenvalues. It therefore suffices to consider the eigenvalues of

$$\bar{\mathcal{L}}f(\mathbf{r}, \mathbf{p}, t) = \boldsymbol{\nabla}_{\mathbf{p}} \cdot \left[\int d\mathbf{r}' d\mathbf{p}' g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t) \big(\tfrac{1}{N-1}\mathbf{1} + \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \big) (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t) \right]$$

$$=: \boldsymbol{\nabla}_{\mathbf{p}} \cdot \bar{\mathbf{Z}}(\mathbf{r}, t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) f(\mathbf{r}, \mathbf{p}, t).$$

Suppose $-\bar{\mathcal{L}}\psi_{n,j} = \lambda_{n,j}\psi_{n,j}$; then

$$\lambda_{n,j} \int d\mathbf{r} d\mathbf{p}\, f_0^{-1}(\mathbf{r}, \mathbf{p}, t) |\psi_{n,j}(\mathbf{r}, \mathbf{p}, t)|^2$$

$$= - \int d\mathbf{r} d\mathbf{p}\, f_0^{-1}(\mathbf{r}, \mathbf{p}, t) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t) \boldsymbol{\nabla}_{\mathbf{p}} \cdot \bar{\mathbf{Z}}(\mathbf{r}, t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t)$$

$$= \int d\mathbf{r} d\mathbf{p}\, f_0^{-1}(\mathbf{r}, \mathbf{p}, t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t) \cdot \bar{\mathbf{Z}}(\mathbf{r}, t)(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t)$$

$$= \int d\mathbf{r} d\mathbf{p} d\mathbf{r}' d\mathbf{p}'\, f_0^{-1}(\mathbf{r}, \mathbf{p}, t) g(\mathbf{r}, \mathbf{r}') f_0(\mathbf{r}', \mathbf{p}', t)$$
$$\times (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t) \cdot \big(\tfrac{1}{N-1}\mathbf{1} + \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \big) (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t)$$

$$= \int d\mathbf{r} d\mathbf{p} d\mathbf{r}' d\mathbf{p}'\, f_0(\mathbf{r}, \mathbf{p}, t) f_0(\mathbf{r}', \mathbf{p}', t) g(\mathbf{r}, \mathbf{r}')$$
$$\times \mathbf{v}(\mathbf{r}, \mathbf{p}, t) \cdot \big(\tfrac{1}{N-1}\mathbf{1} + \mathbf{Z}_1(\mathbf{r}, \mathbf{r}') \big) \mathbf{v}(\mathbf{r}, \mathbf{p}, t),$$

where the second equality follows via integration by parts and since $f_0^{-1}(\mathbf{r}, \mathbf{p}, t) = \rho_0^{-1}(\mathbf{r}, t) Z \exp(|\mathbf{p}|^2/2)$, and we denote $\mathbf{v}(\mathbf{r}, \mathbf{p}, t) = (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t) f_0^{-1}(\mathbf{r}, \mathbf{p}, t)$. Now note that Lemma 3.1 holds when $\mathbf{Z}_2$ is set to zero since it requires only that $\boldsymbol{\Gamma}$ be positive definite with the correct symmetry. Since $\boldsymbol{\Gamma}_{11}$ is a principal minor of $\boldsymbol{\Gamma}$, it is positive definite, and by symmetry so are all $\boldsymbol{\Gamma}_{jj}$. It therefore follows that the block diagonal matrix with entries $\boldsymbol{\Gamma}_{jj}$ is also positive definite, with the same required symmetry as $\boldsymbol{\Gamma}$, and we have

$$\lambda_{n,j} \int d\mathbf{r} d\mathbf{p}\, f_0^{-1}(\mathbf{r}, \mathbf{p}, t) |\psi_{n,j}(\mathbf{r}, \mathbf{p}, t)|^2$$

$$\geq \delta \int d\mathbf{r} d\mathbf{p}\, f_0(\mathbf{r}, \mathbf{p}, t) |\mathbf{v}(\mathbf{r}, \mathbf{p}, t)|^2 = \delta \int d\mathbf{r} d\mathbf{p}\, f_0^{-1}(\mathbf{r}, \mathbf{p}, t) |(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t)|^2$$

$$= -\delta \int d\mathbf{r} d\mathbf{p}\, f_0^{-1}(\mathbf{r}, \mathbf{p}, t) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t) \boldsymbol{\nabla}_{\mathbf{p}} \cdot (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t).$$

We now compute $\boldsymbol{\nabla}_{\mathbf{p}} \cdot (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \psi_{n,j}(\mathbf{r}, \mathbf{p}, t)$:

$$\boldsymbol{\nabla}_{\mathbf{p}} \cdot (\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) \, e^{-|\mathbf{p}|^2/2} \, P_{n,\mathbf{a}}(\mathbf{p}) = \sum_{j=1}^{3} \partial_{p_j} (p_j + \partial_{p_j}) \, e^{-|\mathbf{p}|^2/2} \, P_{n,\mathbf{a}}(\mathbf{p})$$

$$= \sum_{j=1}^{3} \partial_{p_j} \big(e^{|\mathbf{p}|^2/2} \, \partial_{p_j} P_{n,\mathbf{a}}(\mathbf{p}) \big) = \sum_{j=1}^{3} \partial_{p_j} \big(e^{|\mathbf{p}|^2/2} \, a_j P_{n,\mathbf{a}-\mathbf{e}_j}(\mathbf{p}) \big)$$

$$= \sum_{j=1}^{3} e^{|\mathbf{p}|^2/2} \, a_j (-p_j + \partial_{p_j}) P_{n,\mathbf{a}-\mathbf{e}_j}(\mathbf{p}) = - e^{|\mathbf{p}|^2/2} \, P_{n,\mathbf{a}}(\mathbf{p}) \sum_{j=1}^{3} a_j$$

$$= -n \, e^{|\mathbf{p}|^2/2} \, P_{n,\mathbf{a}}(\mathbf{p}).$$

The required identities for operators on $P_{n,\mathbf{a}}$ follow from its product form and the equivalent one-dimensional identities. Note that $\mathbf{e}_j$ is the jth unit vector. Thus we have

$$\lambda_{n,j} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r},\mathbf{p},t)|\psi_{n,j}(\mathbf{r},\mathbf{p},t)|^2 \geq n\delta \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{p} f_0^{-1}(\mathbf{r},\mathbf{p},t)|\psi_{n,j}(\mathbf{r},\mathbf{p},t)|^2,$$

and the result follows. $\square$

A.4. Proof of Lemma 3.12. From (2.6a) and Lemma 3.4, for $\mathcal{L}_0 f$ and $\mathcal{N}_0(f_0,f)$, it is sufficient to consider a general operator

$$\overline{\mathcal{L}} := \boldsymbol{\nabla}_{\mathbf{p}} \cdot \mathbf{Z}(\mathbf{r})(\mathbf{p} + \boldsymbol{\nabla}_{\mathbf{p}}) = \sum_{i,j} Z_{ij}\partial_{p_i}(p_j + \partial_{p_j}).$$

We have, using the standard identity $\partial_x H_n(x) = nH_{n-1}(x)$,

(A.2) $\qquad (p + \partial_p)[H_a(p)\,\mathrm{e}^{-p^2/2}] = \mathrm{e}^{-p^2/2}\,\partial_p H_a(p) = \mathrm{e}^{-p^2/2}\,aH_{a-1}(p),$

and using $H_{n+1}(x) = xH_n(x) - \partial_x H_n(x)$, we find

$$\partial_p[H_a(p)\,\mathrm{e}^{-p^2/2}] = \mathrm{e}^{-p^2/2}\,(-p + \partial_p)H_a(p) = -H_{a+1}(p)\,\mathrm{e}^{-p^2/2}.$$

It is therefore clear that $\overline{\mathcal{L}}$ preserves $|\mathbf{a}| = n$, with the possibility of the new coefficients all being zero.

It remains to consider $\mathcal{N}_0(f,f_0)$, which by Lemma 3.4 is given by

$$\mathcal{N}_0(f,f_0) = -\tfrac{1}{mk_BT}f_0(\mathbf{r},\mathbf{p},t)\int \mathrm{d}\mathbf{r}'\mathrm{d}\mathbf{p}'g(\mathbf{r},\mathbf{r}')\mathbf{Z}_2(\mathbf{r},\mathbf{r}')(\mathbf{p}' + \boldsymbol{\nabla}_{\mathbf{p}'})f(\mathbf{r}',\mathbf{p}',t)\cdot \mathbf{p}.$$

Note that $P_0 = 1$. Using (A.2), $f_0 = Z^{-1}\,\mathrm{e}^{-p^2/(2mk_BT)}\,\rho_0(\mathbf{r},t)$, and the fact that the $P_{n,\mathbf{a}}$ are orthogonal, it is clear that the integral gives zero for any terms not proportional to $p_i = H_{1,i}$ and in this case returns something of the form $\alpha(\mathbf{r}) \cdot \mathbf{p}$.

Hence $\tilde{\mathcal{L}}$ preserves n and it remains to show that $\tilde{\mathcal{L}} \sum_{|\mathbf{a}|=n} \alpha_{n,\mathbf{a}}(\mathbf{r},p)P_{n,\mathbf{a}}(\mathbf{p}) = 0$ if and only if $\alpha_{n,\mathbf{a}} = 0$ for all $\mathbf{a}$. This follows from the null space of $\tilde{\mathcal{L}}$ being $\mathrm{e}^{-|\mathbf{p}|^2/2}\,\phi(\mathbf{r},t)$ (see Lemma 3.8), and thus containing only P_0, and the orthogonality of the $P_{n,\mathbf{a}}$. $\square$

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