Hydrodynamic behavior of Brownian particles in a position-dependent constant force-field.

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The diffusion equation in physical space-time for a Brownian particle driven by an external force-field has been derived by Smoluchowski in the two particular cases where the external field is uniform or varies linearly with position (elastic force). In more general cases, correction terms must be added to the Smoluchowski equation.

We show here how to use a multi-scale Chapman-Enskog expansion to obtain, in the hydrodynamic limit, the first corrective terms to the Smoluchowski equation, without any restriction on the friction coefficient, and for any sufficiently small position-dependent constant force-field. We also compare our approach with the works of Wilemski, Titulaer and van Kampen.

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I. INTRODUCTION

In 1915, Smoluchowski[1,2] raised the important question of how the usual diffusion equation should be modified in order to describe properly the collective motion of Brownian particles under the influence of a given external force-field. He proved that, for at least two special choices of the force-field, the diffusion process could be described by a common new equation, which now carries the name of the Austrian physicist. Some sixty years later, the problem was reconsidered by various authors[3–9], who applied to Kramers' equation an asymptotic procedure similar in spirit to the Chapman-Enskog expansion introduced for solving Boltzmann equation. The Smoluchowski equation could then be recovered as the first approximation to a more general diffusion equation and higher order correction terms have also been derived[3,4].

We believe that these important results may be improved in at least one significant direction. First, for the problem at hand, the most general asymptotics susceptible of a treatment by a Chapman-Enskog expansion actually involve, in the one-dimensional case, three *a priori* distinct small parameters; however, the results presented so far in the literature have always been obtained under the tacit assumption that a single infinitesimal quantity is sufficient to properly address the problem. The principal aim of this article is to propose a fresh investigation of this matter. In particular, we will present, using as examples cases where the external force field is sufficiently small, a new implementation of the Chapman-Enskog procedure which will hopefully make clearer the number and physical significance of the involved small parameters. An approach similar in spirit to the one adopted here has already been used successfully to derive a diffusion equation from the Relativistic Ornstein-Uhlenbeck Process[10,11]. We will also deduce from our investigations the first correction terms to the Smoluchowski equation for at least one possible set of asymptotics (see Sections IV and V).

This article is organized as follows. In Section II, we review rapidly some fundamentals about the dynamical model of diffusion which will be used in the rest of this work. Section III elaborates on the general philosophy underlying the Chapman-Enskog expansion and presents the various asymptotics considered in this article. Section IV is devoted to obtaining perturbative solutions to Kramers' equation along the lines presented in the preceding section. Section V derives the actual "reduced" transport equation corresponding to all solutions obtained in Section IV. The work in Sections IV and V is restricted to the few perturbation orders which are necessary to obtain the first correction terms to the Smoluchowski equation and to compare them, in Section VI, with those presented in the existing literature. In the conclusion of the article, we review rapidly our main results and mention some problems left open for further study.

II. FUNDAMENTALS

A. The microscopic model of diffusion

To describe the motion of a Brownian particle of mass m under the influence of a given force-field $\mathbf{f}$, we start with the Langevin-like system :

$$\begin{cases} \frac{d}{dt}\mathbf{x} &= \mathbf{v} \\ \frac{d}{dt}\mathbf{v} &= -\alpha\mathbf{v} + \frac{1}{m}\mathbf{G} + \frac{1}{m}\mathbf{f}, \end{cases} \tag{1}$$

Here, $\mathbf{x}$ and $\mathbf{v}$ are respectively the instantaneous position and velocity of the particle, α is a constant positive coefficient, and $\mathbf{G}$ is a stochastic force. The friction-like term $-\alpha\mathbf{v}$ and the stochastic force $\mathbf{G}$ model the collisional interaction of the Brownian particle with the particles of the surrounding fluid. As usual, we will suppose that $\mathbf{G}$ is actually a centered Gaussian white-noise or, somewhat more precisely, the derivative of a Wiener-process multiplied by a constant coefficient $(m^2\alpha^2\chi)$ [1], so that :

$$< G^i(t_1)G_j(t_2) > = -2m^2\alpha^2\chi \ \delta(t_2 - t_1)\delta^i_j, \quad \chi > 0,$$

Let $\Pi(t, \mathbf{x}, \mathbf{v})$ be the probability distribution function (in phase space) associated to the stochastic process defined by (1). It can be shown[12,5,102] that Π satisfies the following differential equation, known as Kramers' equation :

$$\partial_t\Pi + \nabla_{\mathbf{x}} \cdot (\mathbf{v}\Pi) + \nabla_{\mathbf{v}} \cdot (-\alpha\mathbf{v}\Pi) + \nabla_{\mathbf{v}} \cdot (\frac{1}{m}\mathbf{f}\Pi) = \alpha^2\chi\Delta_{\mathbf{v}}\Pi. \tag{2}$$

B. Dimensionless Kramers' equation

To lighten further algebraic manipulations, we feel convenient to express the external force $\mathbf{f}$ and the variables t, $\mathbf{x}$ and $\mathbf{v}$ in terms of natural units of force, time, position and velocity.

The natural time unit that comes directly out of (1) is α^{-1}. It represents the typical microscopic relaxation time of the stochastic process. We therefore choose as dimensionless time variable :

$$\underline{t} \equiv \alpha t.$$

The typical "thermal" velocity $\sqrt{\alpha\chi}$ will be chosen as velocity unit. The dimensionless velocity variable is consequently :

$$\underline{\mathbf{v}} \equiv \frac{1}{\sqrt{\alpha\chi}}\mathbf{v}.$$

A temperature T can be defined by $k_BT = m\alpha\chi$, so that the thermal velocity $\sqrt{\alpha\chi}$ is given by the usual expression $\sqrt{k_BT/m}$ (k_B is the Boltzmann constant).

The natural space unit $\sqrt{\frac{\chi}{\alpha}}$ is simply the ratio of the velocity unit to the time unit. The dimensionless position variable is then :

$$\underline{\mathbf{x}} \equiv \sqrt{\frac{\alpha}{\chi}}\mathbf{x}.$$

From the mass m and the natural units of space and time defined above, it is straightforward to obtain the natural unit of force : $m\sqrt{\alpha^3\chi}$. The dimensionless external force is thus defined as :

---

[1] The coefficient χ is the diffusion coefficient in physical space; see Section IV.

[2] In Ref.[10], the derivation of Kramers' equation has been carried out in the (special) relativistic framework, which straightforwardly degenerates, in the proper limit, into the Galilean case.

$$\underline{\mathbf{f}} \equiv \frac{1}{m\sqrt{\alpha^3\chi}}\mathbf{f}.$$

In term of these dimensionless variables, Kramers' equation reads :

$$\partial_{\underline{t}}\Pi + \nabla_{\underline{\mathbf{x}}} \cdot (\underline{\mathbf{v}}\Pi) - \nabla_{\underline{\mathbf{v}}} \cdot (\underline{\mathbf{v}}\Pi) + \nabla_{\underline{\mathbf{v}}} \cdot (\underline{\mathbf{f}}\Pi) = \Delta_{\underline{\mathbf{v}}}\Pi. \tag{3}$$

C. Hypotheses and restrictions

Throughout this article, we will make the following assumptions :

1. When $|\mathbf{v}|$ tends to infinity, the probability distribution $\Pi(t, \mathbf{x}, \mathbf{v})$ and all its derivatives with respect to $\mathbf{v}$ vanish more rapidly than any power of $\mathbf{v}$, for all time and position :

$$\forall\ k, l, t, \mathbf{x}, \qquad \lim_{|\mathbf{v}|\to\infty} \mathbf{v}^k \partial_{v^l}\Pi(t, \mathbf{x}, \mathbf{v}) = 0.$$

2. For technical simplicity reasons, we restrict our study to the one-dimensional case. We therefore work with a one-dimensional probability distribution $\Pi(t, x, v)$ and with a one-dimensional version of (2) :

$$\partial_t\Pi + \partial_x(v\Pi) - \partial_v(\alpha v\Pi) + \partial_v(\frac{1}{m}f\Pi) - \alpha^2\chi\partial_{vv}\Pi = 0. \tag{4}$$

   The dimensionless form of (4) reads :

$$\partial_{\underline{t}}\Pi + \partial_{\underline{x}}(\underline{v}\Pi) - \partial_{\underline{v}}(\underline{v}\Pi) + \partial_{\underline{v}}(\underline{f}\Pi) - \partial_{\underline{vv}}\Pi = 0. \tag{5}$$

3. The typical linear size $\mathcal{L}$ of the accessible region in physical space will be assumed to be finite although very large compared to any physically relevant length scale of the problem. This gives a well-defined meaning to the notion of uniform particle-density in physical space.

4. The probability distribution $\Pi(t, x, v)$, the external force f and all their derivatives are supposed to exist for any value of (t, x, v).

The main points of this article are, first, present a new implementation of Chapman-Enskog method applied to Kramers' equation, and, second, to derive from (4), under the above listed hypotheses, an evolution equation for the particle density in physical space, in the so-called "hydrodynamic" limit.

D. The momentum hierarchy

Let us now consider the hierarchy of (evolution-)equations obtained by multiplying (5) by $\underline{v}^k$, $k \in \mathbb{N}$ and integrating the result over velocity space. Using the hypotheses presented in Section II C, the level k of the hierarchy takes the form :

$$\begin{aligned} &\partial_{\underline{t}}(n< \underline{v}^k >) \ + \ \partial_{\underline{x}}(n< \underline{v}^{k+1} >) \ + \ kn< \underline{v}^k > - \\ &k\underline{f}n< \underline{v}^{k-1} > \ - \ k(k-1)n< \underline{v}^{k-2} > \ = \ 0, \qquad \text{for} \quad k \geq 0, \end{aligned} \tag{6}$$

where the symbol $< >$ designates the average over $\underline{v}$:

$$n< \psi > \equiv \int_{\mathbb{R}} \psi(\underline{v})\Pi(\underline{t}, \underline{x}, \underline{v})d\underline{v}.$$

In particular, the levels $k = 0$ and $k = 1$ provide the balance equations for the particle and momentum density :

$$\begin{aligned} &\partial_{\underline{t}}(n) \ + \ \partial_{\underline{x}}(n< \underline{v} >) \ = \ 0, \\ &\partial_{\underline{t}}(n< \underline{v} >) \ + \ \partial_{\underline{x}}(n< \underline{v}^2 >) \ + \ n< \underline{v} > \ - \ \underline{f}n \ = \ 0. \end{aligned} \tag{7}$$

The hierarchy (6) will be of crucial importance in establishing the possible asymptotic behaviors of the system in the next section.

III. THE PRINCIPLE BEHIND THE CHAPMAN-ENSKOG EXPANSION AND THE VARIOUS POSSIBLE ASYMPTOTICS

The general idea behind the Chapman-Enskog expansion as we implement it in this article is to solve perturbatively the dimensionless Kramers' equation (5) by searching for "slowly varying" solutions in space and time which correspond to a given spatial probability density $n(\underline{t},\underline{x})$. To be more specific, let us choose a "sufficiently regular" function $n(\underline{t},\underline{x})$ and try to find solutions of (5) which give back this density when integrated over the whole velocity space. The evolution equations for the various macroscopic "hydrodynamical" quantities will appear in this perspective as solvability conditions.

It is to be noted that this whole approach of solving Kramers' equation differs fundamentally from the more usual "physical" one which consists in fixing initial and boundary conditions and use them to generate the solution at all (subsequent) time. If one follows this more traditional approach one is naturally led to distinguish, for example, between a transient regime and long time behavior. Both types of solutions appear however on equal footing if one uses the Chapman-Enskog method, since the very notion of "initial condition" is absent from the whole formalism. From the Chapman-Enskog point of view the data of the problem are Kramers' equation, and a given spatial probability density *at all time*.

In the special case where $\underline{f}$ vanishes identically, a possible solution of (5) corresponding to the constant density $n(\underline{t},\underline{x}) = n_0$ is the global equilibrium distribution :

$$\Pi^{(eq)}(\underline{v}) = n_0 \frac{1}{\sqrt{2\pi}} \exp\,(-\frac{\underline{v}^2}{2}). \tag{8}$$

It seems therefore reasonable, for densities $n(\underline{t},\underline{x})$ which vary sufficiently slowly in space and time, to search for possible solutions of Kramers' equation in the form of an expansion around the local equilibrium distribution (9), at least if the force-field $\underline{f}$ is sufficiently small and has also slow spatial variations :

$$\Pi^{(loc)}(\underline{v}) = n(\underline{t},\underline{x}) \frac{1}{\sqrt{2\pi}} \exp\,(-\frac{\underline{v}^2}{2}). \tag{9}$$

Without restricting for the moment the choice of $\underline{f}$, let us pick two "small parameters" (*i.e.* infinitesimal quantities) ϵ and η, and consider a spatial density $n(\underline{t},\underline{x})$ which, in the domain of space-time under consideration, verifies the scaling relations :

$$\begin{aligned} \frac{\partial_{\underline{x}}\, n}{n} &= \mathcal{O}(\epsilon), \\ \frac{\partial_{\underline{t}}\, n}{n} &= \mathcal{O}(\eta). \end{aligned} \tag{10}$$

These relations state that the spatial variation scale of n is large compared to the mean free path $\sqrt{k_B T/m\alpha^2}$, and that its time variation scale is large compared to the mean flight time α^{-1}. The two parameters ϵ and η are *a priori* independent. However, in the degenerated case of free diffusion ($\mathbf{f} = 0$), both parameters are naturally linked by $\eta = \epsilon^2$ (see the discussion in Section VI B).

To make proper use of these relations, it is best to turn our attention again to the hierarchy (6). Because $n< \underline{v}^{2k+1} >$, $k \in \mathbb{N}$, vanishes for the local equilibrium distribution (9), let us introduce a new "small parameter" ϵ' and search for solutions of Kramers' equation which verify :

$$n< \underline{v} > = \mathcal{O}(\epsilon'). \tag{11}$$

If one supposes, in agreement with the preceding paragraph, that the sought-for distribution function can be written as an expansion around (9), a simple integration by part shows that the scaling (11) is actually valid for any odd power of the velocity :

$$n< \underline{v}^{2k+1} > = \mathcal{O}(\epsilon'), \qquad k \in \mathbb{N}. \tag{12}$$

The level $k = 0$ in the hierarchy (6) then delivers immediately that $\eta = \epsilon\epsilon'$ and the level $k = 1$ implies that $\underline{f}$ has also to be a vanishingly small quantity, whose order will be hereafter denoted by ν. One is therefore left with the following possibilities for balancing properly the terms in (7) :

$$
\begin{aligned}
&(i) \quad \epsilon' = \epsilon \quad \text{and} \quad \nu \leq \epsilon, \\
&(ii) \quad \nu = \epsilon \quad \text{and} \quad \epsilon' \leq \epsilon, \\
&(iii) \quad \nu = \epsilon' \quad \text{and} \quad \epsilon \leq \nu.
\end{aligned}
\tag{13}
$$

Each one of these alternatives defines a particular family of solutions of Equation (5). It is already apparent at this stage that each family subdivides into at least two sub-families; the first one corresponds to a strict inequality in (13) and the second one to the case in which all three small parameters ϵ, ϵ' and ν are actually identical. The situation is however more complicated than this because ϵ, ϵ', $\eta = \epsilon\epsilon'$ and ν are not the only infinitesimal quantities *a priori* involved in the problem. Indeed, (7) also constrains the spatial variations of n with respect to those of $\underline{f}$. To investigate this matter further, it is convenient to introduce a fifth (infinitesimal) quantity ϵ'' such that:

$$
\frac{\partial_{\underline{x}} \underline{f}}{\underline{f}} = \mathcal{O}(\epsilon''),
\tag{14}
$$

and to derive the level k of the hierarchy (6) with respect to x^p, $p \in \mathbb{N}$:

$$
\begin{aligned}
&\partial_{\underline{t}} \partial_{\underline{x}^p} (n < \underline{v}^k >) \; + \; \partial_{\underline{x}^{p+1}} (n < \underline{v}^{k+1} >) \; - \; k \partial_{\underline{x}^p} (\underline{f} n < \underline{v}^{k-1} >) \; = \\
&k(k-1) \partial_{\underline{x}^p} (n < \underline{v}^{k-2} >) \; - \; k \partial_{\underline{x}^p} (n < \underline{v}^k >) \qquad \text{for} \quad k \geq 0.
\end{aligned}
\tag{15}
$$

If k is even, then both terms on the right-hand side of (15) are of order ϵ^p and the first two terms on the left-hand side are clearly of a higher order. Let us suppose that ϵ'' is strictly superior to ϵ. Then, the main contribution to the third term on the left-hand side of (15) is of order $\nu \epsilon''^p$ and this quantity has to be inferior to ϵ^p, for all even integers p. Let us now specialize the discussion according to the family under consideration. For family *(i)*, one can always find a non-negative integer q such that:

$$
\epsilon^{q+1} \leq \nu \leq \epsilon^q.
$$

This implies that, for all even integers p:

$$
\epsilon'' \leq \epsilon^{(p-q-1)/p},
$$

which obviously contradicts the hypothesis $\epsilon'' > \epsilon$. Similarly, for family *(iii)*, one can again introduce a non-negative integer q verifying:

$$
\epsilon^{-q} \leq \nu \leq \epsilon^{-(q+1)},
$$

so that, for all even integers p, one has:

$$
\epsilon'' \leq \epsilon^{(p+q)/p},
$$

which again contradicts the hypothesis $\epsilon'' > \epsilon$. Finally, for family *(ii)*, one has directly that, for all even integers p:

$$
\epsilon'' \leq \epsilon^{(p-1)/p},
$$

which delivers the same result as the one obtained for the other families. A similar argument for odd values of k delivers the same conclusion. We can therefore conclude that in all cases ϵ has to be superior or equal to ϵ''.

Taking into account all four parameters ϵ, ϵ', ϵ'' and ν as well as their possible relationships to one another, we can sum up the preceding discussion in the following way. There are three main families of solutions which are a priori susceptible of a treatment by the Chapman-Enskog method. Each of these families can be subdivided into four different sub-families of solutions. In each family, the first sub-family encompasses three-parameters solutions, the second and third sub-family both represent two-parameters solutions, the second sub-family being actually common to all main families. The fourth sub-family is also common to the three families and involves a single infinitesimal quantity. These conclusions are displayed in a more compact form in Table I and II.

One of the advantages of the Chapman-Enskog procedure is to provide an elegant way to recover the Smoluchowski equation and its various corrections. Indeed, it actually turns out that, for a given spatial density, the whole expansion is only feasible if, at any order, the involved coefficients satisfy various constraints or solvability conditions in the form of partial differential equations. The density and the force f are the only "data" of the problem. It therefore follows that all coefficients in the expansion depend only on the density, the imposed force-field f and their various time- and space-derivatives. Consequently, the constraints satisfied by these coefficients can be transcribed, at any order, into differential equations which are to be verified by the density, the force f and their various derivatives for the whole expansion procedure to exist. These differential equations are actually the evolution equations for the spatial density one is usually looking for.

IV. PERTURBATIVE RESOLUTION OF KRAMERS' EQUATION FOR FAMILY *(II)*

A. The general framework

Let us now present, in the "hydrodynamic" limit, a direct resolution of Kramers' equation by a generalized multi-scale Chapman-Enskog expansion. The dimensionless Kramers' equation reads:

$$\partial_{\underline{t}}\Pi + \underline{v}\partial_{\underline{x}}\Pi + \underline{f}\partial_{\underline{v}}\Pi = \partial_{\underline{v}}\Big((\underline{v} + \partial_{\underline{v}})\Pi\Big). \tag{16}$$

Each family and/or sub-family envisaged in Section III corresponds to a different physical situation, the only exception being the most degenerated case, which is common to all three main families (see Table I). A complete examination of the problem at hand should therefore involve a detailed study of ten different cases. This lies clearly outside the scope of the present article; we will now present the perturbative solutions to Kramers' equation belonging to one of the three main families only. Let us point out however that preliminary calculations seem to indicate that the major steps in the Chapman-Enskog procedure are similar for all three families. There is no physical or mathematical *a priori* reason to prefer one family to the other two. In what follows, we will concentrate on family *(ii)*.

B. The asymptotic expansion for sub-family *(ii.1)*

Using the small parameters presented in Section III, we introduce new rescaled space-, time-, and force- variables $X = \epsilon\underline{x}, Y = \epsilon''\underline{x}, T = \epsilon\epsilon'\underline{t}, \underline{f} = \nu F$ and rewrite (16) as:

$$\epsilon\epsilon'\partial_T\Pi + \underline{v}\Big(\epsilon\partial_X\Pi + \epsilon''\partial_Y\Pi\Big) + \nu F\partial_{\underline{v}}\Pi = \partial_{\underline{v}}\Big((\underline{v} + \partial_{\underline{v}})\Pi\Big). \tag{17}$$

A reasonable form for the Chapman-Enskog expansion corresponding to sub-family *(ii.1)* is:

$$\Pi = \sum_{k,l,m\in\mathbb{N}^3} \epsilon^k \left(\frac{\epsilon'}{\epsilon}\right)^l \left(\frac{\epsilon''}{\epsilon}\right)^m \Pi_{klm}, \tag{18}$$

where Π_{klm} is a function of X, Y, T and $\underline{v}$. Substituting Expression (18) in Equation (17) and collecting all terms of order $\epsilon^k \left(\frac{\epsilon'}{\epsilon}\right)^l \left(\frac{\epsilon''}{\epsilon}\right)^m$, we obtain:

$$\begin{aligned}\partial_T\Pi_{k-2l-1m} + \underline{v}\Big(\partial_X\Pi_{k-1lm} + \partial_Y\Pi_{k-1lm-1}\Big) + F\partial_{\underline{v}}\Pi_{k-1lm} \\ = \partial_{\underline{v}}\Big((\underline{v} + \partial_{\underline{v}})\Pi_{klm}\Big),\end{aligned} \tag{19}$$

with the convention that Π_{klm} vanishes for any strictly negative value of either k, l or m. We rewrite Equation (19) in the more condensed form:

$$\begin{aligned}\partial_T\Big(P\cdot\overset{\circ}{\Pi}_{k-2l-1m}\Big) + \underline{v}\partial\cdot\overset{\circ}{\Pi}_{k-1lm} + F\partial_{\underline{v}}\Big(P\cdot\overset{\circ}{\Pi}_{k-1lm}\Big) \\ = \partial_{\underline{v}}\Big((\underline{v} + \partial_{\underline{v}})P\cdot\overset{\circ}{\Pi}_{klm}\Big),\end{aligned} \tag{20}$$

where the vector-like quantity $\overset{\circ}{\Pi}_{klm}$ is defined by: $\overset{\circ}{\Pi}_{klm} = \begin{pmatrix}\Pi_{klm} \\ \Pi_{klm-1}\end{pmatrix}$. In (20), P and ∂ represent respectively the adjoint-vectors $(1;0)$ and $(\partial_X;\partial_Y)$. The usefulness of this rather abstract vectorial formalism and of the covariant derivative $\mathcal{D}$ to be introduced below in Section IV B 2 may not be quite apparent at this stage. The principle advantage provided by these notations is to furnish cumbersome results obtained in this article in the most possible tractable form. The conversion back to more usual notations will be carried out at the end of all calculations in Section V A. We will now present in full detail the resolution of (20) for $0 \leq k \leq 4$.

1. Order $k = 0$

Setting $k = 0$ in Equation (20) gives :

$$\partial_{\underline{v}}\Big((\partial_{\underline{v}} + \underline{v})P\cdot \overset{\circ}{\Pi}_{0lm}\Big) = 0. \tag{21}$$

The only solutions of (21) compatible with Hypothesis 1 are of the form :

$$\overset{\circ}{\Pi}_{0lm} = \overset{\circ}{A}_{0lm} e^{-\frac{\underline{v}^2}{2}}, \tag{22}$$

where $\overset{\circ}{A}_{0lm}$ is a function of X, Y et T. The A_{0lm}'s and the other similar functions to be introduced below are not arbitrary. Their link to the spatial density n will be discussed in Section IV F.

2. Order $k = 1$

Setting k equal to one in (5) and using the expression (22) for $\overset{\circ}{\Pi}_{0lm}$, we obtain :

$$\underline{v}\mathcal{D}\cdot \overset{\circ}{A}_{0lm} e^{-\frac{\underline{v}^2}{2}} = \partial_{\underline{v}}\Big((\underline{v} + \partial_{\underline{v}})P\cdot \overset{\circ}{\Pi}_{1lm}\Big), \tag{23}$$

where $\mathcal{D}$ is a "covariant"-derivative defined by $\mathcal{D} = \partial - FP$. Hypothesis 1 leads us to retain, as only solutions of (23) :

$$\overset{\circ}{\Pi}_{1lm} = \Big\{\overset{\circ}{A}_{1lm} - \underline{v}\,\overset{\circ}{(}\mathcal{D}\cdot \overset{\circ}{A}_{0lm})\Big\} e^{-\frac{\underline{v}^2}{2}}, \tag{24}$$

where $\overset{\circ}{A}_{1lm}$ is a function of X, Y and T. Up to this order, no solvability condition has to be imposed to obtain solutions of (20) verifying Hypothesis 1.

3. Order $k = 2$

We set k equal to two in (20) and use for $\overset{\circ}{\Pi}_{0lm}$ and $\overset{\circ}{\Pi}_{1lm}$ the expressions (22) and (24) to obtain :

$$\begin{aligned}\Big\{\partial_T\Big(P\cdot \overset{\circ}{A}_{0l-1m}\Big) - F\Big(P\cdot \overset{\circ}{(}\mathcal{D}\cdot \overset{\circ}{A}_{0lm})\Big) + \underline{v}\Big(\mathcal{D}\cdot \overset{\circ}{A}_{1lm}\Big) \\ -\underline{v}^2\mathcal{D}\cdot \overset{\circ}{\Big(}\mathcal{D}\cdot \overset{\circ}{A}_{0lm}\Big)\Big\} e^{-\frac{\underline{v}^2}{2}} = \partial_{\underline{v}}\Big((\underline{v} + \partial_{\underline{v}})P\cdot \overset{\circ}{\Pi}_{2lm}\Big).\end{aligned} \tag{25}$$

The commutation relation $[\mathcal{D}; P] = 0$ has also been used in deriving (25). To satisfy Hypothesis 1, the integral over $\underline{v}$ of the left-hand side must vanish. Indeed, let us take the primitive of both sides of (25). The primitive of the right-hand side clearly vanishes when $\underline{v}$ tends to infinity if Hypothesis 1 is verified. Thus, the primitive of the left-hand side must also vanish when $\underline{v}$ tends to infinity. Hence, the integral over $\underline{v}$ of the left-hand side must be zero. We thus find that a necessary (and sufficient) condition for the integral over $\underline{v}$ of the left-hand side of (25) to vanish is :

$$\partial_T(P\cdot \overset{\circ}{A}_{0l-1m}) - \partial\cdot \overset{\circ}{(}\mathcal{D}\cdot \overset{\circ}{A}_{0lm}) = 0. \tag{26}$$

Assuming this solvability condition to be fulfilled, the only solutions of (25) which verify Hypothesis 1 are :

$$\overset{\circ}{\Pi}_{2lm} = \Big\{\overset{\circ}{A}_{2lm} - \underline{v}\,\overset{\circ}{(}\mathcal{D}\cdot \overset{\circ}{A}_{1lm}) + \frac{\underline{v}^2}{2}\overset{\circ}{\Big(}\mathcal{D}\cdot \overset{\circ}{(}\mathcal{D}\cdot \overset{\circ}{A}_{0lm})\Big)\Big\} e^{-\frac{\underline{v}^2}{2}}, \tag{27}$$

where $\overset{\circ}{A}_{2lm}$ is again a function of X, Y and T.

4. Order $k = 3$

With $k = 3$ in (20) and the expressions (24) and (27) for $\overset{\circ}{\Pi}_{1lm}$ and $\overset{\circ}{\Pi}_{2lm}$ respectively, we obtain :

$$\begin{aligned}
\Big\{ \quad & \partial_T\Big(P\cdot\overset{\circ}{A}_{1l-1m}\Big) - F\Big(P\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{1lm})\Big) \\
& + \underline{v}\Big(\mathcal{D}\cdot\overset{\circ}{A}_{2lm} - \overset{\circ}{[}\mathcal{D},\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm}) - \mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big)\Big) \\
& - \underline{v}^2\Big(\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{1lm})\Big) \\
& + \frac{\underline{v}^3}{2}\Big(\mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big)\Big)\Big\}e^{-\frac{\underline{v}^2}{2}} \\
& = \partial_{\underline{v}}\Big((\underline{v}+\partial_{\underline{v}})P\cdot\overset{\circ}{\Pi}_{3lm}\Big),
\end{aligned} \tag{28}$$

where the condensed notation $\overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{A}$ stands for :

$$\overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{A} = \mathcal{D}\cdot\overset{\circ}{(}\partial\cdot\overset{\circ}{A}) - \partial\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}).$$

Note that the straightforward commutation relations $[\mathcal{D};P] = 0$ and $[\partial_T;\mathcal{D}] = 0$ have been used in deriving (28). The solvability condition associated to (28) is :

$$\partial_T(P\cdot\overset{\circ}{A}_{1l-1m}) - \partial\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{1lm}) = 0. \tag{29}$$

The corresponding solutions compatible with Hypothesis 1 are :

$$\begin{aligned}
\overset{\circ}{\Pi}_{3lm} = \Big\{ \quad & \overset{\circ}{A}_{3lm} \\
& - \underline{v}\Big(\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{2lm}) - \overset{\circ}{\Big(}\overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big)\Big) \\
& + \frac{\underline{v}^2}{2}\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{1lm})\Big) \\
& - \frac{\underline{v}^3}{6}\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big)\Big)\Big\}e^{-\frac{\underline{v}^2}{2}},
\end{aligned} \tag{30}$$

where $\overset{\circ}{A}_{3lm}$ is a function of X, Y and T.

5. Order $k = 4$

If we set $k = 4$ in (20) and give to $\overset{\circ}{\Pi}_{2lm}$ and $\overset{\circ}{\Pi}_{3lm}$ the form (27) and (30), we have :

$$\begin{aligned}
\Big\{ \quad & \partial_T\Big(P\cdot\overset{\circ}{A}_{2l-1m}\Big) - F\Big(P\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{2lm})\Big) \\
& + \partial\cdot\overset{\circ}{\Big(}\overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big) - \mathcal{D}\cdot\overset{\circ}{\Big(}\overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big) \\
& + \underline{v}\Big(\mathcal{D}\cdot\overset{\circ}{A}_{3lm} - \overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{1lm}) - \mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{1lm})\Big)\Big) \\
& - \underline{v}^2\Big(\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{2lm}) - \mathcal{D}\cdot\overset{\circ}{\Big(}\overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big) \\
& - \frac{1}{2}\overset{\circ}{[}\mathcal{D}^2;\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm}) - \frac{1}{2}\mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big)\Big)\Big)
\end{aligned}$$

$$+\frac{\underline{v}^3}{2}\Big(\mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{1lm})\Big)\Big)$$
$$-\frac{\underline{v}^4}{6}\Big(\mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{\Big(}\mathcal{D}\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big)\Big)\Big)\Big\}e^{-\frac{\underline{v}^2}{2}}$$
$$=\partial_{\underline{v}}\Big((\underline{v}+\partial_{\underline{v}})P\cdot\overset{\circ}{\Pi}_{4lm}\Big). \tag{31}$$

In deriving this expression, we have used the solvability conditions (26) and (29) and the commutation relations $[\mathcal{D};P]=0$ and $[\partial_T;\mathcal{D}]=0$. Following again the logic of Sections IV B 3 and IV B 4, we obtain the solvability condition :

$$\partial_T\Big(P\cdot\overset{\circ}{A}_{2l-1m}\Big)-\partial\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{2lm})=-\partial\cdot\Big(\overset{\circ}{[}\mathcal{D};\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm})\Big)$$
$$-\frac{1}{2}\overset{\circ}{[}\mathcal{D}^2;\partial]\cdot\overset{\circ}{(}\mathcal{D}\cdot\overset{\circ}{A}_{0lm}). \tag{32}$$

To obtain the desired corrections terms to Smoluchowski equation, we will only need at this order the solvability condition (32) and no explicit expression for $\overset{\circ}{\Pi}_{4lm}$.

C. The asymptotic expansion for sub-family *(ii.2)*

The sub-family *(ii.1)* formally degenerates into *(ii.2)* if $\epsilon'=\epsilon$. The Chapman-Enskog expansion of Π takes therefore the form :

$$\Pi=\sum_{k,m\in\mathbb{N}^2}\epsilon^k\left(\frac{\epsilon''}{\epsilon}\right)^m\Pi_{km}, \tag{33}$$

where

$$\Pi_{km}=\sum_{l\in\mathbb{N}}\Pi_{klm}.$$

The expressions for Π_{klm}, $0\leq k\leq 3$, obtained in the preceding section furnish directly by summation over l the form of the desired Π_{km}, $0\leq k\leq 3$. The solvability conditions associated to (33) can also be straightforwardly deduced from those given in Section IV B and do not need to be explicitly presented here.

D. The asymptotic expansion for sub-family *(ii.3)*

Contrary to *(ii.2)*, sub-family *(ii.3)* cannot be directly deduced from sub-family *(ii.1)*. The main reason for this is simply that *(ii.1)* and *(ii.2)* involve two spatial scales whereas *(ii.3)* only involves one. The corresponding dimensionless Kramers' equation is :

$$\epsilon\epsilon'\partial_T\Pi+\underline{v}\epsilon\partial_X\Pi+\epsilon F\partial_{\underline{v}}\Pi=\partial_{\underline{v}}\Big((\underline{v}+\partial_{\underline{v}})\Pi\Big), \tag{34}$$

where Π_{kl} is now a function of X, T and $\underline{v}$ only. As in Sections IV B and IV C, solutions of (34) are to be sought for under the form :

$$\Pi=\sum_{k,l\in\mathbb{N}^2}\epsilon^k\left(\frac{\epsilon'}{\epsilon}\right)^l\Pi_{kl}. \tag{35}$$

Substituting (35) in Equation (34) and collecting all terms of order $\epsilon^k\Big(\frac{\epsilon'}{\epsilon}\Big)^l$, we obtain :

$$\partial_T\Pi_{k-2l-1}+\underline{v}\partial_X\Pi_{k-1l}+F\partial_{\underline{v}}\Pi_{k-1l}=\partial_{\underline{v}}\Big((\underline{v}+\partial_{\underline{v}})\Pi_{kl}\Big), \tag{36}$$

with the convention that Π_{kl} vanishes if k or l is strictly negative. There is however an important technical difference between (36) and (19). In (19), the index m traced back the presence of two spatial scales and motivated the introduction of the vectorial formalism used in Sections IV B and IV C. Because (34) involves only one spatial scale, this formalism is of no use for solving (36). Solving (36) is consequently an essentially similar but simpler process than solving (19) for sub-family *(ii.1)*. It seems therefore unnecessary to present here the corresponding cumbersome algebra.

E. The asymptotic expansion for sub-family *(ii.4)*

Because sub-family *(ii.3)* degenerates into sub-family *(ii.4)* for $\epsilon' = \epsilon$, all results pertaining to this last sub-family can be formally deduced from those discussed in Section IV D by a summation over l.

F. Choice of the coefficients in the Chapman-Enskog expansion

One usually fixes the coefficients in the Chapman-Enskog expansion by normalizing the lowest order term to the given spatial density n and by requiring consequently that all superior orders are normalized to zero[13,11]. When applied to sub-family *(ii.4)*, this delivers:

$$\int_{\mathbb{R}} \Pi_0 \, d\underline{v} = n, \tag{37}$$

and

$$\int_{\mathbb{R}} \Pi_k \, d\underline{v} = 0, \text{ for } k > 0. \tag{38}$$

Equation (37) clearly determines A_0 in terms of n and (38) delivers A_k once the lower order coefficients A_j, $j < k$, are known.

The implementation of this procedure for sub-families *(ii.3)*, *(ii.2)* and *(ii.1)* is more subtle. Let us start with sub-family *(ii.3)*. There are *a priori* two different ways of fixing the coefficients of the expansion. The first one would be to normalize Π_{00} to n and all other Π_{kl}'s to 0. This would have the direct consequence of actually setting to zero all Π_{k0}'s, for $k > 0$, and the corresponding expansion would not involve ϵ' but only ϵ. It is easy to check that such an expansion cannot satisfy Equation (34). This means that the retained normalization condition is too restrictive and therefore unsuitable to sub-family *(ii.3)*. The correct way of relating the A_{kl}'s to n is to impose:

$$\int_{\mathbb{R}} \sum_{l \in \mathbb{N}} \Big(\frac{\epsilon'}{\epsilon}\Big)^l \Pi_{0l} \, d\underline{v} = n, \tag{39}$$

and

$$\int_{\mathbb{R}} \sum_{l \in \mathbb{N}} \Big(\frac{\epsilon'}{\epsilon}\Big)^l \Pi_{kl} \, d\underline{v} = 0, \text{ for } k > 0. \tag{40}$$

(39) and (40) solve the problem at the price of not fixing unambiguously every A_{kl} but only their weighed sum over l: $\sum_{l \in \mathbb{N}} \Big(\frac{\epsilon'}{\epsilon}\Big)^l A_{kl}$.

A similar reasoning leads for sub-family *(ii.2)* to the conditions:

$$\int_{\mathbb{R}} \sum_{m \in \mathbb{N}} \Big(\frac{\epsilon''}{\epsilon}\Big)^m \Pi_{0m} \, d\underline{v} = n, \tag{41}$$

and

$$\int_{\mathbb{R}} \sum_{m \in \mathbb{N}} \Big(\frac{\epsilon''}{\epsilon}\Big)^m \Pi_{km} \, d\underline{v} = 0, \text{ for } k > 0, \tag{42}$$

and, for sub-family *(ii.1)*, to :

$$\int_{\mathbb{R}} \sum_{l,m\in\mathbb{N}^2} \left(\frac{\epsilon'}{\epsilon}\right)^l \left(\frac{\epsilon''}{\epsilon}\right)^m \Pi_{0lm}\, d\underline{v} = n, \tag{43}$$

and

$$\int_{\mathbb{R}} \sum_{l,m\in\mathbb{N}^2} \left(\frac{\epsilon'}{\epsilon}\right)^l \left(\frac{\epsilon''}{\epsilon}\right)^m \Pi_{klm}\, d\underline{v} = 0, \text{ for } k > 0. \tag{44}$$

V. THE FIRST CORRECTION TERMS TO SMOLUCHOWSKI EQUATION

In classical Brownian motion theory without external force-field, the spatial density n verifies, in the long-time limit the (dimensionless) diffusion equation :

$$\partial_{\underline{t}} n - \partial_{\underline{x}\underline{x}} n = 0. \tag{45}$$

When an external (time-independent) force-field is present, (45) has obviously to be modified. Smoluchowski has proved that, if $\underline{f}$ is uniform[1] or varies linearly with position[2], the exact transport equation for n is :

$$\partial_{\underline{t}} n - \partial_{\underline{x}\underline{x}} n + \partial_{\underline{x}}(n\underline{f}) = 0. \tag{46}$$

For a more general force-field, (46) is only valid approximately. Wilemski[3] and Titulaer[4] have obtained, for a sufficiently small force field, the lowest order correction terms to (46), which becomes :

$$\partial_{\underline{t}} n - \partial_{\underline{x}\underline{x}} n + \partial_{\underline{x}}(n\underline{f}) = -\partial_{\underline{x}}\{\partial_{\underline{x}}\underline{f}(\partial_{\underline{x}} n - \underline{f} n)\}. \tag{47}$$

Our approach, at least for family *(ii)*, leads to the same equation for n. We will now present in full detail how to reach (47) for sub-family *(ii.1)*. Sub-families *(ii.2)*, *(ii.3)* and *(ii.4)* will be also briefly discussed.

Let us define Δ as the correction to the Smoluchowski equation (46). We can write :

$$\Delta = \partial_{\underline{t}} n - \partial_{\underline{x}\underline{x}} n + \partial_{\underline{x}}(n\underline{f}) \tag{48}$$

Since all preceding developments involve the phase-space distribution Π, it is convenient to introduce the analogue of Δ in phase-space, δ, defined by :

$$\delta = \partial_{\underline{t}}\Pi - \partial_{\underline{x}\underline{x}}\Pi + \partial_{\underline{x}}(\Pi\underline{f}). \tag{49}$$

Δ can be recovered by a direct integration of δ over the velocity $\underline{v}$.

A. Sub-family *(ii.1)* and *(ii.2)*

Using the rescaled space-, time- and force-variables defined in Section IV A, Equation (49) becomes :

$$\begin{aligned}\delta = \epsilon\epsilon'\partial_T\Pi - \Big(&\epsilon^2\partial_{X^2}\Pi + 2\epsilon\epsilon''\partial_{XY}\Pi + (\epsilon'')^2\partial_{Y^2}\Pi\Big) \\ &+ \epsilon\Big(\epsilon\partial_X(\Pi F) + \epsilon''\partial_Y(\Pi F)\Big).\end{aligned} \tag{50}$$

The Chapman-Enskog expansion for Π introduced in Section IV B leads naturally to the following form for δ :

$$\delta = \sum_{k,l,m\in\mathbb{N}^3} \epsilon^k \left(\frac{\epsilon'}{\epsilon}\right)^l \left(\frac{\epsilon''}{\epsilon}\right)^m \delta_{klm}, \tag{51}$$

Replacing in (50) Π and δ by their respective expressions (18) and (51), we obtain :

$$\delta_{klm} = \partial_T \Pi_{k-2l-1m} - \Big(\partial_{X^2}\Pi_{k-2lm} + 2\partial_{XY}\Pi_{k-2lm-1} + \partial_{Y^2}\Pi_{k-2lm-2}\Big) + F\Big(\partial_X \Pi_{k-2lm} + \partial_Y \Pi_{k-2lm-1}\Big) + F_Y \Pi_{k-2lm-1}. \tag{52}$$

Because Π_{klm} vanishes for negative values of k, so do δ_{0lm} and δ_{1lm}. With the help of the vectorial formalism introduced in Section IV B, Equation (52) takes the more compact form :

$$\delta_{klm} = \partial_T(P\cdot^{\circ}\Pi_{k-2l-1m}) - \partial\cdot^{\circ}(\mathcal{D}\cdot^{\circ}\Pi_{k-2lm}). \tag{53}$$

Making use of Equation (22) and of the solvability condition (26), Equation (53) implies that δ_{2lm} vanishes identically for all l and m. This already proves that, at this order, the density n verifies indeed The Smoluchowski equation (S0.2). The correction terms in Equation (S0.3) will now be obtained by evaluating δ_{3lm} and δ_{4lm}. Considering the solvability condition (29) and the expression (24) for $^{\circ}\Pi_{1lm}$, we obtain from (53) :

$$\delta_{3lm} = -\underline{v}\Big(\partial_T\Big(P\cdot^{\circ}(\mathcal{D}\cdot^{\circ}A_{0lm})\Big) - \partial\cdot^{\circ}\Big(\mathcal{D}\cdot^{\circ}(\mathcal{D}\cdot^{\circ}A_{0lm})\Big)\Big)e^{-\frac{v^2}{2}}. \tag{54}$$

After integration over $\underline{v}$, this term provides an identically vanishing contribution to Δ. Finally, the substitution of (27) in (53) leads to :

$$\begin{aligned}\delta_{4lm} = \Big\{ & \partial_T(P\cdot^{\circ}A_{2l-1m}) - \partial\cdot^{\circ}(\mathcal{D}\cdot^{\circ}A_{2lm}) \\ & + \frac{\underline{v}^2}{2}\Big(\partial_T\Big(P\cdot^{\circ}\Big(\mathcal{D}\cdot^{\circ}(\mathcal{D}\cdot^{\circ}A_{0l-1m})\Big)\Big) \\ & -\partial\cdot^{\circ}\Big(\mathcal{D}\cdot^{\circ}\Big(\mathcal{D}\cdot^{\circ}(\mathcal{D}\cdot^{\circ}A_{0lm})\Big)\Big)\Big) \\ & + K(\underline{v})\Big\}e^{-\frac{v^2}{2}},\end{aligned} \tag{55}$$

where $K(\underline{v})$ represents a polynomial expression in $\underline{v}$ which only involves odd powers of this variable. $K(\underline{v})$ will therefore not contribute to Δ. With the help of the solvability conditions (26) and (32) and considering the commutation relations $[P;\mathcal{D}] = 0$ and $[\partial_T;\mathcal{D}] = 0$, (55) becomes, after integration over $\underline{v}$:

$$\int_{\mathbb{R}} \delta_{4lm}\, d\underline{v} = -\sqrt{2\pi}\partial\cdot^{\circ}\Big(^{\circ}[\mathcal{D};\partial]\cdot^{\circ}\Big(\mathcal{D}\cdot^{\circ}A_{0lm}\Big)\Big). \tag{56}$$

Multiplying (56) by $\epsilon^4\Big(\frac{\epsilon'}{\epsilon}\Big)^l\Big(\frac{\epsilon''}{\epsilon}\Big)^m$ and summing over l and m yields the following expression for Δ :

$$\Delta = -\sqrt{2\pi}\epsilon^4\partial\cdot^{\circ}\Big(^{\circ}[\mathcal{D};\partial]\cdot^{\circ}\Big(\sum_{l,m\in\mathbb{N}^2}\Big(\frac{\epsilon'}{\epsilon}\Big)^l\Big(\frac{\epsilon''}{\epsilon}\Big)^m\mathcal{D}\cdot^{\circ}A_{0lm}\Big)\Big) + \mathcal{O}(\epsilon^5). \tag{57}$$

According to the discussion at the end of Section IV, the spatial density is :

$$n = \int_{\mathbb{R}}\sum_{l,m\in\mathbb{N}^2}\Big(\frac{\epsilon'}{\epsilon}\Big)^l\Big(\frac{\epsilon''}{\epsilon}\Big)^m\Pi_{0lm}\, d\underline{v}. \tag{58}$$

Consequently, the summation over l and m in Equation (57) can be expressed in terms of the density n :

$$\epsilon\sqrt{2\pi}\sum_{l,m\in\mathbb{N}^2}\Big(\frac{\epsilon'}{\epsilon}\Big)^l\Big(\frac{\epsilon''}{\epsilon}\Big)^m(\mathcal{D}\cdot^{\circ}A_{0lm}) = \underline{D}n, \tag{59}$$

where the operator $\underline{D}$ is :

$$\underline{D} = \partial_{\underline{x}} - \underline{f}.$$

Substituting (59) in (57), we have :

$$\Delta = -\partial_{\underline{x}}[\underline{D};\partial_{\underline{x}}](\underline{D}n). \tag{60}$$

Using the identity $[\underline{D};\partial_{\underline{x}}] = \partial_{\underline{x}}\underline{f}$, (60) leads immediately to the expression (47) for the corrected Smoluchowski equation. We have seen in Section IV C that the Chapman-Enskog expansion for sub-family *(ii.2)* can be formally deduced from the one obtained in Section IV B for sub-family *(ii.1)* by a simple summation over l. By its very definition (51), δ involves a summation over l and therefore does also Δ. The correction terms to Smoluchowski equation for sub-family *(ii.2)* are consequently identical to those obtained in this Section for sub-family *(ii.1)*.

B. Sub-family *(ii.3)* and *(ii.4)*

Contrary to the preceding case, it is not necessary here to introduce a vectorial formalism. The physical reason is that sub-family *(ii.1)* involves two space-scales, whereas sub-family *(ii.3)* only involves one. The corresponding expression for δ in terms of Π is then :

$$\delta = \epsilon\epsilon'\partial_T\Pi - \epsilon^2\partial_{X^2}\Pi + \epsilon^2\partial_X(\Pi F). \tag{61}$$

Considering the form of the Chapman-Enskog expansion for Π introduced in Section IV D, we write for δ :

$$\delta = \sum_{k,l\in\mathbb{N}^2} \epsilon^k \left(\frac{\epsilon'}{\epsilon}\right)^l \delta_{kl}, \tag{62}$$

With the help of expressions (35) for Π and (62) for δ, (61) becomes :

$$\delta_{kl} = \partial_T\Pi_{k-2l-1} - \partial_{X^2}\Pi_{k-2l} + F\partial_X\Pi_{k-2l} + F_X\Pi_{k-2l}. \tag{63}$$

The solution of (36) leads, through (63), to an expression for Δ identical to the one obtained in the preceding section (Equation (60)). Finally, since all results obtained in Section IV E can be formally deduced from those concerning sub-family *(ii.3)* by summation over l, Equation (47) is also valid for sub-family *(ii.4)*.

C. A brief summary of the preceding results

Using expression (60) for the correction Δ, the dimensionless Smoluchowski equation with the first correction terms reads :

$$\partial_{\underline{t}}n - \partial_{\underline{x}\underline{x}}n + \partial_{\underline{x}}(n\underline{f}) + \partial_{\underline{x}}\{\partial_{\underline{x}}\underline{f}(\partial_{\underline{x}}n - \underline{f}n)\} = 0. \tag{64}$$

In terms of the original physical variables, Equation (64) can be re-written as :

$$\partial_t n - \chi\partial_{xx}n + \frac{1}{m\alpha}\partial_x(nf) + \frac{\chi}{m\alpha^2}\partial_x\{\partial_x f(\partial_x n - \frac{1}{\chi m\alpha}nf)\} = 0. \tag{65}$$

VI. DISCUSSION

A. Comparison with previous works

We would like now to discuss thoroughly the results presented in both preceding sections and compare them with those already available in the literature. Some typical recent and frequently quoted references on the topic are Refs.[3–9]. Actually, Ref.[4] proposes a systematic generalization of the work presented in[3] and discusses most of the literature before 1978 at great length. Ref.[5] is essentially based on Ref.[4]; it proposes a derivation of Smoluchowski equation only, without any correction terms, but it also contains some original very important physical discussions which render its reading essential to any proper evaluation of the issues raised by the present article. References[6–9] elaborate on a relatively different perturbation scheme, where the unperturbed phase space distribution is actually a *drifting* Maxwellian, in contradistinction to the unperturbed local equilibrium retained in the present article and Refs.[3–5]. References[6–9] will therefore be discussed at the end of this section, after having compared carefully our results with those of Titulaer[4] and van Kampen[5].

Titulaer envisages the problem of finding the correct solution to Kramers' equation (2) for given initial conditions. To this end, he considers different classes of solutions, each class being labeled by a non-negative integer n (which, naturally, must not be confused with the notation n used in the present article for the spatial density). In any of these classes[3], each solution has to be obtained by a Chapman-Enskog expansion about which more will be said later on. After having constructed or explained how to construct at least the first expansion terms for solutions belonging to each class, Titulaer argues rather convincingly that the solutions which belong to a class characterized by a strictly positive integer n will decay exponentially in time on a typical time-scale $(n\alpha)^{-1}$; this apparently remains true even for spatially homogeneous solutions in homogeneous force field. This makes clear that the solutions considered in the present article, which describe the system in the hydrodynamic limit, have to be compared only with the solutions of Ref.[4] labeled by $n = 0$.

As already alluded to before, these solutions are obtained by Titulaer (and van Kampen) by means of a Chapman-Enskog expansion which involves a single "small parameter", the inverse of the friction coefficient α. While the corresponding expansion is formally well-defined, its physical interpretation does not seem straightforward to us.

Indeed, if one wants to interpret the asymptotics presented in[4,5] by introducing the scale over which the distribution function Π varies in space, one can follow van Kampen (Ref.[5], Remark at the end of p. 218) and obtain the following inequality (recast in our notation) :

$$\frac{1}{\alpha}\left|\frac{\partial_x\,\Pi_0}{\Pi_0} - f\right| \ll 1. \tag{66}$$

This essentially states that the small parameter used by Titulaer and van Kampen may be considered as some kind of mixture of two fundamentally different quantities, which are respectively the ratio of the "mean free path" $\sqrt{\chi/\alpha}$ to the scale over which Π varies spatially and the ratio of the external force-field to the natural force-unit $m\sqrt{\alpha^3\chi}$. Moreover, both these quantities may be large and their difference may still verify (66). To our eyes, the uneasiness one might feel in trying to interpret physically (66) any further is a sign that the general asymptotics to be used in a Chapman-Enskog expansion applied to Kramers' equation does not a priori depend on a single small parameter only. Indeed, (66) alone strongly suggests that it might be wise to consider at least the two independent small parameters $\sqrt{\chi/\alpha}\partial_x\Pi/\Pi$ and $f/m\sqrt{\alpha^3\chi}$, namely ϵ and ν introduced in Section III. As it turns out from the investigation presented in that Section and from the results of Section IV, the most general collective behavior of point particles diffusing in a time independent vanishingly small force-field involves, in the hydrodynamic limit, three independent small parameters. This general situation can naturally degenerate into various two-parameters problems and even into a one-parameter case. Except in this last situation, the corresponding Chapman-Enskog expansions, presented in part in Section IV, are naturally more cumbersome than those proposed by Titulaer and van Kampen.

Let us now compare briefly our results with the work presented in Refs.[6–9]. If[6] and[7] contain very interesting original material, a most useful source for the procedure they introduced seems to us to be Refs.[8] and[9], where some mathematical and physical points are more extensively discussed. What is envisaged in these references is a Chapman-Enskog expansion around a *drifting* Maxwellian, with a drift velocity v_d related to the force f by $v_d = f/\alpha$ (in our notation). Following the procedure of Refs.[3,4], the expansion in Refs.[8,9] still involves as single small parameter, the inverse of the friction coefficient α. The possible limitations imposed by this last choice have already been discussed at great length in the preceding paragraph and will not be mentioned again. The choice of a drifting Maxwellian as local "equilibrium" around which the expansion is carried out seems to be useful in at least two potentially different (non-exclusive) situations. The first one would involve a non vanishingly small external force field. It is not obvious to us that the choice of v_d retained in Refs.[8,9] would then exhaust all physically interesting solutions. In particular, this choice seems to correspond only to an over-damped mean microscopic motion of the particle. Anyhow, the corresponding physics cannot be compared to the one presented in this article, where the force field is supposed to be vanishingly small, as in Refs.[3–5]. According to Ref.[9], the choice of a drifting Maxwellian may also be useful in describing transient regimes of at least some solutions of Kramers' equation. From this point of view, it might be interesting to compare the solutions presented in Ref.[9] to those of families *(iii.1)* and *(iii.3)* of the present article. As was already discussed before, this cannot be done in the framework of the Chapman-Enskog formalism, and lies therefore outside the scope of this work.

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[3]Each class is actually associated to an eigenvalue of the differential operator which appears, *e.g.*, in the right-hand side of Equation (16); n labels the various eigenvalues of this operator.

B. An interpretation of the various sub-families

The aim we shared with Titulaer[4] in performing these voluminous expansions was to derive from them (for a sufficiently small force field) the Smoluchowski equation and at least the first correction terms to it. The fact that Equation (47) formally agrees with the one obtained by Titulaer (Ref.[4]) hides that our derivation is more general than his, and that Equation (47) is also valid in various physically different situations not investigated in Reference[4] (see in particular Equation (66) and the discussion thereafter). The physical meaning of Expression (60) for Δ can be investigated by evaluating the order of magnitude of the various terms which appear in Equation (65) for the four different sub-families *(ii.1)* to *(ii.4)*. The results are gathered in Table III. This table also indicates the order of magnitude of the same terms for family *(i)* and *(iii)*, because preliminary calculations indicate that (47) is indeed valid for all three families. We will thus be able to discuss them together via (47) only.

This clearly reveals that, although formally identical for the three families, Equation (65) actually describes substantially different Physics in each case.

There are two main groups of sub-families. The first one (Group A) encompasses the sub-families (see Table IV) for which the spatial variations of the density $n(\underline{x},\underline{t})$ occur on much smaller scales than those of the external force field, which can therefore be considered as nearly homogeneous ($\epsilon'' < \epsilon$). The other group (Group B) contains the sub-families for which the density $n(\underline{x},\underline{t})$ and the external force vary on the same spatial scale ($\epsilon'' = \epsilon$).

Each sub-family will be discussed with the help of Table III, retaining in each case the dominant terms in Equation (64).

1. Free diffusion regime

Sub-families *(i.1)* in Group A and *(i.3)* in Group B represent cases where the magnitude of the force is so small that (64) practically degenerates into the usual diffusion equation:

$$\partial_{\underline{t}} n - \partial_{\underline{x}\underline{x}} n = 0. \tag{67}$$

This means that these regimes essentially describe free diffusion phenomena.

2. Barostatic regime

For sub-families *(ii.1)* in Group A and *(ii.3)* in Group B, Equation (64) degenerates respectively into:

$$-\partial_{\underline{x}\underline{x}} n + \underline{f}\,\partial_{\underline{x}}(n) = 0, \tag{68}$$

and

$$-\partial_{\underline{x}\underline{x}} n + \partial_{\underline{x}}(n\underline{f}) = 0. \tag{69}$$

Quite logically, Equation (68) is essentially identical to (69) specialized to a situation where the force field is nearly homogeneous. Equation (69) admits as solution:

$$n(\underline{x},\underline{t}) = n_0 \exp\big(-\Phi(x)\big),$$

where $\Phi(x) = -\int^x f(y)dy$ is the potential associated to f, and n_0 normalizes n to unity. We therefore call this regime "barostatic".

3. Over-damped regime

For sub-families *(iii.1)* in Group A and *(iii.3)* in Group B, Equation (64) degenerates respectively into

$$\partial_{\underline{t}} n + \underline{f}\,\partial_{\underline{x}}(n) = 0, \tag{70}$$

and

$$\partial_{\underline{t}} n + \partial_{\underline{x}}(n\underline{f}) = 0. \tag{71}$$

As before, (70) can be considered as a special case of (71) for nearly uniform force fields. Mathematically, Equation (71) can be obtained from the microscopic equation of motion (1) by neglecting $\frac{d}{dt}\mathbf{v}$. The mean motion of the particle is then an over-damped motion in the force field $\mathbf{f}$.

4. Driven diffusion regime

For sub-family *(–.4)* in Group B, Equation (64) essentially becomes the standard Smoluchowski equation :

$$\partial_{\underline{t}} n - \partial_{\underline{x}\underline{x}} n + \partial_{\underline{x}} (\underline{f} n) = 0, \tag{72}$$

and sub-family *(–.2)* in Group A is characterized by a nearly homogeneous force field version of (72) :

$$\partial_{\underline{t}} n - \partial_{\underline{x}\underline{x}} n + \underline{f} \partial_{\underline{x}} (n) = 0. \tag{73}$$

These equations obviously describe regimes where both diffusion and forcing effects are of comparable importance. This justify the terminology "driven diffusion".

VII. CONCLUSION

We have proposed a fresh investigation of the collective motion of point particles which diffuse stochastically under the influence of a time independent force-field. In the hydrodynamic limit, there are three different families of solutions susceptible of a treatment by the Chapman and Enskog expansion method. Each of these families subdivides into four sub-families characterized by the nature and number of the small parameters involved in the expansion. More precisely, each family contains a class of three (small) parameters solutions, two different classes of two parameters solutions and a single class of one parameter solutions which is actually common to the three main families. We have presented in full the Chapman-Enskog expansions corresponding to the four sub-cases for one of the three main families. We have then derived from our results, for each sub-case, the Smoluchowski equation and the first correction terms to it. These are actually identical for each of the studied sub-families. We have also discussed at great length the physical significance of our results and compared them with the ones already existing in the literature.

Various extensions of this work are possible and are currently the object of active investigation. One should first of all perform the Chapman-Enskog expansions for all sub-families corresponding to the two main families which were not dealt with in Section IV. Preliminary results indicate that these expansions also lead to Smoluchowski equation and to the same correction terms as those derived in this article. Considering the apparent "genericity" of these terms as well as the natural emergence of covariant derivatives (D and $\mathcal{D}$) and commutators in the Chapman-Enskog expansions, it seems to us quite possible that an elegant geometric structure exists behind all the results derived in this paper. A better understanding of the problem in this direction should prove most enlightening. This could be linked with an extension of our results to the case of a time-dependent force-field or to the General-Relativistic realm with the help of the Relativistic Ornstein- Uhlenbeck Process introduced in[10].

As already noted, the Chapman-Enskog method does not permit an investigation of the possible transient nature and dynamical stability of the various regimes envisaged in this article. This could be at least partially achieved by direct numerical simulations.

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TABLE I. *The definition of all the infinitesimal parameters introduced in this article are recalled here.*

Quantity	$\partial_{\underline{x}}\Pi/\Pi$	$\partial_{\underline{t}}\Pi/\Pi$	$n< v^{2k+1} >$	f	$\partial_{\underline{x}}f/f$
Order	$\mathcal{O}(\epsilon)$	$\mathcal{O}(\eta)$	$\mathcal{O}(\epsilon')$	$\mathcal{O}(\nu)$	$\mathcal{O}(\epsilon'')$

TABLE II. *The characteristics of the three families of solutions and of the four sub-families are summed up here. Each column of this table corresponds to a family, and each line to a sub-family.*

	(i)	*(ii)*	*(iii)*
(−.1)	$\epsilon' = \epsilon, \quad \nu < \epsilon, \quad \epsilon'' < \epsilon$	$\nu = \epsilon, \quad \epsilon' < \epsilon, \quad \epsilon'' < \epsilon$	$\nu = \epsilon', \quad \epsilon < \epsilon', \quad \epsilon'' < \epsilon$
(−.2)	$\nu = \epsilon, \quad \epsilon' = \epsilon, \quad \epsilon'' < \epsilon$		
(−.3)	$\epsilon' = \epsilon, \quad \nu < \epsilon, \quad \epsilon'' = \epsilon$	$\nu = \epsilon, \quad \epsilon' < \epsilon, \quad \epsilon'' = \epsilon$	$\nu = \epsilon', \quad \epsilon < \epsilon', \quad \epsilon'' = \epsilon$
(−.4)	$\nu = \epsilon', \quad \epsilon = \epsilon', \quad \epsilon'' = \epsilon$		

TABLE III. *This table gives the orders of magnitude of all the terms in the corrected Smoluchowski equation for all the sub-families. Each column corresponds to a sub-family, each line to a term.*

Term	*(i.1)*	*(ii.1)*	*(iii.1)*	*(−.2)*	*(i.3)*	*(ii.3)*	*(iii.3)*	*(−.4)*
$\partial_{\underline{t}}n$	ϵ^2	$\epsilon'\nu$	$\epsilon\nu$	ν^2	ϵ''^2	$\epsilon'\nu$	$\nu\epsilon''$	ν^2
$\partial_{\underline{xx}}n$	ϵ^2	ν^2	ϵ^2	ν^2	ϵ''^2	ν^2	ϵ''^2	ν^2
$\underline{f}\partial_{\underline{x}}n$	$\epsilon\nu$	ν^2	$\epsilon\nu$	ν^2	$\nu\epsilon''$	ν^2	$\nu\epsilon''$	ν^2
$n\partial_{\underline{x}}\,\underline{f}$	$\nu\epsilon''$	$\nu\epsilon''$	$\nu\epsilon''$	$\nu\epsilon''$	$\nu\epsilon''$	ν^2	$\nu\epsilon''$	ν^2
$\partial_{\underline{xx}}\,\underline{f}\partial_{\underline{x}}n$	$\epsilon\nu\epsilon''^2$	$\nu^2\epsilon''^2$	$\epsilon\nu\epsilon''^2$	$\nu^2\epsilon''^2$	$\nu\epsilon''^3$	ν^4	$\nu\epsilon''^3$	ν^4
$n\underline{f}\partial_{\underline{xx}}\,\underline{f}$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	ν^4	$\nu^2\epsilon''^2$	ν^4
$\partial_{\underline{x}}\,\underline{f}\partial_{\underline{xx}}n$	$\epsilon^2\nu\epsilon''$	$\nu^3\epsilon''$	$\epsilon^2\nu\epsilon''$	$\nu^3\epsilon''$	$\nu\epsilon''^3$	ν^4	$\nu\epsilon''^3$	ν^4
$n(\partial_{\underline{x}}\,\underline{f})^2$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	$\nu^2\epsilon''^2$	ν^4	$\nu^2\epsilon''^2$	ν^4
$\underline{f}\partial_{\underline{x}}\,\underline{f}\partial_{\underline{x}}n$	$\epsilon\nu^2\epsilon''$	$\nu^3\epsilon''$	$\epsilon\nu^2\epsilon''$	$\nu^3\epsilon''$	$\nu^2\epsilon''^2$	ν^5	$\nu^2\epsilon''^2$	ν^5

TABLE IV. *This table sums up the physical discussion presented in Section VI B. Each row corresponds to one of the four physical regimes; Columns one and two help distinguish between cases where the force can be considered nearly homogeneous or not.*

	quasi-homogeneous force Group A. ($\epsilon'' < \epsilon$)	heterogeneous force Group B. ($\epsilon'' = \epsilon$)
Free diffusion regime ($\epsilon = \epsilon'$; $\epsilon > \nu$)	Family *(i.1)* $\partial_{\underline{t}} n - \partial_{\underline{xx}} n = 0$	Family *(i.3)* $\partial_{\underline{t}} n - \partial_{\underline{xx}} n = 0$
Barostatic regime ($\epsilon' < \epsilon$; $\epsilon = \nu$)	Family *(ii.1)* $-\partial_{\underline{xx}} n + \underline{f} \partial_{\underline{x}}(n) = 0$	Family *(ii.3)* $-\partial_{\underline{xx}} n + \partial_{\underline{x}}(n \underline{f}) = 0$
Over-damped regime ($\epsilon < \epsilon'$; $\epsilon' = \nu$)	Family *(iii.1)* $\partial_{\underline{t}} n + \underline{f} \partial_{\underline{x}}(n) = 0$	Family *(iii.3)* $\partial_{\underline{t}} n + \partial_{\underline{x}}(n \underline{f}) = 0$
Driven diffusion regime ($\epsilon' = \epsilon$; $\epsilon = \nu$)	Family *(–.2)* $\partial_{\underline{t}} n - \partial_{\underline{xx}} n + \underline{f} \partial_{\underline{x}}(n) = 0$	Family *(–.4)* $\partial_{\underline{t}} n - \partial_{\underline{xx}} n + \partial_{\underline{x}}(n \underline{f}) = 0$