# A STOCHASTIC APPROACH TO THERMODIFFUSION

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# ABSTRACT

In presence of thermal inhomogeneities, particle diffusion is known to occur even without concentration gradients. This phenomenon, known as "thermodiffusion" or "Ludwig-Soret effect" is well documented since the nineteenth century, and has numerous current applications which range from astrophysics to polymer physics and chemistry. In this contribution, we propose new microscopic stochastic models of thermodiffusion. These models are variants of the Ornstein-Uhlenbeck process; they remedy problems encountered with earlier models and their large scale behaviour coincides with the predictions of standard continuous media theories.

### 1. INTRODUCTION

Stochastic process theory is one of the most popular tools used in modelling time-asymmetric phenomena, with applications as diverse as economics (Shreve, 2004a,b), traffic management (Schreckenberg et al., 1995; Mitra and Wang, 2005), biology (Goel and Richter-Dyn, 2004; Allen, 2003), physics and chemistry (van Kampen, 2007) and cosmology (Chevalier and Debbasch, 2007). Stochastic models of thermodiffusion (Landau and Lifschitz, 1987) have been proposed in the late 80's (van Kampen, 1988; Widder and Titulaer, 1989) and have been recently reconsidered in (Bringuier and Bourdon, 2007). These are based on generalizations of the Langevin equation and model the microscopic motion of a colloidal particle undergoing Brownian motion in a fluid with non-uniform temperature field. The most elaborate description (Widder and Titulaer, 1989; Bringuier and Bourdon, 2007) assumes that the colloidal particle diffuses under the influence of three forces. The first one is the thermophoresis force, independent of the particle velocity and directly proportional to the temperature gradient. The second force is a viscous fluid force and the third one is stochastic, proportional to a Gaussian white noise. The friction and noise coefficients are allowed to depend on position through the temperature field and they obey a local fluctuation-dissipation relation. An expansion in the inverse friction coefficient then predicts that the particle current contains a contribution linear in the temperature gradient, as in traditional maccroscopic models of thermodiffusion.

Observations of thermodiffusion reveal however that these models are not fully realistic. For example, these models predict that Soret coefficients can only take positive values, whereas positive and negative values have been measured experimentally (Lenglet, Bourdon, Bacri and Demouchy, 2002; Demouchy et al, 2004; Wittko and Köhler, 2005; Ning and Wiegand, 2006). Also, the order of magnitude of observed coefficients (Bringuier and Bourdon, 2007) often differs substantially from theoretical predictions based on (Widder and Titulaer, 1989). Explanations for these discrepancies have been proposed (Bringuier and Bourdon, 2007) but an important and simple one seems to have been overlooked. In a non uniform temperature field, friction and noise coefficients generally depend on position, not only through the temperature field, but also through its derivatives. For small enough temperature inhomogeneities, the friction and noise coefficients thus contain contributions directly proportional to the temperature gradient. These terms, which are a priori of the same order of magnitude as the thermophoresis force, have until now been ignored in the literature.

This contribution remedies the problem by presenting new stochastic models of thermodiffusion. These are fully consistent and take into account all first order corrections induced by temperature gradients. The traditional macroscopic description of thermodiffusion is recovered from these new models by a Chapman-Enskog expansion. The new stochastic models are much more flexible than the previous ones (Widder and Titulaer, 1989) and allow for example, for positive as well as negative values of Soret coefficients.

# 2. GENERAL STOCHASTIC MODELS

Let us consider a Brownian<sup>1</sup> particle undergoing 1D motion with position x(t) and momentum p(t) and diffusing through its interactions with a surrounding medium. We suppose that the medium is "isotropic" and globally at rest in the chosen reference frame. Let  $\theta(x)$  be its in-

<sup>&</sup>lt;sup>1</sup>A particle whose size is much larger than the microscopic scale of the surrounding medium, but much smaller than any macroscopically relevant scale.

homogeneous temperature field. We focus on generalized Ornstein-Uhlenbeck models of the form :

$$\begin{cases} dx = \frac{p}{m}dt \\ dp = -\kappa(x,p)\frac{d\theta}{dx}dt - \alpha^{(S)}(x,p)p\,dt \qquad (1) \\ +\sqrt{2D(x,p)}dB_t. \end{cases}$$

Here,  $B_t$  is a standard Brownian motion and the superscript (s) indicates that these equations are to be understood in the Stratonovich sense (Øksendal, 1998). The term proportional to the temperature gradient is a generalized thermophoresis force. It coincides with the traditional velocity-independent thermophoresis force if  $\kappa$  is a constant. The Stratonovich process (1) is identical in law with the following Klimontovich process:

$$\begin{cases} dx = \frac{p}{m}dt \\ dp = -\kappa(x,p)\frac{d\theta}{dx}dt - \alpha(x,p)p\,dt \\ +\sqrt{2D(x,p)}dB_t, \end{cases}$$
(2)

with  $\alpha(x,p) = \alpha^{(S)}(x,p) + \frac{1}{2p}\partial_p D(x,p)$ . The density f(t,x,p) of the process with respect to the phase-space measure dxdp obeys the forward Kolmogorov equation:

$$\partial_t f + \partial_x \left(\frac{p}{m}f\right) = \partial_p \left(\kappa(x,p)\frac{d\theta}{dx}f\right) + Lf,\qquad(3)$$

$$Lf = \partial_p \Big( \alpha(x, p) \, p \, f + D(x, p) \partial_p \, f \Big). \tag{4}$$

The spatial density n(t, x) of the diffusing particle is defined by:

$$n(t,x) = \int_{\mathbb{R}} f(t,x,p)dp.$$
 (5)

#### 3. HYDRODYNAMIC LIMIT

#### 3.1 Scaling laws

Loosely speaking, the hydrodynamical limit corresponds to near equilibrium situations where all considered fields vary slowly in space and time. This definition can be made precise in the following way. The near equilibrium character of the hydrodynamical regimes is taken into account by assuming that the distribution f of the diffusing particle in phase space can be expanded around the Maxwellian local equilibrium distribution of density n(t, x), temperature  $\theta(x)$  and vanishing mean velocity:

$$f_0(t,x,p) = n(t,x) \frac{\exp\left(-\frac{p^2}{2mk_B\theta(x)}\right)}{\sqrt{2\pi mk_B\theta(x)}}$$
(6)

We thus introduce an infinitesimal parameter  $\epsilon$  and a collection of functions  $f_k(t, x, p)$  for k > 0 such that the solution of the Kolmogorov equation (3) reads:

$$f(t,x,p) = \sum_{k=0}^{\infty} \epsilon^k f_k(t,x,p).$$
(7)

We treat (7) as a Chapman-Enskog expansion and therefore impose that the  $f_k$ 's for k > 0 do not contribute to the particle density:

$$\int_{\mathbb{R}} f_k(t, x, p) dp = 0, \quad \text{for} \quad k > 0.$$
(8)

The functions  $f_k$  will be obtained by solving the transport equation (3) and condition (8) implies that the all  $f_k$ 's depend on n and  $\theta$  only.

We now assume that the temperature field  $\theta(x)$  of the surrounding fluid fluctuates only weakly around its mean value. In other words, we suppose there exists a typical temperature  $\theta_{\star}$ , such that for all x,  $|\theta(x) - \theta_{\star}| << \theta_{\star}$ . It is thus natural to restrict the choice of the model's parameters  $\alpha(x,p)$  and D(x,p) by imposing that there exist also two quantities  $\alpha_{\star}$  and  $D_{\star} = mk_B\alpha_{\star}\theta_{\star}$ , such that  $|\alpha(x,p) - \alpha_{\star}| << \alpha_{\star}$  and  $|D(x,p) - D_{\star}| << D_{\star}$ , for any value of the particle position x and for any value of the momentum p, accessible with non-vanishing probability.

It is then possible to define a microscopic time-scale  $\tau = 1/\alpha_{\star}$ , which can be interpreted as a mean free-flight time. In the same manner, a microscopic momentum typical scale  $\rho$  can be defined as  $\rho = \sqrt{D_{\star}/\alpha_{\star}}$ . Consequently, the typical length scale  $\lambda = \tau \rho/m$  emerges naturally, and can be interpreted as the mean free path of the diffusing particle.

Let us now suppose (i) that the temperature field varies on a typical length scale  $\lambda/\tilde{\epsilon}$  much larger than the mean free path (ii) that the distribution function f varies on the same length scale as  $\theta$  and on a time-scale  $\tau/\nu$  much larger than the mean flight time. The infinitesimals  $\tilde{\epsilon}$  and  $\nu$  are at this stage a priori unrelated to each other and to  $\epsilon$ .

The transport equation (3) is then best solved by introducing the dimensionless time and space variables  $T = \nu t/\tau$  and  $X = \tilde{\epsilon}x/\lambda$  as well as the dimensionless momentum  $P = p/\rho$ . We also define a dimensionless density N(T, X) as:

$$N(T,X) = \frac{\lambda}{\tilde{\epsilon}} n\left(\frac{T\tau}{\nu}, \frac{X\lambda}{\tilde{\epsilon}}\right),\tag{9}$$

and a dimensionless phase space density F(T, X, P) as:

$$F(T, X, P) = \frac{\lambda \rho}{\tilde{\epsilon}} f\left(\frac{T\tau}{\nu}, \frac{X\lambda}{\tilde{\epsilon}}, \rho P\right), \tag{10}$$

so that the following simple normalization relations hold:

$$\int_{\mathbb{R}} N(T, X) dX = 1, \tag{11}$$

$$\int_{\mathbb{R}^2} F(T, X, P) dX dP = 1$$
(12)

In term of the dimensionless variables, the Kolmogorov equation (3) reads:

$$\nu \partial_T F + \tilde{\epsilon} \partial_X \left( P F \right) = \tilde{\epsilon} \partial_P \left( K(X, P) \frac{d\Theta}{dX} F \right) + \mathcal{L}F \quad (13)$$

where

$$K(X,P) = \frac{\kappa\left(\frac{X\lambda}{\tilde{\epsilon}}, P\rho\right)}{k_B}, \quad \text{and} \quad \Theta(X) = \frac{\theta\left(\frac{X\lambda}{\tilde{\epsilon}}\right)}{\theta_{\star}}.$$
 (14)

The linear operator  $\mathcal{L}$  is defined by:

$$\mathcal{L}F = \partial_P \left( \mathcal{A}(X, P) P F + \mathcal{D}(X, P) \partial_P F \right), \qquad (15)$$

with

$$\mathcal{A}(X,P) = \frac{\alpha\left(\frac{X\lambda}{\tilde{\epsilon}}, P\rho\right)}{\alpha_{\star}} \tag{16}$$

and

$$\mathcal{D}(X,P) = \frac{D\left(\frac{X\lambda}{\tilde{\epsilon}}, P\rho\right)}{D_{\star}} \tag{17}$$

The first two moments of (13) read:

$$\begin{cases}
\nu \partial_T N + \tilde{\epsilon} \partial_X J = 0 \\
\nu \partial_T J + \tilde{\epsilon} \partial_X S = -\tilde{\epsilon} K(X, P) \frac{d\Theta}{dX} N \\
- \int_{\mathbb{R}} \mathcal{A}(X, P) P F dP \\
+ \int_{\mathbb{R}} \partial_P \mathcal{D}(X, P) F dP,
\end{cases}$$
(18)

where J is the dimensionless particle current  $\int_{\mathbb{R}} P F dP$ , and S is  $\int_{\mathbb{R}} P^2 F dP$ . Since all odd-order momenta of the Maxwell distribution vanish, all odd-order momenta of the distribution F given by (7) are  $\mathcal{O}(\epsilon)$ . Thus, J and the integrals in the right-hand side of (18) are  $\mathcal{O}(\epsilon)$ , but S is  $\mathcal{O}(1)$ . The first equation in (18) thus implies that  $\nu = \tilde{\epsilon}\epsilon$ but the second one does not enforce any particular relation between  $\epsilon$  and  $\tilde{\epsilon}$ . Three types of scalings are thus compatible with (18). The first one is  $\tilde{\epsilon} < \epsilon$ , the second one is  $\tilde{\epsilon} > \epsilon$  and the third one is evidently  $\tilde{\epsilon} = \epsilon$ . Solutions which obey this last scaling law will exhibit a richer macroscopic physics because the choice  $\tilde{\epsilon} = \epsilon$  maximizes the number of term of equal order in (18). We therefore focus on this scaling law and thus retain :

$$\tilde{\epsilon} = \epsilon$$
 and  $\nu = \epsilon^2$  (19)

for the remainder of this contribution. Note that these scaling laws are identical to those obeyed by the hydrodynamic regimes of the Ornstein-Uhlenbeck process in uniform temperature fields.

#### 3.2 First order Chapman-Enskog expansion

We now write down the Kolmogorov equation at first order in  $\epsilon$ . According to (13), we need K at zeroth order and  $\mathcal{L}$ , *i.e.*  $\mathcal{A}$  and  $\mathcal{D}$ , at first order. We suppose that K, at zeroth order in  $\epsilon$ , is a constant  $K_0$  independent of x and p. The force proportional to  $\frac{d\theta}{dx}$  is then exactly identical to the standard thermophoresis force. The friction and noise coefficients are expanded into:

$$\mathcal{A}(X,P) = \mathcal{A}_0(X,P^2) + \epsilon \mathcal{A}_1(X,P^2) P \frac{d\Theta}{dX}, \qquad (20)$$

and

 $\epsilon$ :

$$\mathcal{D}(X,P) = \mathcal{D}_0(X,P^2) + \epsilon \mathcal{D}_1(X,P^2) P \frac{d\Theta}{dX}.$$
 (21)

We further assume that  $\mathcal{A}_0$  and  $\mathcal{D}_0$  obey the local fluctuation-dissipation relation:

$$\frac{\mathcal{D}_0(X, P^2)}{\mathcal{A}_0(X, P^2)} = \Theta(X), \quad \text{for all } P \text{ and } X, \qquad (22)$$

The terms  $\mathcal{A}_1$  and  $\mathcal{D}_1$  have been omitted by previous authors but are clearly necessary if one wants to ensure the consistency of the first order treatment.

The Kolmogorov equation then reads, at first order in

$$\epsilon \partial_X (P F) - \epsilon K_0 \frac{d\Theta}{dX} \partial_P F =$$

$$\mathcal{L}_0 F + \epsilon \frac{d\Theta}{dX} \mathcal{L}_1 F,$$
(23)

with

$$\mathcal{L}_0 F = \partial_P \left( \mathcal{A}_0(X, P^2) P F + \mathcal{D}_0(X, P^2) \partial_P F \right), \quad (24)$$

and

$$\mathcal{L}_1 F = \partial_P \left( \mathcal{A}_1(X, P^2) P^2 F + \mathcal{D}_1(X, P^2) P \partial_P F \right).$$
(25)

The dimensionless local equilibrium distribution:

$$F_0(T, X, P) = N(T, X) \frac{\mathrm{e}^{-\frac{P^2}{2\Theta(X)}}}{\sqrt{2\pi\Theta(X)}}$$
(26)

solves (23) at order zero in  $\epsilon$ . The first order terms of (23) collect into:

$$\mathcal{L}_0 F_1 = \partial_X \left( P F_0 \right) - \frac{d\Theta}{dX} \left( K_0 \partial_P F_0 + \mathcal{L}_1 F_0 \right), \qquad (27)$$

where  $F_1$  is the dimensionless version of  $f_1$ , the first order term of the Chapman-Enskog expansion (7). Equation (27) can be integrated over P to deliver:

$$\mathcal{A}_{0}(X, P^{2})PF_{1} + \mathcal{D}_{0}(X, P^{2})\partial_{P}F_{1} = -\partial_{X} \Big(\Theta(X)F_{0}(T, X, P)\Big) - \frac{d\Theta}{dX} \bigg(K_{0}F_{0} + \mathcal{A}_{1}(X, P^{2})P^{2}F_{0} + \mathcal{D}_{1}(X, P^{2})P\partial_{P}F_{0}\bigg).$$
(28)

The function  $H_1(T, X, P)$ , defined by:

$$H_1(T, X, P) = F_1(T, X, P) \frac{\sqrt{2\pi\Theta(X)}}{e^{-\frac{P^2}{2\Theta(X)}}},$$
 (29)

for all T, X and P, verifies:

$$\partial_P H_1 = -\frac{\Theta(X)}{\mathcal{D}_0(X, P^2)} \partial_X N \\ -\left(\frac{\frac{1}{2} + K_0 + P^2 \mathcal{A}_1(X, P^2)}{\mathcal{D}_0(X, P^2)}\right) N \frac{d\Theta}{dX} \\ -\left(\frac{\frac{1}{2} - \mathcal{D}_1(X, P^2)}{\mathcal{D}_0(X, P^2)}\right) P^2 \frac{N}{\Theta(X)} \frac{d\Theta}{dX}.$$

$$(30)$$

The first order contribution  $J_1$  to the particle current can be expressed in terms of the function  $H_1$  as:

$$J_{1}(T,X) = \int_{\mathbb{R}} H1(T,X,P) \frac{e^{-\frac{P^{2}}{2\Theta(X)}}}{\sqrt{2\pi\Theta(X)}} PdP$$
  
$$= \Theta(X) \int_{\mathbb{R}} \partial_{P} H1(T,X,P) \frac{e^{-\frac{P^{2}}{2\Theta(X)}}}{\sqrt{2\pi\Theta(X)}} dP.$$
 (31)

Taking into account expression (30) for  $\partial_P H_1$ , the first order contribution to the particle current can be related as follows to the gradients of the particle density and of the temperature:

$$J_1(T,X) = -\Phi(X) \frac{\partial N}{\partial X} - \Psi(X) N(T,X) \frac{d\Theta}{dX}, \quad (32)$$

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where the diffusion coefficient  $\Phi$  and the thermal diffusion coefficient  $\Psi$  have the following expressions :

$$\begin{cases}
\Phi = \Theta^{2}(X) \int_{\mathbb{R}} \frac{1}{\mathcal{D}_{0}(X, P^{2})} \frac{e^{-\frac{P^{2}}{2\Theta(X)}}}{\sqrt{2\pi\Theta(X)}} dP, \\
\Psi = \Theta(X) \int_{\mathbb{R}} \frac{\frac{1}{2} + K_{0}}{\mathcal{D}_{0}(X, P^{2})} \frac{e^{-\frac{P^{2}}{2\Theta(X)}}}{\sqrt{2\pi\Theta(X)}} dP \\
+\Theta(X) \int_{\mathbb{R}} \frac{\mathcal{A}_{1}(X, P^{2})}{\mathcal{D}_{0}(X, P^{2})} \frac{e^{-\frac{P^{2}}{2\Theta(X)}}}{\sqrt{2\pi\Theta(X)}} P^{2} dP \\
+ \int_{\mathbb{R}} \frac{\frac{1}{2} - \mathcal{D}_{1}(X, P^{2})}{\mathcal{D}_{0}(X, P^{2})} \frac{e^{-\frac{2P^{2}}{2\Theta(X)}}}{\sqrt{2\pi\Theta(X)}} P^{2} dP
\end{cases}$$
(33)

The first term in (32) is the usual contribution of the particle density gradient to the particle current (Fick's law). The second is the traditional macroscopic description of the Ludwig-Soret effect. Previous stochastic models (Widder and Titulaer, 1989) neglect the  $\mathcal{A}_1$  and  $\mathcal{D}_1$ contributions to  $\Psi$  and also assume that  $\mathcal{D}_0$  is independent of  $P^2$ . It is clear that the new models are much more flexible and allow, for example, for positive as well as negative values of the Soret coefficient.

To fully test these new models, one should have experimental access, not only to Soret coefficients and to  $\mathcal{A}_0$ and  $\mathcal{D}_0$  coefficients, but also to  $\mathcal{A}_1$  and  $\mathcal{D}_1$  coefficients. To the best of our knowledge, no  $\mathcal{A}_1$  and  $\mathcal{D}_1$  coefficient has yet been measured, if only because previous theoretical efforts had not taken these coefficients into account and thus, presumably, drawn the attention of experimentalists away from measuring them. A less satisfactory alternative would be to compute these coefficients from kinetic theoretical models. Unfortunately, no such computation exists at present in the literature.

#### 4. CONCLUSION

We have introduced new microscopic stochastic models for the Ludwig-Soret effect. These models are essentially Ornstein-Uhlenbeck processes with position dependent noise and friction coefficients and an additional thermophoresis force term. They differ from the previously introduced models by consistently taking into account all possible contributions generated by a non-vanishing temperature gradient. The hydrodynamic limit of these models has been studied with a first order Chapman-Enskog expansion. The new models predict that large scale temperature inhomogeneities induce a non vanishing particle current proportional to the temperature gradient. This prediction concurs with standard macroscopic models from continuous media theories (Landau and Lifschitz, 1987). The new models are also flexible enough to describe thermodiffusion in a much more realistic way than earlier stochastic approaches, allowing in particular for both positive and negative Soret coefficients.

This work can and should be extended in various directions. A first task is to consider several practically important applications of the Ludwig-Soret effect (Schimpf and Giddings, 1987, 1989; Perronace et al., 2002; Clusius and Dickel, 1938; Joly et al., 2000) and to determine, for each of these applications, which model(s) in the class introduced above best fits the experimental situations. Since the new microscopic models contain contributions linear in temperature gradients which are absent form the older models, it is for example natural to wonder if these extra terms suffice to explain all the documented discrepancies between measured values of the Soret coefficients and previous theoretical predictions. In particular, the  $\mathcal{A}_1$  and  $\mathcal{D}_1$ coefficients should be experimentally determined or, if possible, theoretically computed in a realistic manner and the value of the Soret coefficient predicted by (33) compared to the value deduced from experiments.

On the more theoretical side, the work presented in this article should be extended to both special and general relativistic realms, if only to model temperature driven diffusions in stars (Kippenhahn and Weigert, 1990).

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