A Generalized Smoluchowsky Equation: 
The Hydrodynamical and Thermodynamical 
Picture of Brownian Motion

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Abstract

We present a systematic expansion of Kramers equation in the high friction limit. The latter is expanded within an operator continued fraction scheme. The relevant operators include both temporal and spatial derivatives and a covariant derivative or gauge like operator associated to the potential energy. Trivially, the first order term yields the Smoluchowsky equation. The second order term is readily obtained, known as the corrected Smoluchowsky equation. Further terms are computed in compact and straightforward fashion. As an application, the nonequilibrium thermodynamics and hydrodynamical schemes for the one dimensional Brownian motion is presented.

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I. Introduction

Kramers equation [1] (although originally proposed by Klein [2]) and the associated Smoluchowsky equation [3] have been widely applied to diverse problems, namely: Brownian motion in potential wells, chemical reactions rate theory, nuclear dynamics, general stochastic processes, just to mention a few. A relevant overview (by no means complete) can be found in the review articles [4]-[7], books [8]-[11] and references therein. The passage from Kramers to Smoluchowsky equation was informal and qualitative in the early days (see for example [1] and [4]). Only fairly recently a well founded relationship was established [12]-[22]. A multiple time scale approach has been put forward more recently [23],[24].

In section II we present a differential recursive expansion for Kramers equation, with respect to the inverse of the friction coefficient, within a continued fraction formalism [23]-[27]. In a compact and straightforward fashion we retrieve the original and the corrected Smoluchowsky equation [4],[10], in first and third order respectively. Higher order corrections are eased out with reasonable algebra, amenable to a diagrammatic framework. In section III some applications are presented, namely the hydrodynamical and non-equilibrium thermodynamical picture associated to the Brownian motion. Finally, in section IV concluding remarks include further applications and future work.

II. Continued fraction expansion

Kramers [1] considers the probability distribution $P(x,v,t)$ in phase space for the Brownian particle (mass $m$) in an external potential $V(x,t)$. The friction coefficient is $\gamma$, $D$ the diffusion coefficient and $T$ the bath temperature. Kramers equation (in the one dimensional case to simplify our analysis) is given by

$$\left( \frac{\partial}{\partial t} + v \frac{\partial}{\partial x} - \frac{1}{m} \frac{\partial V}{\partial x} \frac{\partial}{\partial v} \right) P = \gamma \frac{\partial}{\partial v} \left( v + \gamma D \frac{\partial}{\partial v} \right) P$$

(1)
If $l$ denotes a characteristic length scale (associated to $V$ for instance) and with Einstein relation $D = \zeta k_B T$ \[^{[28]}\] ($\zeta$ is the mobility coefficient, $\zeta^{-1} = m\gamma$), we introduce dimensionless variables \[^{[24]}\]

$$
\tau = \frac{t}{l} \sqrt{\frac{k_B T}{m}}, \quad q = \frac{x}{l}, \quad \xi = \frac{v}{\sqrt{2k_B T}}, \quad A(q, \tau) = \frac{V(x, t)}{k_B T}
$$

and a dimensionless friction coefficient inverse $\varepsilon$ given by

$$
\varepsilon = \frac{1}{\gamma l} \sqrt{\frac{k_B T}{m}} = \frac{1}{\gamma v_T} l
$$

Thus, Kramers equation is cast as

$$
\partial \frac{\partial}{\partial \xi} \left( \xi + \frac{1}{2} \frac{\partial}{\partial \xi} \right) P(q, \xi, \tau) = \varepsilon \left( \frac{\partial}{\partial \tau} + \sqrt{2} \xi \frac{\partial}{\partial q} - \frac{1}{\sqrt{2}} \frac{\partial A}{\partial q} \frac{\partial}{\partial \xi} \right) P(q, \xi, \tau)
$$

(2)

Following \[^{[12]}\] we use the ansatz

$$
P(q, \xi, \tau) = C \exp \left( -\frac{1}{2} \xi^2 \right) \sum_{n=-\infty}^{\infty} \Psi_n(q, \tau) \Phi_n(\xi)
$$

(3)

where the $\Phi_n(\xi)$’s are the orthonormal Hermite functions \[^{[29]}\] with $\Psi_n(q, \tau) \equiv \Phi_n(\xi) \equiv 0$ for $n < 0$ and $C$ is an overall constant. Substitution yields a set of differential recursive (difference) equations for the $\Psi_n$’s. Introducing the compact notation

$$
\partial = \frac{\partial}{\partial \tau}; \quad \nabla = \frac{\partial}{\partial q} \quad D = \nabla + (\nabla A)
$$

we obtain the recursion relation \[^{[29]}\]

$$
(\varepsilon \partial + n) \Psi_n(q, \tau) = -\varepsilon \sqrt{n} D \Psi_{n-1}(q, \tau) - \varepsilon \sqrt{n + 1} \nabla \Psi_{n+1}(q, \tau)
$$

(4)

This recursion relation can be readily generalized to higher dimensions via the tensor Hermite polynomials \[^{[30]}\]. The first relation obtained from equation (4) with $n = 0$ is a continuity equation, defined here as the generalized Smoluchowsky equation.

$$
\partial \Psi_0(q, \tau) + \nabla \Psi_1(q, \tau) = 0
$$

(5)
For the $\Psi_n(x, \tau)$ we may define an associated function $\Psi_n(x, s)$ via the transformation

$$\Psi_n(q, \tau) = \int_{\Gamma} ds \Psi_n(q, s) \exp(-s \tau) \quad (6)$$

Both the integration path $\Gamma$ and particulars of the $s$ variable are to be defined adequately (Laplace or Fourier like), assuring among other things that the expansion

$$G_n = -\varepsilon_n \left(1 + \frac{\varepsilon_n}{n} \partial\right)^{-1} = \sum_{k=0}^{\infty} \left(-\frac{\varepsilon_n}{n}\right)^{1+k} \partial^k \quad n > 0 \quad (7)$$

is a well defined expression. Here we do not require the transformed functions, as in previous works [13]-[22]. Furthermore we define

$$\alpha_n = \sqrt{nD}G_n, \quad \beta_n = \sqrt{n+1} \nabla G_n$$

yielding a compact form for the recursion relation (4)

$$\Psi_n(q, \tau) = \alpha_n \Psi_{n-1}(q, \tau) + \beta_n \Psi_{n+1}(q, \tau)$$

With standard recursive relations and continued fractions techniques ([25]-[27]) the above differential recursion relation can be solved as

$$\Psi_n(q, \tau) = \mathcal{L}_n(\partial, \nabla, D) \Psi_{n-1}(q, \tau) \quad n > 0$$

$$\partial\Psi_0(q, \tau) = -\nabla \Psi_1(q, \tau) = -\nabla \mathcal{L}_1 \Psi_0(q, \tau) \quad (8)$$

$$\mathcal{L}_n(\partial, \nabla, D) = (1 - \alpha_n \mathcal{L}_{n+1}(\partial, \nabla, D))^{-1} \beta_n$$

The last equation is an infinite continued fraction differential operator

$$\mathcal{L}_n(\partial, \nabla, D) = \left(1 - \alpha_n \left(1 - \alpha_{n+1} \left(1 - \alpha_{n+2} \left(1 - \cdots \right)^{-1} \beta_{n+3} \right)^{-1} \beta_{n+2} \right)^{-1} \beta_{n+1} \right)^{-1} \beta_n$$

This equation can be expanded into a sum of separable products of the kind
\[
\mathcal{L}_n(\partial, \nabla, D) = \sum_{l=1}^{\infty} B_{n,l}(\nabla, D) R_{n,l}(\partial), \quad R_{n,l}(\partial) = \sum_{\alpha_1, \ldots, \alpha_l} C_{n,l}^{\alpha_1 \cdots \alpha_l} G_{\alpha_1} \cdots G_{\alpha_l} \tag{9}
\]

On the other hand, from equations (5), (7) and (8) we obtain the identity

\[
G_{\alpha} \Psi_0 = -\frac{\varepsilon}{\alpha} \left( 1 - \frac{\varepsilon}{\alpha} \nabla \mathcal{L}_1 \right)^{-1} \Psi_0 \tag{10}
\]

Let us write the ansatz (an \(\varepsilon\) expansion in purely spatial operators)

\[
\mathcal{L}_n(\nabla, D) = \sum_{l=1}^{\infty} Z_{n,l}(\nabla, D) \varepsilon^l \tag{11}
\]

With this ansatz for \(n = 1\) we compute (10), then (9), obtaining an iterative relation for the \(Z_{1,l}\). Once the latter are computed we are able to compute all the \(\mathcal{L}_n\)'s in terms of purely spatial operators. We present the expansion results up to \(\varepsilon^5\) for the first five \(\Psi_n\)’s

\[
\begin{align*}
\Psi_1 &= -\varepsilon \left( 1 + \varepsilon^2 \Gamma_3 + \varepsilon^4 \Gamma_5 \right) D \Psi_0 \\
\sqrt{2} \Psi_2 &= \varepsilon^2 \left( D + \varepsilon^2 D \left( \Gamma_3 - \frac{1}{2} \nabla D \right) + \frac{1}{2} \varepsilon^3 \nabla D^3 \right) D \Psi_0 \\
\sqrt{3} \Psi_3 &= -\varepsilon^3 D \left( D + \varepsilon^2 D \left( \Gamma_3 - \frac{1}{2} \nabla D \right) \right) D \Psi_0 \\
\sqrt{4} \Psi_4 &= \varepsilon^4 D^4 \Psi_0, \quad \sqrt{5} \Psi_5 = -\varepsilon^5 D^5 \Psi_0
\end{align*}
\]

where

\[
\Gamma_3 = \nabla^2 A, \quad \Gamma_5 = 2 \left( \nabla^2 A \right)^2 + \frac{1}{2} \nabla^4 A + (\nabla^3 A) \nabla A + \frac{3}{2} (\nabla^3 A) \nabla
\]

The generalized Smoluchowsky equation, from equation (9) and up to \(\varepsilon^5\) terms is therefore

\[
\partial \Psi_0 = \varepsilon \nabla \left( \mathcal{L}_0 + \varepsilon^2 \Gamma_3 + \varepsilon^4 \Gamma_5 \right) D \Psi_0 \tag{12}
\]
This expression coincides with previous results, see for example [16]. We believe to have presented a simpler and compact version for the recursion relations, amenable to further diagrammatic manipulations. Little attention has hitherto been paid to the evolution of moments higher than $\Psi_0$. Within an effective medium scheme [31] we assume $\Psi_0$ to be known via the solution of (12) and thus compute the higher moments $\Psi_n$ ($n > 0$) in terms of $\Psi_0$. In this fashion we explore both formal and practical consequences (the usual scheme is to solve the recursion relations in a hierarchical form, $\Psi_n$ in terms of $\Psi_{n+1}$).

III. Applications

We show that the recursive relations (4) correspond to hydrodynamic balance equations derived from the Kramers kinetic equation (for equations other than Kramers see [30]-[33]). We may picture the Brownian motion problem as a one dimensional fluid. Restoring to the original (dimensional) variables, let us write some macroscopic densities (moments) of interest [30], namely: the mass $\rho$ (particle $n$), the momentum flow $J_m$ (velocity density $J$ and mean velocity $u$), the hydrodynamic pressure $\Pi$, the kinetic energy $E$ and the heat flow $J_E$, given respectively by (hereafter we denote $P(x,v,t) = P(q,\xi,\tau)$, given by (3))

\[
\begin{align*}
\rho(x,t) & = m \int dv P(x,v,t) = mn(x,t) \\
J_m(x,t) & = m \int dvvP(x,v,t) = mJ(x,t) \\
& = \rho(x,t)u(x,t), \quad u(x,t) = \frac{\int_{-\infty}^{\infty} dvvP(x,v,t)}{\int_{-\infty}^{\infty} dvP(x,v,t)} \\
\Pi(x,t) & = m \int dvv^2P(x,v,t) = 2E(x,t) \\
J_E(x,t) & = \frac{1}{2} m \int dvv^3P(x,v,t)
\end{align*}
\]
The overall constant $C$ in equation (3) can be adjusted such that we have the correspondence $\rho(x, t) = \Psi_0(x, t)$ (4), yielding $C^{-2} = 2\sqrt{\pi}mk_BT$. From the properties of the Hermite functions [29] we have

\[
\begin{align*}
\rho(x, t) &= \Psi_0(x, t) = mn(x, t) \\
J_m(x, t) &= v_T\Psi_1(x, t) = mn(x, t)u(x, t) \\
E(x, t) &= \frac{1}{2}\Pi(x, t) = \frac{1}{2}v_T^2\left(\Psi_0(x, t) + \sqrt{2}\Psi_2(x, t)\right) \\
J_E(x, t) &= \frac{1}{\sqrt{2}}v_T^3\left(\frac{3}{2}\Psi_1(x, t) + \sqrt{3}\Psi_3(x, t)\right)
\end{align*}
\]

From the recursion relation (4), we obtain

\[
\begin{align*}
\frac{\partial \rho}{\partial t} &= -\frac{\partial J_m}{\partial x} = -\frac{\partial \rho u}{\partial x} \\
\frac{\partial J_m}{\partial t} &= -\frac{\partial \Pi}{\partial x} + K\rho - \gamma J_m, \quad K(x) = -\frac{1}{m}\frac{\partial V}{\partial x} \\
\frac{\partial E}{\partial t} &= -\frac{\partial J_E}{\partial x} + KJ_m - 2\gamma(E - E_0), \quad E_0 = \frac{1}{2}v_T^2\rho
\end{align*}
\]

These are balance equations for the mass, momentum and kinetic energy density respectively. The RHS of these equations are the diffusive (gradient), convective (proportional to the external force) and dissipative terms, respectively. The first equation is the continuity (generalized Smoluchowsky) equation, the only one having a conserving character (no dissipation). There is a convective (drift) term hidden in $\Psi_1$. The dissipative terms appear in a relaxation time form. The inverse relaxation times are $0, \gamma, 2\gamma, ... n\gamma$ (as we compute higher order terms). Here we connect to the recent multiple time scale analysis mentioned in the introduction [23,24]. Our formalism can be readily generalized for multiple components undergoing chemical reactions, provided the reaction terms are cast in the linear approximation. This is the case for inhomogeneous semiconductors in the semiclassical approximation [35]-[36].

We write down the Boltzmann entropy density (hereafter $k_B = 1$)
\[ \sigma(x, t) = -\int dv P(x, v, t) \ln \frac{h}{me} P(x, v, t) = \int dv \sigma(x, v, t) \]

with \( \ln e = 1 \). This entropy density is slightly different to the interpolative entropy defined in [4]. The equilibrium state is defined as the stationary state \( \partial \Psi_{eq}^n \equiv 0, n = 0, 1, 2 \cdots \), where all flows have ceased, \( i.e. \ \Psi_{eq}^n \equiv 0, n = 1, 2, \cdots \). In this context the equilibrium state is properly defined by \( D \Psi_{eq}^0 = 0 \), and we readily obtain (see [32] for a one dimensional ideal gas)

\[ \sigma_{eq}(n_{eq}(x), T) = n_{eq}(x) \left( \frac{3}{2} - \ln \frac{n_{eq}(x)}{n_{Q}(T)} \right) \quad n_{Q}(T) = \sqrt{\frac{2\pi m T}{h^2}} \]

Some nonequilibrium stationary states (ss) may exist and will be reported elsewhere.

With the above defined entropy, our hydrodynamical picture allow us define the local nonequilibrium densities for the energy \( E \), free energies \( F \) and \( G \); the kinetic temperature \( \theta \) and the internal chemical potential \( \mu_{int} \) as

\[
\begin{align*}
E(x, t) &= \frac{1}{2} \theta(x, t) n(x, t) \\
F(x, t) &= E(x, t) - \theta(x, t) \sigma(x, t) \\
G(x, t) &= F(x, t) + \Pi(x, t) = \mu_{int}(x, t) n(x, t)
\end{align*}
\]

These definitions do not assume the local equilibrium assumption [33, 34] and will prove to be consistent. The total chemical potential is \( \mu = \mu_{int} + V(x) \) [32]. From our hydrodynamical results, and hereafter to second order in \( \varepsilon \), we obtain for the kinetic temperature and the particle flow density

\[
\begin{align*}
\theta(x, t) &= T \left( 1 + \varepsilon^2 \varepsilon^2 \frac{D^2 n(x, t)}{n(x, t)} \right) \\
J(x, t) &= -\varepsilon v_T D n(x, t)
\end{align*}
\]

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We estimate the temperature fluctuations $|\theta - T|/T$ for constant external fields, for two cases at room temperature. First, for an Aluminum Brownian particle (radius $\sim 10^{-3}$ mm) in water, the constant force being gravity. The temperature fluctuation is of the order of $10^{-6}$. Second, for a typical semiconductor under an external electric field $\sim 10^{5}$ Vcm$^{-1}$, temperature fluctuations are of the order $10^{-5}$. In the latter case, temperature fluctuations reach a few percents of the bath temperature only for extremely large fields of the order of the breakdown field $\sim 10^{5}$ Vcm$^{-1}$ where the semiclassical picture may fail for a typical semiconductor.

The entropy is given by

$$\sigma(x, t) = \sigma_{eq}(n(x, t), \theta(x, t)) - \frac{\varepsilon^2}{2n(x, t)} \left| \left( \frac{\partial}{\partial x} + A(x, t) \right) n(x, t) \right|^2$$

where we have separated the local equilibrium contribution from the gradient corrections term, the latter with a Ginzburg-Landau reminiscence, here with the covariant derivative (the external force is the affine connection). Indeed, the local equilibrium approximation is valid up to linear corrections in $\varepsilon$. The total chemical potential is given by

$$\mu(x, t) = \varphi(x, t) + \frac{\varepsilon^2}{2n^2(x, t)} \left( \frac{\partial}{\partial x} + A(x, t) \right) n(x, t) \right|^2$$

$$\varphi(x, t) = \theta(x, t) \ln \frac{n^*(x, t)}{n_Q(T)} \quad n^*(x, t) = n(x, t) \exp \left( \frac{V(x)}{\theta(x, t)} \right)$$

As expected for the total chemical potential

$$J(x, t) = -\zeta n(x, t) \frac{\partial \mu(x, t)}{\partial x}$$

Finally, we multiply (1) by the integrating factor $1 + \ln \left( \frac{\rho}{n e P(x, v, t)} \right)$ and integrate over the velocity. We obtain the entropy balance equation (integrating by parts, we assume $v^n P(x, v, t) \to 0$, $v \to \pm \infty$).
\[
\frac{\partial \sigma(x, t)}{\partial t} + \frac{\partial J_\sigma(x, t)}{\partial x} = P_\sigma(x, t)
\]

The computed entropy flow density \( J_\sigma(x, t) \) (associated to entropy exchange with the bath) and entropy production \( P_\sigma(x, t) \) (associated to internal entropy production) are given respectively by

\[
J_\sigma(x, t) = \int dv \sigma(x, v, t) = \sigma(x, t)J(x, t)
\]

\[
P_\sigma(x, t) = \gamma \int dv P(x, v, t) \left( 1 - \gamma \mathbb{D} \left( \frac{\partial \ln P(x, v, t)}{\partial v} \right)^2 \right)
\]

\[
= \frac{2\gamma}{T} (E(x, t) - E_0(x, t)) + 2\gamma (\sigma(x, t) - \sigma_{\text{eq}}(x, t))
\]

The entropy production density is related to both an energy and entropy gradient ‘excess’ in a relaxation time fashion.

From Smoluchowsky’s equation (12) we also have the identity

\[
P_\sigma(x, t) = \frac{\partial n(x, t)}{\partial t} + J(x, t) \frac{\partial \ln n(x, t)}{\partial x}
\]

**IV. Concluding remarks**

The recursive method presented is compact and amenable to diagrammatic techniques (work in progress, as an \( \varepsilon \) expansion). We did show that the moment (\( \Psi \)) recursion relations are nothing but the hydrodynamical balance equations. Therefore we can readily obtain the diffusive, convective and dissipative contributions for any relevant quantity such as particle number, momentum and energy density.

Furthermore, we presented a nonequilibrium thermodynamical picture of Brownian motion, hinging on the knowledge of the fundamental moment, namely the density \( \rho \), calculated via the generalized Smoluchowsky equation.
With the introduction of a generalized entropy we derived the nonequilibrium thermodynamic density functions, the kinetic temperature and the chemical potential. We obtain gradient corrections to the local equilibrium approximation. We presented some relations among the computed quantities, namely: particle flow to chemical potential and entropy gradient corrections to entropy production.

Work in progress will probe and renormalize the thermodynamics presented here, as higher order terms in the $\varepsilon$ expansions are considered. Other interesting applications are related to nonisothermal oscillatory chemical reactions [39, 40]

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