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A Colored-Noise Approach to Brownian Motion in Position Space. Corrections to the Smoluchowski Equation

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The contraction of the description of Brownian motion from phase space to position space is discussed by means of non-Markovian Langevin equations in position space. A Fokker–Planck equation valid for any time is derived for the harmonic oscillator, and the overdamped, critical, and infradamped cases are discussed. For anharmonic potentials systematic corrections to the Smoluchowski equation are derived. Such corrections can be interpreted in this context as an expansion in powers of the correlation time of the "colored" stochastic noise appearing in the Langevin equation. The breakdown of the Fokker–Planck approximation is also discussed.

KEY WORDS: Smoluchowski equation; Fokker–Planck equation; Langevin equation; non-Markovian process; white and colored noise; adiabatic elimination.

1. INTRODUCTION

An old question posed by Uhlenbeck and Orstein[1] is to find the exact equation satisfied by the probability distribution in position space of a Brownian particle that can be in general under the influence of some potential. An approximate answer to this question is given by the Smoluchowski equation[2] valid for long times and high frictions. Generally speaking, the role of these two limits is not yet completely clear. The exact answer for free Brownian motion has recently been discussed by Mazo[3] and an extensive history of the problem is contained in a paper by Wilemski.[4] Besides its intrinsic interest in specific cases,[5] this problem provides a real physical and not too complicated example which allows important subjects to be discussed, such as adiabatic elimination of variables[6] and non-Markovian stochastic differential equations. Thus, its analysis seems worthwhile. Indeed, the elimination of momentum variables is the simplest example of adiabatic

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elimination and we shall show that it leads to Langevin equations with colored noise,[7] for which Fokker–Planck equations will be derived. This is also related to the "irrelevant" non-Markovian terms appearing in the renormalization group approach to critical dynamics.[8]

The Fokker–Planck equation in the complete phase space is the common starting point of the formalisms which have been proposed[4,9–13] to obtain systematic corrections to the Smoluchowski equation. Brinkman[9] derived an equation involving a second-order derivative with respect to time that has been criticized by Hemmer,[14] who showed that, in the case of free Brownian motion, it does not give for small times a better result than the Smoluchowski equation (see also Ref. 12). Wilemski[4] obtained a first correction to the Smoluchowski equation in an expansion in powers of the inverse of the friction coefficient of the momentum. The corrected equation is a Fokker–Planck equation in position space in which a transient regime is neglected. Such an equation had been obtained earlier by Stratonovich[10] without explicit reference to the problem of Brownian motion.[2] This corrected Smoluchowski equation has been recently reobtained by Titulaer,[11] Skinner and Wolynes,[12] and Chaturvedi and Shibata.[13]

In Refs. 11 and 13 it is stressed that the contraction of the description performed implies some kind of projection in position space. Titulaer's[11] work is based on the Chapman–Enskog method, while Chaturvedi and Shibata's[13] work relies on a natural projector formalism.[15] These developments allow one to evaluate corrections to the Smoluchowski equation to any desired order in the inverse of the friction coefficient. Nevertheless, we feel that the mathematical procedures used in Refs. 9–13 obscure the actual physical process in position space and they make it difficult in general to deal with simple problems such as the harmonic oscillator.[11]

The approach we present here is not based on the Fokker–Planck equation in phase space, but on the Langevin equation in position space obtained by projecting in this space the original phase-space Langevin equations. In this way we start from the very beginning with a problem only in position space. The most naive way of doing this is by eliminating adiabatically the momentum by setting its time derivative equal to zero.[6] This just leads to the Smoluchowski equation. If we perform this elimination exactly (it only remains an initial condition for the momentum), the resulting Langevin equation in position space defines a non-Markovian process. The non-Markovicity is caused by the reduction of the number of variables and it is reflected in the appearance of memory kernels and colored stochastic noise. As long as the Langevin equation remains linear, it is possible to associate to it a Fokker–Planck equation valid for any time. Following this

[2] This result [Eq. (4.245) of Stratonovich's monograph] seems to have been overlooked in the recent literature[4,11–13] on this topic.

method, we solve the problem of a harmonic potential in Section 2, deriving an equation for the exact one-time probability density in position space which is valid for any value of the friction constant. Approximate Fokker–Planck equations for nonlinear problems which reproduce earlier results are obtained in Section 3. The picture of a non-Markovian process in position space followed in this approach gives a deeper physical insight into the meaning of such equations. On the other hand, the breakdown of the existence of a Fokker–Planck equation in some order of approximation is very clearly related to the colored behavior of the stochastic force. The basis of the derivation of such Fokker–Planck equations is reviewed in Appendix A.

Finally, it should be stressed that the existence of Fokker–Planck equations is not in disagreement with the non-Markovian character of the stochastic process. The Fokker–Planck equation we shall use is not, although it has the same form, a bona fide Fokker–Planck equation for a nonstationary Markov process[16] in the sense that its fundamental solution is not the conditional probability for any time. The solution of such a Fokker–Planck equation for a non-Markovian process is only valid to evaluate one-time averages and it is no help in multitime averages.[17]

Our main conclusions are summarized in Section 4.

2. HARMONIC POTENTIAL

In this section we derive a Fokker–Planck equation, valid for any time and any value of the friction constant, for the probability density in position space of a Brownian particle in a harmonic potential. It had been earlier claimed[11] that it is impossible to derive such an equation valid for the transitional regime in which initial conditions have not yet decayed. The derivation presented here shows the inexactness of this statement, at least for this special case. By means of that exact equation the overdamped, critical, and infradamped cases will be considered separately and some differences between the long-time limit and high-friction limit will be discussed. The implications of assuming Maxwellian initial conditions for the momentum will also be discussed. Some final remarks will be made for free Brownian motion considered as a limiting case.

2.1. Fokker–Planck Equation

The stochastic equations of motion for the position q and momentum p of a brownian harmonic oscillator are

$$\dot{p}(t) = -\lambda p(t) - \omega^2 q(t) + \zeta(t) \tag{2.1}$$

$$\dot{q}(t) = p(t) \tag{2.2}$$

where we have taken a unity mass and $\zeta(t)$ is the stochastic driving force, assumed to be Gaussian and white noise, with zero mean and correlation function

$$\langle\zeta(t)\zeta(t')\rangle = 2D\,\delta(t - t') \tag{2.3}$$

where, according to the fluctuation-dissipation relation,

$$D = \lambda k_B T \tag{2.4}$$

The contraction of the description from phase space to position space is carried out by formally solving (2.1) and substituting into (2.2)

$$\dot{q}(t) = -\omega^2 \int_0^t e^{-\lambda(t-t')} q(t')\,dt' + \int_0^t e^{-\lambda(t-t')}\zeta(t')\,dt' + P_0 e^{-\lambda t} \tag{2.5}$$

where p_0 is the initial value of the momentum $p(0) = p_0$. This equation can be rewritten as

$$\dot{q}(t) = -\int_0^t \beta(t - t')q(t')\,dt' + F(t) + \bar{\zeta}(t) \tag{2.6}$$

where the following notation has been introduced:

$$\beta(t - t') = \omega^2 e^{-\lambda(t-t')}, \qquad t > t' \tag{2.7}$$

$$\bar{\zeta}(t) = \int_0^t e^{-\lambda(t-t')}\zeta(t')\,dt' \tag{2.8}$$

$$F(t) = p_0 e^{-\lambda t} = (p_0/\omega^2)\beta(t) \tag{2.9}$$

Due to the projection made in position space, the Markovian character of Eqs. (2.1) and (2.2) is absent in Eq. (2.6). This is reflected in the appearance of the memory kernel $\beta(t - t')$ and also in the stochastic force $\bar{\zeta}(t)$, which is no longer white, but colored. The new force $\bar{\zeta}(t)$ is still Gaussian with zero mean, and its correlation function is now

$$\begin{aligned}\sigma(t, t') = \langle\bar{\zeta}(t)\bar{\zeta}(t')\rangle &= k_B T(e^{-\lambda(t-t')} - e^{-\lambda(t+t')}) \\ &= k_B T[\omega^{-2}\beta(t - t') - \omega^{-4}\beta(t)\beta(t')], \qquad t > t'\end{aligned} \tag{2.10}$$

It is also important to note that no fluctuation-dissipation relation between $\sigma(t, t')$ and $\beta(t - t')$ exists in Eq. (2.6). Such a relation is only recovered in the long-time limit $t \to \infty$, $t' \to \infty$.

As a first step toward obtaining a Fokker–Planck equation for the probability density in position space, we transform Eq. (2.6) into another

Langevin equation with no memory kernel in it. This transformation[18–20] follows the procedure used for Langevin equations of the type of Eq. (2.6) but for which a fluctuation-dissipation relation was assumed.[18,19] Laplace-transforming Eq. (2.6), one has

$$\hat{q}(z) = \hat{\chi}(z)[q(0) + \hat{F}(z) + \hat{\bar{\zeta}}(z)] \tag{2.11}$$

so that

$$q(t) = \chi(t)q(0) + \int_0^t \chi(t-s)F(s)\,ds + \int_0^t \chi(t-s)\bar{\zeta}(s)\,ds \tag{2.12}$$

where

$$\hat{\chi}(z) = [z + \hat{\beta}(z)]^{-1} \tag{2.13}$$

with $\hat{\beta}(z)$ the Laplace transform of $\beta(t)$ and $\chi(t)$ the Laplace antitransform of $\hat{\chi}(z)$. The initial condition $q(0)$ can be eliminated by taking the time derivative of Eq. (2.12), and one is then led to the desired Langevin equation:

$$\dot{q}(t) = -\bar{\beta}(t)q(t) + \chi(t)\frac{d}{dt}\int_0^t \frac{\chi(t-s)}{\chi(t)}F(s)\,ds + \chi(t)\frac{d}{dt}\int_0^t \frac{\chi(t-s)}{\chi(t)}\bar{\zeta}(s)\,ds \tag{2.14}$$

where

$$\bar{\beta}(t) = -\dot{\chi}(t)/\chi(t) \tag{2.15}$$

This is a linear Langevin equation of the type studied in Appendix A and according to Eq. (A15) its corresponding Fokker–Planck equation is

$$\frac{\partial P(q,t)}{\partial t} = \frac{\partial}{\partial q}\bar{\beta}(t)qP(q,t) - \frac{\partial}{\partial q}\chi(t)\frac{d}{dt}\int_0^t \frac{\chi(t-s)}{\chi(t)}F(s)\,ds\,P(q,t) + \frac{\partial^2}{\partial q^2}D(t)P(q,t) \tag{2.16}$$

where

$$D(t) = \int_0^t d\tau\,\gamma(t,z)\exp\left[-\int_\tau^t \bar{\beta}(\tau')\,d\tau'\right] = \int_0^t \frac{\gamma(t,\tau)\chi(t)}{\chi(\tau)}\,d\tau \tag{2.17}$$

and

$$\gamma(t,\tau) = \left\langle \chi(t)\frac{d}{dt}\int_0^t \frac{\chi(t-s)}{\chi(t)}\bar{\zeta}(s)\,ds\,\chi(\tau)\frac{d}{d\tau}\int_0^\tau ds'\,\frac{\chi(\tau-s')}{\chi(\tau)}\bar{\zeta}(s')\right\rangle \tag{2.18}$$

Let us now give a more explicit expression for the diffusion $D(t)$. From Eqs. (2.17), (2.18), and (2.10) we have

$$D(t) = \frac{\chi^2(t)}{2} \frac{d}{dt} \chi^{-2}(t)[A_1(t) + A_2(t)] \tag{2.19}$$

$$A_1(t) = \int_0^t ds \int_0^t ds' \, \chi(t-s)\chi(t-s') \frac{k_B T}{\omega^2} \beta(s-s') \tag{2.20}$$

$$A_2(t) = -\int_0^t ds \int_0^t ds' \, \chi(t-s)\chi(t-s') \frac{k_B T}{\omega^4} \beta(s)\beta(s') \tag{2.21}$$

$A_1(t)$ can be evaluated by taking its time derivative

$$\dot{A}_1(t) = -(2k_B T/\omega^2)\chi(t)\dot{\chi}(t) \tag{2.22}$$

where use has been made of the definition (2.13) of $\chi(t)$. By recalling that $\chi(0) = 1$, we obtain

$$A_1(t) = (k_B T/\omega^2)[1 - \chi^2(t)] \tag{2.23}$$

Again by the definition (2.13) of $\chi(t)$ we have

$$A_2(t) = -(k_B T/\omega^4)[\dot{\chi}(t)]^2 \tag{2.24}$$

Substituting (2.23) and (2.24) in (2.19) and replacing (2.15), we obtain

$$D(t) = \frac{k_B T}{\omega^2} \bar{\beta}(t) - \frac{k_B T}{2\omega^4} \chi^2(t) \frac{d}{dt} [\bar{\beta}(t)^2] \tag{2.25}$$

A more explicit expression can also be given for the second term on the right-hand side of Eq. (2.16). Substituting the value (2.9) of $F(s)$ and using once again the definition (2.13) of $\chi(t)$ and Eq. (2.15), we find that this term reduces to

$$\chi(t) \frac{p_0}{\omega^2} \left[\frac{d}{dt} \bar{\beta}(t) \right] \tag{2.26}$$

Substituting (2.25) and (2.26) in (2.16), we find that the Fokker–Planck equation becomes

$$\frac{\partial P(q,t)}{\partial t} = \frac{\partial}{\partial q} \bar{\beta}(t) q P(q,t) - \frac{p_0 \chi(t)}{\omega^2} \left[\frac{d}{dt} \bar{\beta}(t) \right] \frac{\partial P(q,t)}{\partial q} + \frac{k_B T}{\omega^2} \bar{\beta}(t) \frac{\partial^2}{\partial q^2} P(q,t)$$
$$- \frac{k_B T}{2\omega^4} \chi^2(t) \left[\frac{d}{dt} \bar{\beta}(t)^2 \right] \frac{\partial^2}{\partial q^2} P(q,t) \tag{2.27}$$

This is the desired equation, valid for any time and any value of the parameters, given in terms of $\chi(t)$, which has different values depending on the relative strength of the friction λ and of the potential, the latter being measured by the frequency ω. In the following the three possible cases are analyzed. In any case, Eq. (2.27) is a linear Fokker–Planck equation and although it has time-dependent coefficients, the exact solution can always be given in terms of these coefficients.

2.2. Overdamped Case

We study now the particular form of (2.27) under the assumption that

$$\lambda/2 > \omega \tag{2.28}$$

This is the case usually considered in the literature[11] because it is the one for which the high-friction limit becomes more natural. For the range of values (2.28), $\chi(t)$ becomes

$$\chi(t) = e^{-\lambda t/2}[\text{ch}\, at + (\lambda/2a)\, \text{sh}\, at]; \qquad a^2 = (\lambda/2)^2 - \omega^2 \tag{2.29}$$

and so

$$\bar{\beta}(t) = \omega^2\, \text{sh}\, at/[a\, \text{ch}\, at + (\lambda/2)\, \text{sh}\, at] \tag{2.30}$$

$$d\bar{\beta}(t)/dt = [\omega/\chi(t)]^2 e^{-\lambda t} = \omega^2/[\text{ch}\, at + (\lambda/2a)\, \text{sh}\, at]^2 \tag{2.31}$$

Equations (2.27) and (2.29)–(2.31) give the exact and explicit form of the Fokker–Planck equation. Earlier approximate results can be obtained by taking the appropriate limits. If it is assumed that

$$e^{at} \gg e^{-at} \tag{2.32}$$

Eqs. (2.29)–(2.31) become

$$\chi(t) = [1 + (\lambda/2a)]e^{-(\lambda/2 - a)t} \tag{2.33}$$

$$\bar{\beta}(t) = \lambda/2 - a \tag{2.34}$$

$$d\bar{\beta}(t)/dt = \{\omega/[1 + (\lambda/2a)]\}^2 e^{-2at} \tag{2.35}$$

and the Fokker–Planck equation (2.27) reads

$$\frac{\partial P(q, t)}{\partial t} = \left(\frac{\lambda}{2} - a\right) \frac{\partial}{\partial q} qP(q, t) - \frac{2p_0}{1 + \lambda/2a} e^{-(\lambda/2 + a)t} \frac{\partial}{\partial q} P(q, t)$$
$$+ \frac{k_B T}{\omega^2} \left(\frac{\lambda}{2} - a\right)(1 - e^{-\lambda t}) \frac{\partial^2}{\partial q^2} P(q, t) \tag{2.36}$$

It is worth emphasizing that Eq. (2.36), which still contains the p_0 initial value, is only restricted by (2.32), which is satisfied both for long times and high frictions, but which is much less restrictive than the usual assumption[2]

$$\lambda t \gg 1 \tag{2.37}$$

Equation (2.36) contains two terms involving exponentials. One of them depends on the initial condition p_0 and the other does not. These exponentials can be neglected by assuming (2.37). Then,

$$\frac{\partial P(q,t)}{\partial t} = \left(\frac{\lambda}{2} - a\right)\frac{\partial}{\partial q} qP(q,t) + \frac{k_B T}{\omega^2}\left(\frac{\lambda}{2} - a\right)\frac{\partial^2}{\partial q^2} P(q,t) \qquad (2.38)$$

which is the result of Titulaer.[11]

Assumption (2.37) can be satisfied either by considering a fixed, finite value of λ and a long-time t (long-time limit) or by considering a fixed, finite time and a large value of λ (high-friction limit). In the latter case, Eq. (2.38) is somehow misleading, since it contains all orders in λ^{-1}. Therefore Eq. (2.38) should be considered as valid for long times in which the transient regime has already elapsed. In the high-friction limit an expansion of Eq. (2.36) in terms of λ^{-1} has to be made. The same results are of course obtained if Eq. (2.38) is expanded. In first order one obtains the Smoluchowski equation[2]

$$\frac{\partial P(q,t)}{\partial t} = \frac{\omega^2}{\lambda}\frac{\partial}{\partial q} qP(q,t) + \frac{k_B T}{\lambda}\frac{\partial^2}{\partial q^2} P(q,t) \qquad (2.39)$$

and in second order

$$\frac{\partial P(q,t)}{\partial t} = \frac{\omega^2}{\lambda}\left(1 + \frac{\omega^2}{\lambda^2}\right)\frac{\partial}{\partial q} qP(q,t) + \frac{k_B T}{\lambda}\left(1 + \frac{\omega^2}{\lambda^2}\right)\frac{\partial^2}{\partial q^2} P(q,t) \qquad (2.40)$$

which is the correction first given by Stratonovich.[10]

2.3. Critical Case

This is the case in which

$$\omega = \lambda/2 \qquad (2.41)$$

and thus assumption (2.32) can no longer be made. For this value of λ, $\chi(t)$ becomes

$$\chi(t) = e^{-(\lambda/2)}(1 + \lambda t/2) \qquad (2.42)$$

and so

$$\bar{\beta}(t) = (\tfrac{1}{2}\lambda)^2 t/(1 + \tfrac{1}{2}\lambda t) \qquad (2.43)$$

$$(d/dt)\bar{\beta}(t) = (\tfrac{1}{2}\lambda)^2/(1 + \tfrac{1}{2}\lambda t)^2 \qquad (2.44)$$

Therefore, the Fokker–Planck equation (2.27) becomes

$$\frac{\partial P(q,t)}{\partial t} = \frac{(\frac{1}{2}\lambda)^2 t}{1 + \frac{1}{2}\lambda t}\frac{\partial}{\partial q} qP(q,t) - \frac{P_0 e^{-1t/2}}{1 + \frac{1}{2}\lambda t}\frac{\partial P(q,t)}{\partial q}$$
$$+ \frac{k_B T t}{1 + \frac{1}{2}\lambda t}(1 - e^{-\lambda t})\frac{\partial^2}{\partial q^2} P(q,t) \qquad (2.45)$$

In the long-time limit (2.45) can be written as

$$\frac{\partial P(q,t)}{\partial t} = \frac{\lambda}{2}\frac{\partial}{\partial q} qP(q,t) + \frac{2k_BT}{\lambda}\frac{\partial^2}{\partial q^2} P(q,t) \tag{2.46}$$

This equation coincides with (2.38) with $a = 0$. This is in agreement with the physical fact that for any values of λ and ω the stationary state of the oscillator, which is obtained from the long-time limit, must coincide. On the other hand, the leading term in the high-friction limit of (2.45) gives

$$\frac{\partial P(q,t)}{\partial t} = \frac{\lambda}{2}\frac{\partial}{\partial q} qP(q,t) \tag{2.47}$$

which also can be obtained from (2.46) but not as a particular case of the Smoluchowski equation (2.39). We see that in this limit diffusion becomes negligible. The impossibility of deriving (2.47) as a particular case of the overdamped oscillator is due to the critical values attained by the parameters ω and λ. In fact, earlier systematic expansions become divergent at this point.[11]

2.4. Infradamped Case

This is defined by

$$\lambda/2 < \omega \tag{2.48}$$

Then

$$\chi(t) = e^{-\lambda t/2}[\cos at + (\lambda/2a)\sin at] \tag{2.49}$$

$$\bar{\beta}(t) = \omega^2 \sin at/[a\cos at + (\lambda/2)\sin at] \tag{2.50}$$

$$(d/dt)\bar{\beta}(t) = \omega^2/[\cos at + (\lambda/2a)\sin at]^2 \tag{2.51}$$

Equation (2.27) supplemented by (2.49)–(2.51) gives the Fokker–Planck equation for the infradamped oscillator. It is not possible to write down a Fokker–Planck equation in the long-time limit since no such limit exists for $\bar{\beta}(t)$. Even more, there exists a set of times for which $\bar{\beta}(t)$ diverges. For these points the Fokker–Planck operator is not defined. This fact is related to the nonexistence for all times of the inverse of the conditional probability for general non-Markov processes.[17] Nevertheless, this formal Fokker–Planck equation has a time-dependent solution whose long time does exist and it coincides, as it should, with the stationary solution of (2.39).

2.5. Maxwellian Initial Momentum Distribution

In the above developments, the initial value of the momentum has been considered as a fixed parameter in the evolution equation for $q(t)$ [Eq. (2.6)]. Thus, any correlation function or statistical average calculated with the solution of the Fokker–Planck equation (2.27) will depend on p_0. If this is not fixed but has some initial probability distribution, the correct result is obtained by taking the average over such a distribution.

If the initial momentum p_0 is considered from the beginning as a stochastic parameter, it may be interesting to find the master equation associated to (2.14) under these circumstances. The derivation of the Fokker–Planck equation (2.16) given in Appendix A has then to be modified by including a further average of Eq. (A3) over the probability distribution of p_0. This can be in general quite involved. Nevertheless, if we assume the Maxwellian Gaussian probability distribution with zero mean value for p_0, this new average can also be performed by means of Novikov's theorem[21] [see Eq. (A7)]. In practice, this is equivalent to considering in Eq. (2.6) or (2.14) the term

$$\zeta'(t) = F(t) + \bar{\zeta}(t) \tag{2.52}$$

as a new stochastic force whose correlation function over the mutually independent functional distribution of $\zeta(t)$ and the Maxwellian distribution of p_0 is

$$\sigma'(t, t') = \langle \zeta'(t)\zeta'(t')\rangle_{p_0} = (\langle p_0^2\rangle_{p_0} - k_B T)e^{-\lambda(t+t')} + k_B T\, e^{-\lambda(t-t')} \tag{2.53}$$

For the Maxwellian distribution

$$\langle p_0^2\rangle_{p_0} = k_B T \tag{2.54}$$

so that

$$\sigma'(t, t') = k_B T e^{-\lambda(t-t')} \tag{2.55}$$

Therefore, the Fokker–Planck equation (2.16) reduces to

$$\frac{\partial P(q, t)}{\partial t} = \frac{\partial}{\partial q}\bar{\beta}(t) q P(q, t) + \frac{\partial^2}{\partial q^2} D'(t) P(q, t) \tag{2.56}$$

where $D'(t)$ is defined in terms of $\gamma'(t, t')$ in the same way that $D(t)$ was defined in terms of $\gamma(t, t')$ in Eq. (2.17). The quantity $\gamma'(t, t')$ is defined as $\gamma(t, t')$ in Eq. (2.18) by replacing $\bar{\zeta}(t)$ by $\zeta'(t)$ and taking the average also over the Maxwellian distribution of p_0. Comparing (2.10) with (2.55), it becomes clear that $D'(t)$ is given by the same expression (2.19) for $D(t)$ but with $A_2(t) = 0$. Thus, the Fokker–Planck equation for $P(q, t)$ under the assumption of Maxwellian initial conditions for the momentum is

$$\frac{\partial P(q, t)}{\partial t} = \frac{\partial}{\partial q}\bar{\beta}(t) q P(q, t) + \frac{k_B T}{\omega^2}\bar{\beta}(t)\frac{\partial^2}{\partial q^2} P(q, t) \tag{2.57}$$

The assumption of Maxwellian initial conditions as compared to an arbitrary initial distribution for the momentum means physically the neglect of a transient regime in which the momentum distribution reaches thermal equilibrium. This is already seen in the fact that under this assumption $\sigma(t, t')$ [Eq. (2.10)] is replaced by $\sigma'(t, t')$ [Eq. (2.55)] so that Eq. (2.6) satisfies a fluctuation-dissipation relation that, as mentioned earlier, implies a long-time limit. On the other hand, terms that are negligible, after a transient regime, such as the exponential terms in Eq. (2.36), no longer appear under this special initial condition.

2.6. Free Brownian Motion

The Brownian motion problem in a vanishing potential can be very easily treated along the same lines as followed in Section 2.1. The Fokker–Planck equation in position spaces is of course the limit of (2.27) and (2.29)–(2.31) when $\omega^2 \to 0$. This equation can also be obtained by other methods[3,22] and is

$$\frac{\partial P(q, t)}{\partial t} = -\frac{\partial}{\partial q} p_0 e^{-\lambda t} P(q, t) + \frac{\partial^2}{\partial q^2} \frac{k_B T}{\lambda} (1 - 2e^{-\lambda t} + e^{-2\lambda t}) P(q, t) \tag{2.58}$$

It had already been remarked by Wilemski[4] that in this case the corrections to the Smoluchowski equation had to be of a high order in λ^{-1}. From (2.58) we see that such corrections are of order $e^{-\lambda t}$ and that they only exist for the transient regime.

If Maxwellian initial conditions for p_0 are assumed, going through the same steps as in Section 2.5, we obtain that (2.58) reduces to

$$\frac{\partial P(q, t)}{\partial t} = \frac{\partial^2}{\partial q^2} \frac{k_B T}{\lambda} (1 - e^{-\lambda t}) P(q, t) \tag{2.59}$$

This equation clearly shows that part of the transient regime cannot be eliminated by appropriate momentum initial conditions.

3. ANHARMONIC POTENTIALS

The equations of motion for a Brownian particle in an anharmonic potential $\phi(q)$ read

$$\dot{q}(t) = p(t) \tag{3.1}$$

$$\dot{p}(t) = -\lambda p(t) - \phi'(q(t)) + \zeta(t) \tag{3.2}$$

where ϕ' denotes the derivative of ϕ with respect to q and $\zeta(t)$ is the stochastic force defined in Section 2.1. In the same way as in the last section, formally solving (3.2) and substituting in (3.1) gives

$$\dot{q}(t) = e^{-\lambda t} p(0) - \int_0^t e^{-\lambda(t-t')} \phi'(q(t'))\, dt' + \bar{\zeta}(t) \tag{3.3}$$

Due to the nonlinearity contained in ϕ', this equation cannot be reduced to a memory-less form. Thus, we shall look for an approximate master equation for the probability density in q space in powers of λ^{-1}. The point we want to emphasize in this paper is how this can be done starting from the non-Markovian Langevin equation (3.3) and handling consistently the colored noise $\bar{\zeta}(t)$. In this way, the reason why no Fokker–Planck equation exists beyond terms of order λ^{-3} (Refs. 11–13) will clearly appear as related to the colored behavior of $\bar{\zeta}(t)$.

The term containing the initial condition $p(0)$ can be neglected in this approximation. The second term on the right-hand side of (3.3) can be approximated by successive integrations by parts and neglect of transient terms. This is equivalent to the formal expansion

$$e^{-\lambda(t-t')} = 2\lambda^{-1}\,\delta(t-t') + 2\lambda^{-2}\frac{d}{dt'}\,\delta(t-t') + \cdots, \qquad t > t' \tag{3.4}$$

Substituting in (3.3), we obtain

$$\dot{q}(t) = -\lambda^{-1}\phi'(q(t)) + \lambda^{-2}\phi''(q(t))\dot{q}(t) + \bar{\zeta}(t) \tag{3.5}$$

so that

$$\dot{q}(t) = -\lambda^{-1}\phi'(q(t))[1 + \lambda^{-2}\phi''(q(t))] + [1 + \lambda^{-2}\phi''(q(t))]\bar{\zeta}(t) + O(\lambda^{-5}) \tag{3.6}$$

The stochastic force $\bar{\zeta}(t)$ has not yet been approximated. If Eq. (3.4) is introduced in its definition (2.8), ill-behaved functions as $\dot{\zeta}(t)$ appear. Thus a consistent approximation to order λ^{-3} has to be made in its correlation function $\sigma(t, t')$ [Eq. (2.10)], which appears in the master equation associated to (3.6). Such an equation is derived in Appendix A, Eq. (A8), and it becomes in this case

$$\frac{\partial P(q,t)}{\partial t} = \frac{\partial}{\partial q}\lambda^{-1}\phi'(q)[1 + \lambda^{-2}\phi''(q)]P(q,t) + \frac{\partial}{\partial q}[1 + \lambda^{-2}\phi''(q)]\frac{\partial}{\partial q}\int_0^t dt'\,\sigma(t,t')\left\langle \delta(q(t) - q)\frac{\delta q(t)}{\delta\bar{\zeta}(t')}\right\rangle \tag{3.7}$$

where

$$P(q,t) = \langle \delta(q(t) - q)\rangle \tag{3.8}$$

The correlation function $\sigma(t, t')$ can now be expanded analogously to the expansion in Eq. (3.4). The second term $-k_BT\exp[-\lambda(t + t')]$ of $\sigma(t, t')$ will not contribute, when so expanded, to the integral in Eq. (3.7), and so

$$\sigma(t,t') = 2k_BT\left[\lambda^{-1}\,\delta(t-t') + \lambda^{-2}\frac{d}{dt'}\,\delta(t-t') + \cdots\right], \qquad t > t' \tag{3.9}$$

This expansion of $\sigma(t, t')$ is related to the so-called "quasicomplete random processes."[23] With such an expansion,

$$\int_0^t dt'\, \sigma(t, t') \frac{\delta q(t)}{\delta \bar{\zeta}(t')} = k_B T \left[\lambda^{-1} \frac{\delta q(t)}{\delta \bar{\zeta}(t')} - \lambda^{-2} \left(\frac{d}{dt'} \frac{\delta q(t)}{\delta \bar{\zeta}(t')} \right) + O(\lambda^{-3}) \right]_{t'=t} \tag{3.10}$$

The quantity $\delta q(t)/\delta \bar{\zeta}(t')$ is the response of $q(t)$ to the force $\bar{\zeta}(t')$. This response function is analyzed in Appendix B by means of an operator formalism. This formalism gives an analytic expression for it from which its time derivatives are easily evaluated. According to Eqs. (B12) and (B15), one has

$$\frac{\delta q(t)}{\delta \bar{\zeta}(t)} 1 + \lambda^{-2} \phi''(q(t)) \tag{3.11}$$

$$\begin{aligned} \left[\frac{d}{dt'} \frac{\delta q(t)}{\delta \bar{\zeta}(t')} \right]_{t'=t} &= [1 + \lambda^{-2} \phi''(q(t))] \{ \lambda^{-1} \phi'(q(t)) [1 + \lambda^{-2} \phi''(q(t))] \}' \\ &\quad - \lambda^{-1} \phi'(q(t)) [1 + \lambda^{-2} \phi''(q(t))] [1 + \lambda^{-2} \phi''(q(t))]' \end{aligned} \tag{3.12}$$

The second term in Eq. (3.12) does not contribute to Eq. (3.7) in order λ^{-3}. Substituting (3.10)–(3.12) in (3.7) and keeping terms up to order λ^{-3}, we obtain the following Fokker–Planck equation:

$$\begin{aligned} \frac{\partial P(q, t)}{\partial t} &= \frac{\partial}{\partial q} \lambda^{-1} \phi'(q) [1 + \lambda^{-2} \phi''(q)] P(q, t) \\ &\quad + \frac{\partial}{\partial q} [1 + \lambda^{-2} \phi''(q)] \frac{\partial}{\partial q} \lambda^{-1} k_B T P(q, t) \\ &= \frac{\partial}{\partial q} \{ \lambda^{-1} \phi'(q) [1 + \lambda^{-2} \phi''(q)] - \lambda^{-3} k_B T \phi'''(q) \} P(q, t) \\ &\quad + \lambda^{-1} k_B T \frac{\partial^2}{\partial q^2} [1 + \lambda^{-2} \phi''(q)] P(q, t) \end{aligned} \tag{3.13}$$

If we drop terms of order λ^{-3}, Eq. (3.13) becomes the Smoluchowski equation.[2] This corresponds to the limit of white noise for $\bar{\zeta}$ obtained by keeping only the first term in expansions (3.9) and (3.4). Terms of order λ^{-3} give the first correction to the Smoluchowski equation coinciding with the result of Refs. 4 and 10–13. It is worth noting that (3.13) is only completely meaningful when solved for the steady state, since transients have been neglected in its derivation. For natural boundary conditions and from the phase-space Fokker–Planck equation the exact steady-state solution is known to be

$$P_{\rm st}(q) = N e^{-\phi(q)} \tag{3.14}$$

This is also a stationary solution of both the Smoluchowski equation and of (3.13). The main interest with regard to (3.13) is thus in looking for non-equilibrium steady states in which nonnatural boundary conditions have to be imposed.

Equation (3.13) represents a Fokker–Planck approximation to the non-Markovian and nonlinear process defined in (3.3), which is valid for a small correlation time λ^{-1} of $\bar{\zeta}(t)$. Higher order corrections are systematically obtained by considering higher order terms in (3.4) and (3.9). Nevertheless, in the next order to the one considered up to now, that is, $O(\lambda^{-5})$, the Fokker–Planck approximation already breaks down. In this next order one adds to Eq. (3.10) a term

$$k_B T \lambda^{-3} \left[\frac{d^2}{dt'^2} \frac{\delta q(t)}{\delta \bar{\zeta}(t')} \right]_{t'=t} \tag{3.15}$$

This quantity is evaluated in Appendix B [Eq. (B17)] and it contains a term of order λ^{-5} in which $\bar{\zeta}(t)$ appears as a multiplicative factor. When this is substituted in (3.7) a factor $\langle \bar{\zeta}(t)\, \delta(q(t) - q) \rangle$ comes in. The average in this factor is discussed in Appendix A by means of Novikov's theorem[21] and it introduces a new derivative with respect to q. Thus, no Fokker–Planck equation will exist in this approximation since third-order derivatives are present. Nevertheless, it is easily checked that in the special case of a harmonic potential, the second term of (B17) involving $\bar{\zeta}(t)$ vanishes, and thus a Fokker–Planck approximation still exists. Moreover, this is true in any order of approximation. Thus, one has a power series in λ^{-n} that, when summed, becomes a Fokker–Planck equation valid in any order in λ^{-n}. In summing such a series, Eq. (2.38) is reobtained. One of the advantages of the approach presented here is that this equation can be directly obtained as done in the previous section without going through a series expansion. This could not be done in earlier approaches.[11]

4. CONCLUSIONS

We have presented in this paper a new approach to the problem of the corrections to the Smoluchowski equation based on a non-Markovian Langevin equation in position space. As stated in the introduction, this approach is of direct relevance to other related fields. The problem of a harmonic oscillator has been exactly solved in this framework. We have not noticed that this result has been previously published and we have obtained it in our approach by considering as starting equations ones which are already in position space.[3] This exact solution has allowed for a separate

[3] After the completion of this work we became aware of an alternative derivation of the results in Section 2.2 by Risken *et al.*[24] These authors rely on an analogy with quantum mechanics and also reobtain (3.13).

discussion of the overdamped, critical, and infradamped oscillators. Earlier approaches were unable to deal systematically with the last two cases. The specific roles of the long-time limit and of the high-friction limit have also been discussed in the context of this exact result, as well as the implications of special initial conditions.

For the nonlinear case earlier approximate results are reobtained. Our picture of a non-Markovian process in q space gives a more direct physical meaning to the different steps of these calculations. In particular, the expansion in powers of λ^{-1} can be reinterpreted as an expansion in terms of the correlation time of the colored stochastic force driving the process. The breakdown of the Fokker–Planck approximation for other than linear problems is also clearly displayed. On the other hand, Eq. (3.13) is a nonlinear Fokker–Planck equation derived from a nonlinear, non-Markovian Langevin equation, valid for small correlation times. This equation can be useful in other contexts to test the soundness of white noise approximations.

APPENDIX A. FOKKER–PLANCK EQUATIONS FOR COLORED-NOISE LANGEVIN EQUATIONS

We consider a general nonlinear Langevin equation

$$\dot{q}(t) = V(q(t); t) + g(q(t); t)\zeta(t) \tag{A1}$$

where we assume that the stochastic force $\zeta(t)$ is Gaussian with zero mean and arbitrary correlation function

$$\langle \zeta(t)\zeta(t')\rangle = \gamma(t, t') \tag{A2}$$

For each realization of the noise $\zeta(t)$ one can consider[25,26] an ensemble of systems obeying Eq. (A1). The probability of finding the system at the point q at time t is given by the probability density $\rho(q, t)$ which obeys the continuity or "stochastic Liouville equation"

$$\frac{\partial \rho(q, t)}{\partial t} = -\frac{\partial}{\partial q}[V(q; t) + g(q; t)\zeta(t)]\rho(q, t) \tag{A3}$$

where v and g are taken at a fixed point of q space.

The solution $q(t)$ of Eq. (A1) for each realization of $\zeta(t)$ is a functional of $\zeta(t)$ and of the initial condition q^0

$$q(t) = q([\zeta(t)], q^0, t) \tag{A4}$$

and therefore $\rho(q, t)$ can be expressed as the average of $\delta(q(t) - q)$ over the distribution of initial conditions q^0. It can be shown,[26] and it is physically clear, that the probability density $P(q, t)$ for the stochastic process $q(t)$

defined by (A1) is given by the average of $\rho(q, t)$ over the realizations of $\zeta(t)$: Therefore

$$P(q, t) = \langle \delta(q(t) - q) \rangle \tag{A5}$$

where the brackets $\langle \cdots \rangle$ mean average over both the distribution of initial conditions and over the probability distribution of the stochastic forces. Taking the latter in Eq. (A3), we obtain

$$\frac{\partial P(q, t)}{\partial t} = -\frac{\partial}{\partial q} V(q, t)P(q, t) - \frac{\partial}{\partial q} g(q, t)\langle \zeta(t)\, \delta(q(t) - q) \rangle \tag{A6}$$

Since $\delta(q(t) - q)$ is a functional of $\zeta(t)$, Novikov's theorem[21] states that

$$\begin{aligned} \langle \zeta(t)\, \delta(q(t) - q) \rangle &= \int_0^t dt'\, \gamma(t, t') \left\langle \frac{\delta[\delta(q(t) - q)]}{\delta\zeta(t')} \right\rangle \\ &= -\frac{\partial}{\partial q} \int_0^t dt'\, \gamma(t, t') \left\langle \delta(q(t) - q) \frac{\delta q(t)}{\delta \zeta(t')} \right\rangle \end{aligned} \tag{A7}$$

Substituting in (A6), we obtain

$$\begin{aligned} \frac{\partial P(q, t)}{\partial t} = &-\frac{\partial}{\partial q} V(q, t)P(q, t) \\ &+ \frac{\partial}{\partial q} g(q, t) \frac{\partial}{\partial q} \int_0^t dt'\, \gamma(t, t') \left\langle \delta(q(t) - q) \frac{\delta q(t)}{\delta \zeta(t')} \right\rangle \end{aligned} \tag{A8}$$

In general, this equation can be written in Fokker–Planck form only after some approximations such as the ones carried out in Section 3. Nevertheless, there are two cases in which (A8) reduces exactly to a Fokker–Planck equation. The first case is the white noise limit:

$$\gamma(t, t') = 2\gamma(t)\, \delta(t - t') \tag{A9}$$

It is proved in Appendix B that

$$\delta q(t)/\delta\zeta(t) = g(q(t), t) \tag{A10}$$

and therefore Eq. (A8) becomes the usual bona fide Fokker–Planck equation for a nonstationary Markov process:

$$\begin{aligned} \frac{\partial P(q, t)}{\partial t} &= -\frac{\partial}{\partial q} V(q, t)P(q, t) + \gamma(t) \frac{\partial}{\partial q} g(q, t) \frac{\partial}{\partial q} g(q, t)P(q, t) \\ &= -\frac{\partial}{\partial q} \left[V(q, t) + \gamma(t) g(q, t) \frac{\partial g(q, t)}{\partial q} \right] P(q, t) \\ &\quad + \gamma(t) \frac{\partial^2}{\partial q^2} g^2(q, t)P(q, t) \end{aligned} \tag{A11}$$

In the last step we have obtained the spurious drift $\gamma(t)g(q, t)\,\partial g(q, t)/\partial q$ that appears when transforming a Stratonovich stochastic differential equation to its equivalent Itô equation.[27]

The second case in which (A8) can be exactly reduced to a Fokker–Planck equation is the case of a linear Langevin equation (A1):

$$V(q, t) = a(t)q + b(t), \qquad g(q, t) = 1 \tag{A12}$$

In these circumstances

$$q(t) = \left\{\exp\left[\int_0^t a(\tau)\,d\tau\right]\right\}\left(q(0) + \int_0^t d\tau\left\{\exp\left[-\int_0^\tau a(\tau')\,d\tau'\right]\right\}[b(\tau) + \zeta(\tau)]\right. \tag{A13}$$

and so

$$\delta q(t)/\delta\zeta(t') = \exp\left[\int_{t'}^t a(\tau)\,d\tau\right], \qquad t > t' \tag{A14}$$

Therefore (A8) becomes

$$\frac{\partial P(q, t)}{\partial t} = -\frac{\partial}{\partial q}[a(t)q + b(t)]P(q, t) + \frac{\partial^2}{\partial q^2}D(t)P(q, t) \tag{A15}$$

$$D(t) = \int_0^t dt'\,\gamma(t, t')\exp\left[\int_{t'}^t a(\tau)\,d\tau\right] \tag{A16}$$

It is worth mentioning that this result also can be applied to the whole class[28] of Fokker–Planck equations that can be transformed to linear form.

In a related context Adelman[18] and Fox[16,19] have also obtained Fokker–Planck equations for linear Langevin equations with colored noise under the restriction of the existence of a fluctuation-dissipation relation. Adelman obtains the equation from the knowledge of the solution and Fox uses methods which we believe rather complicated, at least for the case of Gaussian noise in which we are interested. A much more general treatment of the nonlinear problem, including non-Gaussian forces, is also given in Ref. 20. Some useful formulas for evaluating the quantity $\langle\zeta(t)\,\delta(q(t) - q)\rangle$ are given in Ref. 29 for a special form of $\gamma(t, t')$.

APPENDIX B

In this appendix we evaluate the quantity $\delta q(t)/\delta\zeta(t')$ appearing in (A8) and its time derivatives used in Section 3. The force $\zeta(t)$ is formally considered in the following as a given function of time and no average over its probability distribution is performed.

Given Eq. (A1), $\delta q(t)/\delta\zeta(t')$ may be interpreted as the nonaveraged, nonlinear response function to the external force $\zeta(t)$. The MSR formalism[30-32] is the formalism best suited to deal with such a response function for classical phenomenological equations. According to it, the following operator equations of motion[31,32] are equivalent to (A1) if an average over initial condition is made at the end of the calculations:

$$\dot{q}(t) = [L(q(t), \hat{q}(t), t), q(t)] \tag{B1}$$

$$\dot{\hat{q}}(t) = [L(q(t), \hat{q}(t), t), \hat{q}(t)] \tag{B2}$$

$$[\hat{q}(t), q(t)] = 1 \tag{B3}$$

$$\hat{q}(0) = \partial/\partial q(0) \tag{B4}$$

where $\hat{q}(t)$ is defined by Eqs. (B2)–(B4), the square bracket stands for a commutator, and

$$L(q(t), \hat{q}(t), t) = \{V(q(t), t) + g(q(t), t)\zeta(t)\}\hat{q}(t) \tag{B5}$$

Going to an interaction picture[31,32] and denoting by $q^0(t)$ the operators whose evolution is governed by $L^0(q, \hat{q}, t) = v(q, t)\hat{q}$, one has

$$q(t) = S(0, t)q^0(t)S(t, 0) \tag{B6}$$

$$\hat{q}(t) = S(0, t)\hat{q}^0(t)S(t, 0) \tag{B7}$$

$$S(0, t) = \bar{T}\exp\left\{\int_0^t dt'\, g(q^0(t'), t')\zeta(t')\hat{q}^0(t')\right\} \tag{B8}$$

where $\bar{T}$ denotes time antiordering.

From Eq. (B6)

$$\frac{\delta q(t)}{\delta\zeta(t')} = \left[\frac{\delta S(0, t)}{\delta\zeta(t')} S(t, 0), q(t)\right] \tag{B9}$$

Since

$$\delta S(0, t)/\delta\zeta(t') = \theta(t - t')S(0, t')g(q^0(t'), t')\hat{q}^0(t')S(t', t) \tag{B10}$$

one finally has

$$\delta q(t)/\delta\zeta(t') = \theta(t - t')[g(q(t'))\hat{q}(t'), q(t)] \tag{B11}$$

which is the operator expression for the response function.[32] In the limit $t' \to t^-$,

$$\delta q(t)/\delta\zeta(t) = g(q(t), t) \tag{B12}$$

The derivative with respect to t' of $\delta q(t)/\delta\zeta(t')$ is easily evaluated once its expression (B11) and the equations of motion (B1)–(B2) are given.

Assuming for simplicity from now on that neither $v(q(t))$ nor $g(q(t))$ depends explicitly on time, as is the case in Section 3, we have for $t > t'$

$$\begin{aligned}\frac{d}{dt'}\frac{\delta q(t)}{\delta\zeta(t')} &= \left[\frac{d}{dt'}g(q(t'))\hat{q}(t'), q(t)\right] \\ &= [[L(q(t'),\hat{q}(t'),t'), g(q(t'))\hat{q}(t')], q(t)] \\ &= -[V'(q(t'))g(q(t'))\hat{q}(t'), q(t)] + [V(q(t'))g'(q(t'))\hat{q}(t'), q(t)]\end{aligned} \tag{B13}$$

where we have used the fact that[31]

$$[\hat{q}(t), f(q(t))] = f'(q(t)) \tag{B14}$$

and $f'(q)$ denotes the derivative of f with respect to q. In the limit $t' \to t$

$$\left.\frac{d}{dt'}\frac{\delta q(t)}{\delta\zeta(t')}\right|_{t'=t} = -g(q(t))V'(q(t)) + V(q(t))g'(q(t)) \tag{B15}$$

Higher order derivatives are analogously evaluated. For example,

$$\begin{aligned}&\frac{d^2}{dt'^2}\frac{\delta(q(t))}{\delta\zeta(t')} \\ &\quad= -[[L(q(t'),\hat{q}(t'),t'), V'(q(t'))g(q(t'))\hat{q}(t')], q(t)] \\ &\qquad+ [[L(q(t'),\hat{q}(t'),t'), V(q(t')g'(q(t'))\hat{q}(t')], q(t)] \\ &\quad= \left[V^2(q(t'))\left\{V(q(t'))\left[\frac{g(q(t'))}{V(q(t'))}\right]'\right\}'\hat{q}(t')\right. \\ &\qquad\left. - g^2(q(t'))\left\{g(q(t'))\left[\frac{V(q(t'))}{g(q(t'))}\right]'\right\}'\zeta(t')\hat{q}(t'), q(t)\right]\end{aligned} \tag{B16}$$

In the limit $t' \to t$

$$\begin{aligned}\left.\frac{d^2}{dt'^2}\frac{\delta q(t)}{\delta\zeta(t')}\right|_{t'=t} &= V^2(q(t))\left\{V(q(t))\left[\frac{g(q(t))}{V(q(t))}\right]'\right\}' \\ &\quad - g^2(q(t))\left\{g(q(t))\left[\frac{V(q(t))}{g(q(t))}\right]'\right\}'\zeta(t)\end{aligned} \tag{B17}$$

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