Langevin equation for the Rayleigh model with finite-range interactions

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Both linear and nonlinear Langevin equations are derived directly from the Liouville equation for an exactly solvable model consisting of a Brownian particle of mass M interacting with ideal gas molecules of mass m via a quadratic repulsive potential. Explicit microscopic expressions for all kinetic coefficients appearing in these equations are presented. It is shown that the range of applicability of the Langevin equation, as well as statistical properties of random force, may depend not only on the mass ratio m/M but also on the parameter Nm/M, involving the average number N of molecules in the interaction zone around the particle. For the case of a short-ranged potential, when $N \ll 1$, analysis of the Langevin equations yields previously obtained results for a hard-wall potential in which only binary collisions are considered. For the finite-ranged potential, when multiple collisions are important ($N \gg 1$), the model describes nontrivial dynamics on time scales that are on the order of the collision time, a regime that is usually beyond the scope of more phenomenological models.

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I. INTRODUCTION

The explicit derivation from first principles of the Langevin equation, describing the evolution of a small number of variables in a complex system, is often necessary since in many cases the statistical properties of the random force, the range of applicability and even the form of the equation are far from evident [1]. The need for microscopic considerations is especially compelling in the case of a nonlinear Langevin equation where the usual phenomenological approach of adding stochastic terms to the deterministic nonlinear equation describing the relaxation of targeted variables may be inadequate [2]. Although statistical properties of the random force in the nonlinear Langevin equation may be deduced phenomenologically in some cases [3], it is generally necessary to start from the microscopic behavior of the system in order to construct the appropriate form of the equation.

The conventional systematic method for deriving the Langevin equation (LE) for a Brownian particle exploits Mori's projection-operator techniques [1], which allow the transformation of the microscopic Liouville equation to a non-Markovian predecessor of the LE, generally known as the generalized Langevin equation. The LE can be then obtained by a subsequent perturbation expansion of the memory kernel appearing in the generalized Langevin equation using the square root ratio of the mass of a bath particle to that of the Brownian particle, $(m/M)^{1/2}$, as a perturbation parameter λ. While the first step in the derivation of the LE, involving rearrangement of the Liouville equation with projection operator methods, is an exact algebraic procedure, the question of convergence of the λ expansion in the second step is subtle and can be strictly justified only under assumptions that are difficult to prove in general [4]. Mazur and Oppenheim developed an alternative projection-operator approach more suitable to analyze the convergence of the λ expansion [5]. The validity of the perturbation analysis in both the Mori and the Mazur-Oppenheim approach has been examined for an exactly solvable model consisting of tagged particle motion in a harmonic lattice [6]. Unfortunately, the convergence properties of the λ series are trivial for this model, since all terms in the λ expansion higher than zeroth order vanish. As a result, the memory kernel for the linear damping force in the LE does not depend on the mass ratio, and all nonlinear terms are identically zero [7].

One goal of this paper is to present and analyze a simplified model that serves as a useful and nontrivial testing ground for examining some of the subtle aspects of the theory of Brownian motion. The model considered here is a generalization of the well-known Rayleigh model of a Brownian particle of mass M constrained to move in one dimension and subjected to collisions with an equilibrium ideal (noninteractive) gas of particles of mass m. The Rayleigh model is perhaps the oldest model of nonequilibrium statistical physics [8], and has attracted much attention over the years, with early work [9–11] focusing on the model as a test of the systematic derivation of macroscopic kinetic equations for the heavy particle from the master equation. More recent investigations have examined the stationary and transient solutions of the asymmetric Rayleigh model in which the thermodynamic parameters characterizing the gas to the left and right of the piston differ [12]. In all these studies, the interaction between the Brownian particle and the bath is assumed to be short ranged with a negligible collision time τ_c. Only binary collisions are considered in this model because the range of interaction is assumed to be short compared to the average distance between bath particles. The usual starting point for analysis of the Rayleigh model with binary collisions is a Markovian master equation for particle's velocity distribution function. The master equation is only an approximate form of the fully microscopic Liouville equation, and valid only for time scales longer than τ_c. To test many aspects of the theory of Brownian motion, it is essential to start from a fully microscopic description of the dynamics directly from the Liouville equation so that any non-Markovian character of bath correlations is properly incorporated and particle dynamics on time scales less than τ_c can be described. To this end, one may generalize the interaction between the bath and Brownian particles from a hard-wall to a parabolic repulsive potential. The generalized

model is analytically solvable, allowing explicit calculation of all terms appearing in the derivation of both linear and nonlinear LE beyond the binary collision approximation. We demonstrate that in addition to the small parameter $\lambda = (m/M)^{1/2}$, the character of the dynamics of a tagged particle can be governed by an additional parameter, $N^{1/2}\lambda$, involving the average number N of bath particles simultaneously interacting with the Brownian particle. For a large Brownian particle, it is shown that when $N \gg 1$ and multiple collisions are important, the parameter of the formal λ expansion is actually $N^{1/2}\lambda$.

The paper is organized as follows. In Sec. II, the Mazur-Oppenheim approach is reviewed to provide groundwork for all subsequent analysis. In Sec. III, the structure of terms in the λ expansion is examined and presented in a convenient form. In Sec. IV, the general formalism is applied to the Rayleigh model with a quadratic repulsive potential describing bath-Brownian particle interactions, and the LE is derived for the heavy particle. The nonlinear LE is obtained in Sec. V and various aspects of this equation are discussed. Finally, a few concluding remarks are made in Sec. VI.

II. BASIC EQUATIONS

The Hamiltonian for a Brownian system composed of a tagged particle of mass M in a bath of point particles of mass m is

$$H = \frac{P^2}{2M} + H_0, \tag{1}$$

$$H_0 = \sum_i \frac{p_i^2}{2m} + U(x,X), \tag{2}$$

where $x = \{x_i\}$ and p_i are positions and momenta of bath particles, X and P are those of the Brownian (or tagged) particle, and H_0 is the Hamiltonian of the bath in the field of the tagged particle fixed at X. One can expect that on average $P \sim \sqrt{Mk_BT}$, where k_B is Boltzmann's constant and T is the temperature, and that the scaled momentum $P_* = \lambda P$, where $\lambda = \sqrt{m/M}$, is of the same order as the average momentum of a bath particle. In terms of scaled momentum, one can write the Liouville operator as

$$\mathcal{L} = \mathcal{L}_0 + \lambda \mathcal{L}_1, \tag{3}$$

$$\mathcal{L}_0 = \sum_i \left\{ \frac{p_i}{m} \frac{\partial}{\partial x_i} + F_i \frac{\partial}{\partial p_i} \right\}, \tag{4}$$

$$\mathcal{L}_1 = \frac{P_*}{m} \frac{\partial}{\partial X} + F \frac{\partial}{\partial P_*}, \tag{5}$$

where $F_i = -\nabla_{x_i} U$ and $F = -\nabla_X U$ are the forces on the ith bath particle and on the Brownian particle, respectively. The operator $\mathcal{L}_0$ dictates the dynamics of the bath in the field of the fixed Brownian particle.

If the mass of the tagged particle is large (i.e., a Brownian particle), one might intuitively expect that inertial effects of

the particle's motion are small and that the force on the particle, $F(t) = e^{\mathcal{L}t}F$, is close to the pressure force, i.e., to the force on the fixed tagged particle,

$$F_0(t) \equiv e^{\mathcal{L}_0 t} F. \tag{6}$$

In the Mazur-Oppenheim approach [5], the force $F(t)$ is decomposed using the projection operator $\mathcal{P}$ which averages a dynamical variable A over the canonical distribution $\rho = Z^{-1} \exp(-\beta H_0)$, for bath variables at fixed position of the tagged particle,

$$\mathcal{P}A = \langle A \rangle \equiv \int \rho A \prod_i dx_i \, dp_i, \tag{7}$$

where Z is the canonical partition function and $\beta = 1/k_BT$. Using the operator identity [13]

$$e^{(\mathcal{A}+\mathcal{B})t} = e^{\mathcal{A}t} + \int_0^t d\tau \, e^{\mathcal{A}(t-\tau)} \mathcal{B} e^{(\mathcal{A}+\mathcal{B})\tau}, \tag{8}$$

with $\mathcal{A} = \mathcal{L}$ and $\mathcal{B} = -\mathcal{P}\mathcal{L}$, one may formally decompose the force $F(t)$ on the tagged particle into a "random" part and a remainder as

$$F(t) = F^\dagger(t) + \int_0^t d\tau \, e^{\mathcal{L}(t-\tau)} \mathcal{P}\mathcal{L}F^\dagger(\tau), \tag{9}$$

where $F^\dagger(t) = e^{\mathcal{Q}\mathcal{L}t}F$ and $\mathcal{Q} = 1 - \mathcal{P}$. The factor $\mathcal{P}\mathcal{L}F^\dagger(\tau)$ in the integral in Eq. (9) can be simplified taking into account the orthogonality of $\mathcal{P}$ and $\mathcal{L}_0$ (i.e., $\mathcal{P}\mathcal{L}_0 = 0$), and the equality

$$\langle \nabla_X F^\dagger(t) \rangle = -\beta \langle F_0 F^\dagger(t) \rangle, \tag{10}$$

which can be derived by integration by parts. As a result, one obtains the following exact equation of motion for the scaled momentum of the tagged particle:

$$\frac{dP_*(t)}{dt} = \lambda F^\dagger(t) + \lambda^2 \int_0^t d\tau \, e^{\mathcal{L}(t-\tau)} \left(\nabla_{P_*} - \frac{\beta}{m} P_* \right)$$
$$\times \langle F F^\dagger(\tau) \rangle, \tag{11}$$

where $F^\dagger(t)$ is a zero-centered random force obeying $\langle F^\dagger(t) \rangle = \mathcal{P} e^{\mathcal{Q}\mathcal{L}t} F = 0$.

The random force $F^\dagger(t) = e^{(\mathcal{L}_0 + \lambda \mathcal{Q}\mathcal{L}_1)t} F$ can be further expanded in terms of the mass ratio parameter λ using identity (8) to obtain

$$F^\dagger(t) = F_0(t) + \lambda \int_0^t dt_1 \, e^{\mathcal{L}_0(t-t_1)} \mathcal{Q}\mathcal{L}_1 F_0(t_1) + \lambda^2 \int_0^t dt_1$$
$$\times \int_0^{t_1} dt_2 \, e^{\mathcal{L}_0(t-t_1)} \mathcal{Q}\mathcal{L}_1 e^{\mathcal{L}_0(t_1-t_2)} \mathcal{Q}\mathcal{L}_1 F_0(t_2) + \cdots. \tag{12}$$

Similarly, the kernel $K(t) = \langle F F^\dagger(t) \rangle$ appearing in the exact equation of motion (11) of the tagged particle may be expanded in a power series in λ,

$$K(t) = \langle FF^{\dagger}(t) \rangle = \sum_l K_l(t), \qquad (13)$$

$$K_0(t) = \lambda^0 \langle FF_0(t) \rangle,$$

$$K_1(t) = \lambda^1 \int_0^t dt_1 C_1(t,t_1),$$

$$K_2(t) = \lambda^2 \int_0^t dt_1 \int_0^{t_1} dt_2 C_2(t,t_1,t_2) \ldots,$$

where the correlation functions C_l are defined to be

$$C_1(t_0,t_1) = \langle F(e^{\mathcal{L}_0(t_0-t_1)}\mathcal{Q}\mathcal{L}_1)F_0(t_1) \rangle, \qquad (14)$$

$$C_2(t_0,t_1,t_2) = \langle F(e^{\mathcal{L}_0(t_0-t_1)}\mathcal{Q}\mathcal{L}_1)(e^{\mathcal{L}_0(t_1-t_2)}\mathcal{Q}\mathcal{L}_1)F_0(t_2) \rangle,$$

$$C_l(t_0,t_1,\ldots,t_l) = \left\langle F\left(\prod_{i=1}^l e^{\mathcal{L}_0(t_{i-1}-t_i)}\mathcal{Q}\mathcal{L}_1\right)F_0(t_l) \right\rangle$$

The truncation of the λ expansion to zeroth order, $K(t) \approx K_0(t)$, leads from Eq. (11) directly to the generalized Langevin equation

$$\frac{dP_*(t)}{dt} = \lambda F^{\dagger}(t) - \lambda^2 \int_0^t d\tau \, M_0(\tau) P_*(t-\tau), \qquad (15)$$

where

$$M_0(t) = \frac{\beta}{m} K_0(t) = \frac{\beta}{M} \langle FF_0(t) \rangle. \qquad (16)$$

This approximation is sensible provided the correlation functions C_l appearing at higher order in the λ-expansion decay on a similar λ-independent time scale τ_c characteristic of motions of the fixed-particle system (i.e., governed by $\mathcal{L}_0$). Mazur and Oppenheim [5] succeeded in proving that this is the case assuming the factorization properties

$$\langle A(t_1)e^{\mathcal{L}_0 t}B(t_2) \rangle \xrightarrow{t>\tau_c} \langle A(t_1) \rangle \langle B(t_2) \rangle.$$

While being formally nonlocal in time, Eq. (15) can actually be written in a form that is local in time by expanding $P_*(t-\tau)$ around $\tau=0$ to obtain

$$\frac{dP_*(t)}{dt} = \lambda F^{\dagger}(t) - \lambda^2 \gamma_0(t) P_*(t), \qquad (17)$$

where $\gamma_0(t) = \int_0^t d\tau \, M_0(\tau)$. The nonlocal correction terms to this approximation are of the form [14]

$$\lambda^2 \int_0^t d\tau \, M_0(\tau) \int_{t-\tau}^t d\tau' \, \dot{P}_*(\tau') \sim \lambda^3, \qquad (18)$$

and are therefore of higher order in λ. Naturally, this analysis is pertinent only if the characteristic time τ_c for decay of $M_0(t)$ does not depend on λ.

The local equation (17) is applicable on arbitrary time scales and for $t \gg \tau_c$ assumes the form of the conventional LE with a time-independent damping coefficient $\gamma_0 = \int_0^\infty dt \, M_0(t)$. It is then evident from Eq. (17) that the autocorrelation function of the momentum of a heavy particle decays on a time scale $\tau_p \sim \lambda^{-2}$ that is much longer than the characteristic time of the bath $\tau_c \sim \lambda^0$. One can therefore expect the local form of the LE with time-independent damping to be a good approximation for Eq. (17) except at short times determined by $t < \tau_c$.

It will be shown below that for homogeneous bath $K_1(t)=0$, so the next approximation for the λ expansion (13) is of the form $K(t) \approx K_0(t) + K_2(t)$. The equation of motion for the momentum of the tagged particle in this case includes a nonlinear damping term of third order in P_*. This equation will be considered in Sec. V.

III. STRUCTURE OF THE TERMS IN λ EXPANSION

To examine the convergence properties of the λ expansion and other features of the projection-operator derivation of the LE, the structure of the correlation functions C_l defined in Eq. (14) which appear in the expansion of the memory function must be analyzed. Although only the functions C_0 and C_2 are needed to obtain the nonlinear LE to lowest order in λ, it is useful to know general properties of C_l.

By inspection of the symmetry properties of the system, it is immediately apparent that the correlation functions C_{2n+1} corresponding to odd powers of λ contain an odd number of factors F and ∇_X, and therefore vanish for isotropic systems. In fact, in the absence of external field, the dependence on the particle coordinate appears only through the difference $x_i - X$, and it is useful to introduce new variables $q_i = x_i - X$. Since the vectors $F = -\nabla_X U = \Sigma_i \nabla_{q_i} U$ and $\nabla_X = -\Sigma_i \nabla_{q_i}$ have negative parity and the Hamiltonian H_0 is invariant with respect to transformation $\{q_i \to -q_i, p_i \to -p_i\}$, the correlation functions $C_{2n+1}(t,t_1,\ldots,t_{2n+1})$ vanish.

Correlation functions of even orders C_{2n} do not vanish and have a rather complicated structure. For notational simplicity, we restrict the analysis to the case of one-dimensional diffusion, expecting no physical features in higher dimensions. For future development, it is convenient to define

$$G_0(t) = F_0(t),$$

$$G_1(t,t_1) = S(t-t_1)F_0(t_1),$$

$$G_2(t,t_1,t_2) = S(t-t_1)S(t_1-t_2)F_0(t_2),$$

$$G_s(t,t_1,\ldots,t_s) = S(t-t_1)\cdots S(t_{s-1}-t_s)F_0(t_s), \qquad (19)$$

where

$$S(t_i-t_l) = e^{\mathcal{L}_0(t_i-t_l)} \frac{\partial}{\partial X}. \qquad (20)$$

Note the property

$$\langle G_{i_1} G_{i_2} \cdots G_{i_l} \rangle = 0 \qquad (21)$$

which holds for arbitrary time arguments when $i_1 + i_2 + \cdots + i_l$ and l have different parities. For example, $\langle G_i \rangle$ and $\langle G_0 G_0 G_i \rangle$ vanish for even i, while $\langle G_0 G_i \rangle$ and $\langle G_0 G_0 G_0 G_i \rangle$ are zero for odd i.

Using the definitions above and according to Eqs. (14), the second-order correlation function C_2 can be written as

$$C_2(t, t_1, t_2) = \left(\frac{P_*}{m} \right)^2 \langle G_0 G_2(t, t_1, t_2) \rangle$$

$$+ \frac{1}{m} \langle G_0 G_0(t - t_1) G_1(t, t_2) \rangle$$

$$- \frac{1}{m} \langle G_0 G_0(t - t_1) \rangle \langle G_1(t, t_2) \rangle. \qquad (22)$$

Using cumulants, denoted by $\langle\langle \cdots \rangle\rangle$ and defined via the relations

$$\langle A \rangle = \langle\langle A_1 \rangle\rangle,$$

$$\langle A_1 A_2 \rangle = \langle A_1 \rangle \langle A_2 \rangle + \langle\langle A_1 A_2 \rangle\rangle,$$

$$\langle A_1 A_2 A_3 \rangle = \langle A_1 \rangle \langle A_2 \rangle \langle A_3 \rangle + \langle A_1 \rangle \langle\langle A_2 A_3 \rangle\rangle + \langle A_2 \rangle \langle\langle A_3 A_1 \rangle\rangle$$

$$+ \langle A_3 \rangle \langle\langle A_2 A_1 \rangle\rangle + \langle\langle A_1 A_2 A_3 \rangle\rangle,$$

$$\vdots$$

the function C_2 can be written as

$$C_2(t, t_1, t_2) = \left(\frac{P_*}{m} \right)^2 \langle\langle G_0 G_2(t, t_1, t_2) \rangle\rangle$$

$$+ \frac{1}{m} \langle\langle G_0 G_0(t - t_1) G_1(t, t_2) \rangle\rangle. \qquad (23)$$

Note that the zeroth-order kernel K_0 is also a cumulant, $K_0 = \langle F F_0(t) \rangle = \langle\langle G_0 G_0(t) \rangle\rangle$. The relevance of cumulant representation for C_l follows from the fact that one expects the cumulants to have similar scaling properties with respect to parameters of the system independent of their order. For example, it will be established in the following section that cumulants $\langle\langle G_{i_1} G_{i_2} \cdots G_{i_l} \rangle\rangle$ of any order are linear functions of the average number N of particles in the interaction zone around the particle for the Rayleigh model. Therefore, for this model, the first two nonvanishing terms in the λ expansion (13), namely, K_0 and K_2, are both linear in N.

Consider the correlation functions C_{2l} with $l \geq 1$. It is tedious, though not difficult to establish that these correlation functions are of order l in cumulants $\langle\langle G_{i_1} G_{i_2} \cdots G_{i_l} \rangle\rangle$. Note that C_{2l} has the contributions that are of order $l + 2$ or less in G_i. First consider the terms of maximal order $l + 2$: One type of these terms are the correlation functions of the form

$$\langle G_0 G_0 G_{i_1} G_{i_2} \cdots G_{i_l} \rangle, \qquad (24)$$

where here and below time arguments have been omitted for brevity. The indices $i_1, \ldots, i_l$ may take any values from the set $\{0, 1, \ldots, l\}$ provided

$$i_1 + i_2 + \cdots + i_l = l. \qquad (25)$$

For example, for $l = 1$ the correlation function of maximal order in G_i which contributes to C_2 is $\langle G_0 G_0 G_1 \rangle$, as can be seen from Eq. (23). For $l = 2$ the function C_4 includes contributions of the forms $\langle G_0 G_0 G_0 G_2 \rangle$ and $\langle G_0 G_0 G_1 G_1 \rangle$, and so on.

All other contributions of order $l + 2$ in G_i can be written as products of correlation functions of lower orders which can be obtained dividing the sequences

$$G_0(\tau_1) G_0(\tau_2) G_{i_1}(\tau_3) G_{i_2}(\tau_4) \cdots G_{i_l}(\tau_{l+2}) \qquad (26)$$

into groups in all possible ways without permuting functions. For example, for $l = 2$ the three remaining terms are

$$\langle G_0(\tau_1) G_0(\tau_2) \rangle \langle G_0(\tau_3) G_2(\tau_4) \rangle,$$

$$\langle G_0(\tau_1) G_0(\tau_2) \rangle \langle G_1(\tau_3) G_1(\tau_4) \rangle,$$

$$\langle G_0(\tau_1) G_0(\tau_2) G_1(\tau_3) \rangle \langle G_1(\tau_4) \rangle,$$

$$\langle G_0(\tau_1) G_0(\tau_2) \rangle \langle G_1(\tau_3) \rangle \langle G_1(\tau_4) \rangle.$$

Clearly, the terms of maximal order in cumulants correspond to the case when all indices i_s in Eqs. (24) and (25) are equal to 1. Then the cumulant expansion of all such terms contains the contribution

$$\langle G_0 G_0 \rangle \langle G_1 \rangle \langle G_1 \rangle \cdots \langle G_1 \rangle, \qquad (27)$$

which contains l factors $\langle G_1(t_i) \rangle$ and, therefore, is of order $l + 1$ in cumulants. However, all terms of order $l + 2$ in G_i described above enter in C_{2l} via combinations

$$\langle G_0 G_0 G_{i_1} G_{i_2} \cdots G_{i_l} \rangle - \langle G_0 G_0 \rangle \langle G_{i_1} G_{i_2} \cdots G_{i_l} \rangle,$$

$$\langle G_0 G_0 G_{i_1} \rangle \langle G_{i_2} \cdots G_{i_l} \rangle - \langle G_0 G_0 \rangle \langle G_{i_1} \rangle \langle G_{i_2} \cdots G_{i_l} \rangle,$$

$$\langle G_0 G_0 G_{i_1} G_{i_2} \rangle \langle G_{i_3} \cdots G_{i_l} \rangle$$

$$- \langle G_0 G_0 \rangle \langle G_{i_1} G_{i_2} \rangle \langle G_{i_3} \cdots G_{i_l} \rangle, \ldots,$$

which is a consequence of the presence of the operator $\mathcal{Q}$ (first from the left) in the definition of C_i [see Eq. (14)]. As a result, contributions (27) involving $l + 1$ cumulant factors cancel, and the maximal order in cumulants of surviving terms in the expression for C_{2l} contain at most l cumulant factors.

Having established that the contributions to C_{2l} from terms of maximal order $l + 2$ in G_i are of order l in cumulants, it is clear that the terms of order $l + 1$ and lower in G_i cannot contain more than l cumulant factors since such terms contain at least one isolated factor of G_0 whose average vanishes. Hence C_{2l} contains *at most* l cumulant factors.

In the following section, we examine the consequences of the cumulant expansions of the memory function for a spe-

cific system, namely, the Rayleigh model with a repulsive parabolic potential. It will be demonstrated that for the Rayleigh model, cumulants are linear functions of the average number N of particles in the interaction zone around the particle. This, in turn, implies that the terms in the λ expansion behave as

$$K_{2l} \sim (N + N^2 + \cdots + N^l)\lambda^{2l} \qquad (28)$$

for $l > 1$. Clearly, for a large particle and/or long-ranged potential leading to $N \gg 1$, these results suggest that $K_{2l} \sim N^l \lambda^{2l}$, demonstrating that the actual parameter of λ expansion for this exactly solvable model is in fact $N^{1/2}\lambda$. On the other hand, for a short-ranged potential, when $N \ll 1$, all terms in the expansion are linear in N, and one sees that $K_{2l} \sim N\lambda^{2l}$, and the effective small parameter of λ expansion is in fact the square root of the mass ratio. To further illustrate this analysis, an explicit form of the cumulant expansion of C_4 is presented in Appendix A.

IV. THE LINEAR LANGEVIN EQUATION FOR A HEAVY PARTICLE OR IDEALIZED PISTON

Consider the random motion of a piston of mass M and cross-sectional area S subjected to collisions with an ideal gas particles of mass m. The gas particles and the piston are constrained to move in one dimension perpendicular to the piston faces. The velocity distribution of incident particles $f_M(v)$ before collision with the piston is Maxwellian with inverse temperature β, namely,

$$f_M(v) = \left(\frac{m\beta}{2\pi}\right)^{1/2} \exp\left(-\frac{1}{2}\beta m v^2\right). \qquad (29)$$

The piston-particle interaction is assumed to be described by a purely repulsive parabolic potential. For particles to the left of the piston the interaction potential between a gas particle and the piston is

$$U_l = \begin{cases} \frac{1}{2}k_f(x - X_l)^2, & x > X_l \\ 0, & x < X_l, \end{cases} \qquad (30)$$

where k_f is a force constant, x is the coordinate of the gas particle, $X_l = X_{lf} - a$ the boundary of the piston-particle interaction zone, X_{lf} is the coordinate of the left face of the piston, and a is the width of the interaction zone. Similarly, the gas particle–piston potential for the particles to the right of the piston has the analogous form

$$U_r = \begin{cases} \frac{1}{2}k_f(x - X_r)^2, & x < X_r \\ 0 & x > X_r, \end{cases} \qquad (31)$$

where $X_r = X_{rf} + a$, and X_{rf} is the position of the piston's right face. We assume that the temperature is low enough (or k_f is sufficiently large) so that the probability for a particle to reach the piston's surface is negligible.

In the previous sections it was established that the dynamics of the piston can be deduced from time correlation functions describing molecular motion in the field of the piston

fixed in space. Below we derive the explicit expressions for these correlation functions in the thermodynamic limit, neglecting recollisions of the piston and gas particles due to the finite size of the bath.

Consider the force on the left side of the fixed piston,

$$F_l(t) = -k_f \sum_i q_i(t)\theta(q_i(t)), \qquad (32)$$

where $q_i = x_i - X_l$ is the position of gas particle i relative to the boundary of the interaction zone, $\theta(x)$ is the step function, and summation over index i is over all particles in the tube of diameter S to the left of the piston. In this section we omit for brevity the subscript 0 for the force on the fixed piston. The simplifying feature of parabolic potential is that the time τ_c that a particle spends in the interaction zone of the fixed piston is independent of the initial velocity of the particle and is given by $\tau_c = \pi/\omega$, where $\omega = \sqrt{k_f/m}$. At a given time t, the only gas particles in the interaction zone are those that had positive velocities and coordinates q in the interval $-v\tau_c < q < 0$ at time $t - \tau_c$. At time t, the position of the gas particle is determined by $q(t) = (v/\omega)\sin \omega(t - t_{in})$, where $t_{in} = t - \tau_c - q/v$ corresponds to the time at which the gas particle enters the interaction region and q is the position of the gas particle at time $t - \tau_c$. It then follows that $q(t) = (v/\omega)\sin \omega(\tau_c + q/v) = -(v/\omega)\sin \omega q/v$, which implies that the total instantaneous force on the left side of the fixed piston at time $t > 0$ can be written as

$$F_l(t) = -k_f \int_0^\infty dv \int_{-v\tau_c}^0 dq \, N(X_l + q, v; t - \tau_c)\frac{v}{\omega}\sin\frac{\omega q}{v}. \qquad (33)$$

In Eq. (33), $q = x - X_l$ and $N(x,v;t)$ is the microscopic linear density of particles defined by

$$N(x,v;t) = \sum_i \delta(x - x_i(t))\delta(v - v_i(t)). \qquad (34)$$

Similarly, the total instantaneous force acting on the right side of the piston is

$$F_r(t) = -k_f \int_{-\infty}^0 dv \int_0^{-v\tau_c} dq \, N(X_r + q, v; t - \tau_c)\frac{v}{\omega}\sin\frac{\omega q}{v}, \qquad (35)$$

where $q = x - X_r$.

For a particle outside the interaction zone of the fixed piston (i.e., for $x < X_l$, and $x > X_r$), the average linear density of particles is $\langle N(x,v) \rangle = nSf_M(v)$, where n is the total (three-dimensional) density of bath particles, and S is the cross-sectional area of the piston. It then follows from Eqs. (33) and (35) that the average force acting on the left $\langle F_l \rangle$ and the right $\langle F_r \rangle$ side of the piston is $\langle F_l \rangle = -\langle F_r \rangle = nS/\beta$.

It is straightforward to show that the stationary distribution in the vicinity of the fixed piston, including the interaction zone, assumes Boltzmann's form

$$\langle N(x,v)\rangle = nSf_M(v)\exp[-U(x)/k_BT]. \qquad (36)$$

To calculate the force correlation functions required to analyze the damping terms in the LE, correlation functions of the form $\langle N(Y_1)N(Y_2)\cdots N(Y_s)\rangle$ must be evaluated, where Y denotes the position-velocity pair (x,v). It is sufficient to consider only the case when time arguments are equal for all functions, since time displacement can be transformed into spatial displacement for a free particle, i.e.,

$$N(x,v;t+t_1)=N(x-vt_1,v;t). \qquad (37)$$

Note that the product $N(Y_1)N(Y_2)$ can be written as

$$N(Y_1)N(Y_2)=\sum_{i,j}\delta(Y_1-Y_i)\delta(Y_2-Y_j)$$

$$=\sum_i\delta(Y_1-Y_i)\delta(Y_2-Y_i)$$

$$+\sum_{i\neq j}\delta(Y_1-Y_i)\delta(Y_2-Y_j).$$

Since $\delta(Y_1-Y_i)\delta(Y_2-Y_i)=\delta(Y_1-Y_2)\delta(Y_1-Y_i)$, for the ideal gas system one obtains

$$\langle N(Y_1)N(Y_2)\rangle = \delta(Y_1-Y_2)\langle N(Y_1)\rangle + \langle N(Y_1)\rangle\langle N(Y_2)\rangle. \qquad (38)$$

For the three-point correlation function, the same arguments lead to the result

$$\langle N(Y_1)N(Y_2)N(Y_3)\rangle = \delta(Y_1-Y_2)\delta(Y_2-Y_3)\langle N(Y_1)\rangle$$

$$+\delta(Y_1-Y_2)\langle N(Y_1)\rangle\langle N(Y_3)\rangle$$

$$+\delta(Y_1-Y_3)\langle N(Y_1)\rangle\langle N(Y_2)\rangle$$

$$+\delta(Y_2-Y_3)\langle N(Y_1)\rangle\langle N(Y_2)\rangle$$

$$+\langle N(Y_1)\rangle\langle N(Y_2)\rangle\langle N(Y_3)\rangle. \qquad (39)$$

Equations (38) and (39) are the cumulant expansions of $\langle N(Y_1)N(Y_2)\rangle$ and $\langle N(Y_1)N(Y_2)N(Y_3)\rangle$, where the cumulants

$$\langle\!\langle N(Y_1)N(Y_2)\rangle\!\rangle = \delta(Y_1-Y_2)\langle N(Y_1)\rangle,$$

$$\langle\!\langle N(Y_1)N(Y_2)N(Y_3)\rangle\!\rangle = \delta(Y_1-Y_2)\delta(Y_2-Y_3)\langle N(Y_1)\rangle, \qquad (40)$$

$$\langle\!\langle N(Y_1)N(Y_2)\cdots N(Y_s)\rangle\!\rangle$$

$$= \delta(Y_1-Y_2)\delta(Y_2-Y_3)\cdots\delta(Y_{s-1}-Y_s)\langle N(Y_1)\rangle$$

are proportional to the equilibrium density n of gas particles.

To order λ^2, the dynamics of the piston is described by the LE (17) with a time-dependent damping coefficient, $\gamma_0(t)=\beta/m\int_0^t d\tau\langle FF_0(\tau)\rangle$, where the evolution of the total
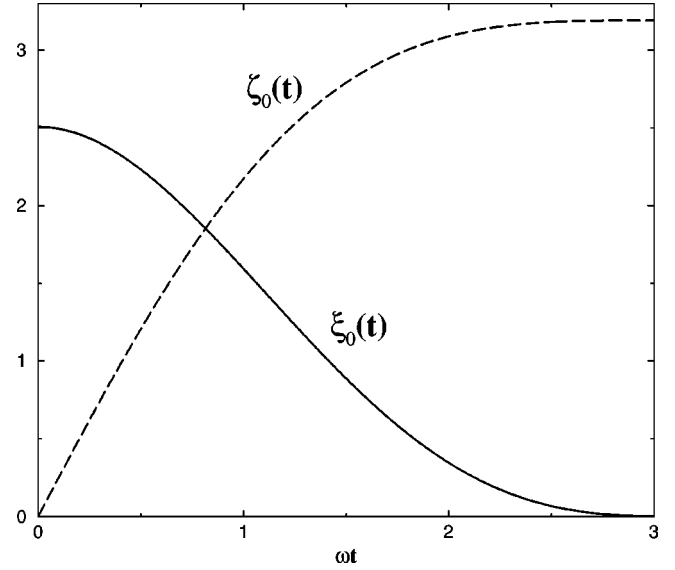

FIG. 1. The function $\xi_0(t)$ (solid line) governing the time dependence of the memory function $M_0(t)=N\omega^2\xi_0(t)$ in the non-Markovian Langevin equation (15), and the damping function $\zeta_0(t)$ (dashed line) in the local Langevin equation (48).

force $F_0=F_l+F_r$ is determined by the constrained piston-bath Liouville operator $\mathcal{L}_0$. Since $\langle F_l\rangle=-\langle F_r\rangle$ and $\langle F_lF_l(t)\rangle=\langle F_rF_r(t)\rangle$,

$$\langle FF_0(t)\rangle = 2\langle F_lF_l(t)\rangle - 2\langle F_l\rangle^2 = 2\langle\!\langle F_lF_l(t)\rangle\!\rangle. \qquad (41)$$

From Eqs. (41) and (33), one sees that $\langle FF_0(t)\rangle$ can be expressed in terms of the cumulant $\langle\!\langle N(x,v;t-\tau_c)N(x',v';-\tau_c)\rangle\!\rangle$. Using property (37) and Eq. (41), the cumulant may be rewritten as

$$\langle\!\langle N(x-vt,v;-\tau_c)N(x',v';-\tau_c)\rangle\!\rangle$$

$$= \delta(x-vt-x')\delta(v-v')nSf_M(v). \qquad (42)$$

Then, using Eqs. (41), (33), and (42), one obtains

$$\langle FF_0(t)\rangle = \frac{nSk_f^2}{\omega^3}\theta(\tau_c-t)\{\sin\omega t+\omega(\tau_c-t)\cos\omega t\}$$

$$\times\int_0^\infty dv\, f_M(v)v^3. \qquad (43)$$

This can be reexpressed in the compact form

$$\langle FF_0(t)\rangle = N\omega^2\frac{m}{\beta}\xi_0(t), \qquad (44)$$

where $\xi_0(t)$ is a dimensionless function (see Fig. 1) given by

$$\xi_0(t)=\sqrt{\frac{2}{\pi}}\theta(\tau_c-t)\{\sin\omega t+(\pi-\omega t)\cos\omega t\}, \qquad (45)$$

and

$$N = nS \frac{\langle v^2 \rangle^{1/2}}{\omega} \qquad (46)$$

is the average number of particles in the shell of thickness $l = \sqrt{\langle v^2 \rangle}/\omega$ around the piston. The parameter l specifies the length at which the average bath particle penetrates the interaction zone, and $N = nSl$ is the average number of particles in the layer of thickness l around the piston. Thus, it is evident that in addition to the mass ratio $\lambda^2 = m/M$, the relevant physics depends strongly on another characteristic parameter of the system, namely, $N\lambda^2$, which can be interpreted as the ratio of total mass $M_* = mnSl$ of bath particles in the layer of thickness l in the vicinity of the piston to the mass M of the piston.

With these definitions in hand, the time-dependent damping coefficient in the LE (17) can be written as

$$\gamma_0(t) = \frac{\beta}{m} \int_0^t d\tau \langle FF_0(t) \rangle = \omega N \zeta_0(t), \qquad (47)$$

and therefore the LE (17) assumes the form

$$\frac{dP_*(t)}{dt} = \lambda F^\dagger(t) - \omega \lambda^2 N \zeta_0(t) P_*(t), \qquad (48)$$

where the damping function $\zeta_0(t) = \omega \int_0^t d\tau \, \xi_0(\tau)$ is given by

$$\zeta_0(t) = \sqrt{\frac{2}{\pi}} \theta(\tau_c - t) \{ 2(1 - \cos \omega t) + (\pi - \omega t) \sin \omega t \}$$

$$+ 4\sqrt{\frac{2}{\pi}} \theta(t - \tau_c) \qquad (49)$$

(see Fig. 1). This expression describes the interesting time development of the dissipative force, an aspect of the dynamics that is outside the scope of more phenomenological models. For $t < \tau_c$, Eq. (48) describes essentially nonexponential relaxation of the momentum $\langle P(t) \rangle = P(0) e^{-\chi(t)}$ with

$$\chi(t) = \lambda^2 N\omega \int_0^t d\tau \zeta_0(\tau)$$

$$= \lambda^2 N \sqrt{\frac{2}{\pi}} \{ \pi + 2\omega t - (\pi - \omega t) \cos \omega t - 3 \sin \omega t \}. \qquad (50)$$

For small ωt, $\chi(t) \sim (\omega t)^2$. On a time scale $t > \tau_c$ the damping function reaches its plateau value and the Markovian limit of the Langevin equation, in which the damping coefficient is independent of time, is recovered:

$$\frac{dP_*(t)}{dt} = \lambda F^\dagger(t) - 4\sqrt{\frac{2}{\pi}} \omega \lambda^2 N P_*(t). \qquad (51)$$

Note that the characteristic time for relaxation of the momentum $\tau_p = \omega^{-1}(\lambda^2 N)^{-1}$ is governed by the parameter $\lambda^2 N$, rather than λ^2. It is also interesting to observe that the aver-

age number of collisions of bath particles with the piston for $t \sim \tau_p$ is of order $nS\langle v^2 \rangle^{1/2} \tau_p = \lambda^{-2}$ and depends neither on temperature nor on the piston's size, but only on the mass ratio.

Equation (51) is obtained under the condition that the characteristic time τ_c for the force autocorrelation function is negligible on a time scale τ_p of the dynamics of the momentum of the piston. Assuming in addition that the random force in this equation is Gaussian, one can obtain the Fokker-Planck equation for the momentum distribution function $f(P)$

$$\frac{\partial f(P)}{\partial t} = nS \frac{4m}{M} \sqrt{\frac{2k_B T}{\pi m}} \left\{ \frac{\partial}{\partial P} [Pf(P)] + M k_B T \frac{\partial^2 f(P)}{\partial P^2} \right\}. \qquad (52)$$

This coincides exactly with the equation for the piston interacting with the bath particles through a hard-wall potential previously obtained by van Kampen from the master equation [10].

The assumption of a Gaussian random force appears to be justified for Eq. (51) describing dynamics on time scales much longer than τ_c. In this case one can use a coarse-grained description of the dynamics with time resolution $\tau_c \ll \Delta t \ll \tau_p$. The coarse-graining procedure corresponds to replacing the instantaneous random force in Eq. (51) by its average over a time window of duration Δt, i.e., $F(t) \rightarrow \hat{F}(t) = \Delta t^{-1} \int_t^{t + \Delta t} F(t) \, dt$. As previously discussed, the number of collisions of bath particles with the piston for the time interval τ_p is of order $\lambda^{-2} \gg 1$. Therefore the resolution time interval Δt may be chosen sufficiently long that the piston experiences many uncorrelated collisions during Δt. Then, according to the central limit theorem, one may expect that $\hat{F}(t)$ is Gaussian distributed.

For the more general LE (17) with time-dependent damping, the random force is generally not Gaussian distributed. However, one can easily demonstrate that the distribution of the random force is approximately Gaussian in the limit $N \gg 1$ where the piston interacts simultaneously with many bath particles. In fact, the cumulant expansion (23) of the multiple-time correlation function $C_{2s} = \langle F(t_1)F(t_2) \cdots F(t_{2s}) \rangle$ contains the products of s pair correlation functions $\langle F(t_i)F(t_j) \rangle = \langle\langle F(t_i)F(t_j) \rangle\rangle$. Since a cumulant of any order is proportional to N, these terms are of order N^s. The other terms in the expansion contain fewer factors of the cumulants and therefore fewer factors of N, and hence may be neglected. Then C_{2s} can be approximately expressed as a linear combination of pair correlation functions, a well-known characteristic of a Gaussian random variable. The explicit form of the distribution function for the random force $f(F_0)$ can be obtained using the inverse Fourier transformation of the generating function (see, for example, Ref. [2]):

$$f(F_0) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dk \exp \left\{ -ikF_0 + \sum_{s=1}^{\infty} \frac{(ik)^s}{s!} \langle\langle F_0^s \rangle\rangle \right\}. \qquad (53)$$

The cumulants of odd orders vanish, and the cumulants of even orders are $\langle\!\langle F_0^{2s}\rangle\!\rangle = 2\langle\!\langle F_l^{2s}\rangle\!\rangle$ where F_l is given by Eq. (33). Using Eq. (41) one finds that

$$\langle\!\langle F_l^{2s}\rangle\!\rangle = \frac{1}{\sqrt{2}} N g^{2s} s! \frac{\Gamma(s+1/2)}{\Gamma(s+1)}, \qquad (54)$$

where $g^2 = k_f^2\langle v^2\rangle/2\omega^2$, and $\Gamma(s)$ is the gamma function. Substituting Eq. (54) in Eq. (53) gives the following integral representation for the distribution function:

$$f(F) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk \, \exp\{-ikF - \sqrt{2\pi}N(1-e^{-(gk)^2})\} \qquad (55)$$

If $N \gg 1$, one can approximately write in the above expression $1 - e^{-(gk)^2} \approx (gk)^2$, which leads immediately to the Gaussian distribution for the force. Similar arguments hold for the distribution function of higher order $f(F(t_1), F(t_2), \ldots F(t_s))$.

V. THE NONLINEAR LANGEVIN EQUATION

We now turn our attention to the terms of higher order in λ in the λ expansion (13) of the memory function $K(t) \equiv \langle FF^{\dagger}(t)\rangle = \Sigma_l K_l(t)$. It was shown in Sec. III that for a homogeneous bath $K_1(t) = 0$, and the first nonzero correction to $K_0(t) = \langle FF_0(t)\rangle$ is $K_2(t)$ which is of second order in λ. From Eqs. (13) and (23) one can see that K_2 has the structure

$$K_2(t) = \frac{\lambda^2}{m^2} A_1(t) P_*^2 + \frac{\lambda^2}{m} A_2(t), \qquad (56)$$

where the functions $A_1(t)$ and $A_2(t)$ are given by

$$A_1(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \langle\!\langle G_0 G_2(t, t_1, t_2)\rangle\!\rangle, \qquad (57)$$

$$A_2(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \langle\!\langle G_0 G_0(t - t_1) G_1(t, t_2)\rangle\!\rangle. \qquad (58)$$

Substitution of $K \approx K_0 + K_2$ into the exact equation of motion (11) leads to the nonlinear generalized LE of the form

$$\frac{dP_*(t)}{dt} = \lambda F^{\dagger}(t) - \lambda^2\int_0^t d\tau \, M_1(\tau) P_*(t-\tau)$$

$$- \lambda^4\int_0^t d\tau \, M_2(\tau) P_*^3(t-\tau), \qquad (59)$$

where the memory functions $M_1(t)$ and $M_2(t)$ are

$$M_1(t) = M_0(t) - \frac{2\lambda^2}{m^2} A_1(t) + \frac{\lambda^2\beta}{m^2} A_2(t), \qquad (60)$$

$$M_2(t) = \frac{\beta}{m^3} A_1(t). \qquad (61)$$

It is interesting to note that a nonlinear LE of this form has previously been obtained using a mode coupling approach [15]. A similar Markovian version of a nonlinear Langevin equation with a cubic damping term was considered by Mac-Donald [16] on purely phenomenological grounds.

Equation (59) differs from the linear LE (15) with memory function $M_0(t) = \beta/m\langle FF_0(t)\rangle$ not only by the presence of nonlinear damping, but also by appearance of correction terms of order λ^2 to the memory function $M_1(t)$ for the linear damping. Note also that the last term on the right-hand side of Eq. (59) can be written in the local form $-\lambda^4 P_*^3(t)\int_0^t d\tau \, M_2(\tau)$ since the nonlocal correction to this expression $\lambda^4\int_0^t d\tau \, M_2(\tau)\int_{t-\tau}^t d\tau' \dot{P}_*^3(\tau')$ is of order λ^5 [14]. However, the linear damping term in Eq. (59) cannot be simply written in the local form $-\lambda^2 P_*(t)\int_0^t d\tau \, M_1(\tau)$ since in this case the nonlocal correction has contributions of order λ^3 and λ^4 which must be retained. This correction can be written in the form

$$\lambda^2\int_0^t d\tau \, M_1(\tau)\int_{t-\tau}^t d\tau' \dot{P}_*(\tau')$$

$$= \lambda^3\int_0^t d\tau \, M_0(\tau)\int_{t-\tau}^t d\tau' F^{\dagger}(\tau')$$

$$- \lambda^4\int_0^t d\tau \, M_0(\tau)\int_{t-\tau}^t d\tau'\int_0^{\tau'} d\tau'' M_0(\tau'')$$

$$\times P_*(\tau' - \tau'') + O(\lambda^5), \qquad (62)$$

where we have used the result that $M_1(t) = M_0(t) + O(\lambda^2)$ according to Eq. (60). The first term in the right-hand side of this expression depends on initial coordinates of the bath and may be treated as a small correction to the random force $F^{\dagger}(t)$. The second term can be written in the local form

$$- \lambda^4 P_*(t)\int_0^t d\tau \, M_0(\tau)\int_{t-\tau}^t d\tau'\int_0^{\tau'} d\tau'' M_0(\tau'') + O(\lambda^5) \qquad (63)$$

to order λ^5. As a result, Eq. (59) can be written in the local form

$$\frac{dP_*(t)}{dt} = \lambda\widetilde{F}^{\dagger}(t) - \lambda^2\gamma_1(t) P_*(t) - \lambda^4\gamma_2(t) P_*^3(t), \qquad (64)$$

with the modified random force

$$\widetilde{F}^{\dagger}(t) = F^{\dagger}(t) + \lambda^3\int_0^t d\tau \, M_0(\tau)\int_{t-\tau}^t d\tau' F^{\dagger}(\tau') \qquad (65)$$

and the damping functions given by

$$\gamma_1(t) = \int_0^t d\tau \, M_1(\tau) + \lambda^2 \int_0^t d\tau \, M_0(\tau) \int_{t-\tau}^t d\tau'$$

$$\times \int_0^{\tau'} d\tau'' \, M_0(\tau''), \tag{66}$$

$$\gamma_2(t) = \int_0^t d\tau \, M_2(\tau). \tag{67}$$

For $t \gg \tau_c$, the time-dependent coefficients $\gamma_i(t)$ attain their limiting time-independent values γ_i, which can be obtained from the expressions above by setting the upper integration limit t to infinity.

It is possible to obtain explicit expressions for the memory functions $M_i(t)$ and the damping functions $\gamma_i(t)$ for the extended Rayleigh model. To accomplish this, explicit expressions for the functions G_i defined by Eqs. (19) must be computed. It is convenient to express these functions as the sum of two parts corresponding to the force acting on the left and right sides of the piston, $G_i = G_{li} + G_{ri}$. It is somewhat problematic to calculate terms such as $G_{l1} = e^{\mathcal{L}_0 \tau} \, \partial F_l(t)/\partial X$ due to the parametric dependence of $F_l(t)$ on X that is evident when the force is expressed in terms of $q_i(t)$ and $v_i(t)$ [see Eq. (33)]. One straightforward, albeit inelegant, way to circumvent this difficulty is to express the force in terms of $N(x,v) \equiv N(x,v;t=0)$. Details of this technique can be found in Appendix B. Using this approach, we obtain for the left-side part of G_1

$$G_{l1}(t_1,t_2) = k_f \int_0^\infty dv \int_{-v(\tau_c+t_1)}^b dq \, N(X_l+q,v;-\tau_c)$$

$$\times \cos \omega \left(t_1 + \frac{q}{v} \right) - k_f \theta(\tau_c - t_2) \cos \omega t_2$$

$$\times \int_0^\infty dv \int_{-v(\tau_c+t_1-t_2)}^{-vt_1} dq \, N(X_l+q,v;-\tau_c), \tag{68}$$

where the integration limit b is

$$b = -v(\tau_c + t_1 - t_2)\theta(\tau_c - t_2) - vt_1\theta(t_2 - \tau_c). \tag{69}$$

The expression for G_{r1} can be obtained from the above one by replacements,

$$\int_0^\infty dv \rightarrow \int_{-\infty}^0 dv, \quad \int_{q_1}^{q_2} dq \rightarrow \int_{q_2}^{q_1} dq. \tag{70}$$

Note that $\langle G_{l1} \rangle = \langle G_{r1} \rangle$.

For the left-side contribution to G_2, we obtain

$$G_{l2}(t,t_1,t_2) = k_f \int_0^\infty dv \, N(X_l - v(t+\tau_c),v;-\tau_c)$$

$$+ k_f \int_0^\infty dv \int_{-v(t+\tau_c)}^b dq \, N(X_l+q,v;-\tau_c)\frac{\omega}{v}$$

$$\times \sin \omega \left(t + \frac{q}{v} \right) + k_f \phi(t_1,t_2)$$

$$\times \int_0^\infty dv \, N(X_l - vt,v;-\tau_c), \tag{71}$$

where the function $\phi(t,t_1,t_2)$ is

$$\phi(t_1,t_2) = \theta(t_2 - \tau_c) - \theta(t_1 - \tau_c)\theta(\tau_c - t_2)\cos \omega t_2$$

$$+ \theta(\tau_c - t_1)\theta(\tau_c - t_2)\cos \omega t_2 \cos \omega t_1, \tag{72}$$

and the upper integration limit b is

$$b = -v(\tau_c + t - t_2)\theta(\tau_c - t_2) - vt\theta(t_2 - \tau_c). \tag{73}$$

A similar expression for G_{r2} can be obtained from that for G_{l2} using replacements (70), and, in addition, by multiplying the first and the last terms on the right-hand side of Eq. (71) by -1. Recall that we anticipate $\langle G_2 \rangle = 0$ by symmetry so that $\langle G_{r2} \rangle = -\langle G_{l2} \rangle$, which can be explicitly verified from the expressions above.

It has been shown above that the damping forces in the nonlinear Langevin equation can be expressed as integrals of the cumulants $\langle\!\langle G_0 G_2 \rangle\!\rangle$ and $\langle\!\langle G_0 G_0 G_1 \rangle\!\rangle$. From Eqs. (68), (71), (33), and (40), one can get explicit expressions for these cumulants. For $t > t_1 > t_2$ we find

$$\langle\!\langle G_0 G_2(t,t_1,t_2) \rangle\!\rangle = 2\langle\!\langle G_{l0} G_{l2}(t,t_1,t_2) \rangle\!\rangle$$

$$= \frac{nSk_f^2}{\omega} \phi_1(t,t_1,t_2) \int_0^\infty dv \, f_M(v)v, \tag{74}$$

where

$$\phi_1(t,t_1,t_2) = 2\theta(\tau_c - t)\sin \omega t \cos \omega t_1 \cos \omega t_2. \tag{75}$$

The second cumulant required is

$$\langle\!\langle G_0 G_0(t-t_1)G_1(t,t_2) \rangle\!\rangle$$

$$= 2\langle\!\langle G_{l0} G_{l0}(t-t_1)G_{l1}(t,t_2) \rangle\!\rangle$$

$$= -nS\left(\frac{k_f}{\omega} \right)^3 \phi_2(t,t_1,t_2) \int_0^\infty dv \, f_M(v)v^3,$$

where the function ϕ_2 has the form

$$\phi_2(t,t_1,t_2) = \theta(\tau_c - t)\cos \omega t_2 \{ \omega(\tau_c - t)\cos \omega(t-t_1)$$

$$+ \tfrac{1}{2}\sin \omega(t+t_1) + \tfrac{1}{2}\sin \omega(t-t_1) \}.$$

These results allow one to calculate the functions $A_i(t)$ defined by Eqs. (57) and (58),

$$A_1(t) = m^2\omega^2 N \xi_1(t), \qquad (76)$$

$$A_2(t) = -m^2\omega^2\beta^{-1}N \xi_2(t), \qquad (77)$$

where dimensionless functions $\xi_i(t)$ are given by

$$\xi_1(t) = \frac{1}{\sqrt{2\pi}}\theta(\tau_c - t)\sin^3 \omega t,$$

$$\xi_2(t) = \frac{1}{\sqrt{2\pi}}\theta(\tau_c - t)\{\sin^3 \omega t + \omega t(\pi - \omega t)\sin \omega t\}. \qquad (78)$$

The memory functions in the non-Markovian LE (59) take the form

$$M_1(t) = N\omega^2\{\xi_0(t) - 2\lambda^2\xi_1(t) - \lambda^2\xi_2(t)\}, \qquad (79)$$

$$M_2(t) = N\omega^2\frac{\beta}{m}\xi_1(t), \qquad (80)$$

Here the function $\xi_0(t)$ is given by Eq. (45) and governs the memory function $M_0(t)$ in the linear LE (15), $M_0(t) = N\omega^2\xi_0(t)$. The additional λ^2-correction terms lead to a faster decay of the linear damping kernel $M_1(t)$ compared to $M_0(t)$. The kernel for nonlinear damping $M_2(t)$ is not a decaying function of time but rather has a maximum at $t = \tau_c/2$.

Explicit expressions for damping functions $\gamma_i(t)$ in the local-in-time LE (64) by integrating $M_i(t)$ according to Eqs. (66) and (67), and the nonlinear LE for the Rayleigh model with a parabolic potential takes the form

$$\frac{dP_*(t)}{dt} = \lambda\widetilde{F}^\dagger(t) - N\lambda^2\omega\left\{\zeta_1(t)P_*(t) + \lambda^2\frac{\beta}{m}\zeta_2(t)P_*^3(t)\right\}, \qquad (81)$$

where the nonlinear damping function $\zeta_2(t)$ is given by

$$\zeta_2(t) = \omega\int_0^t d\tau\,\xi_1(\tau)$$

$$= \frac{1}{\sqrt{2\pi}}\theta(\tau_c - t)\left\{\frac{2}{3} - \cos\omega t + \frac{1}{3}\cos^3\omega t\right\}$$

$$+ \frac{1}{3}\sqrt{\frac{8}{\pi}}\theta(t - \tau_c), \qquad (82)$$

and the linear damping function $\gamma_1(t)$ can be written as

$$\zeta_1(t) = \zeta_0(t) + \lambda^2\varepsilon_1(t) + N\lambda^2\varepsilon_2(t). \qquad (83)$$

The main contribution to the overall damping coefficient $\zeta_1(t)$ is given by the function $\zeta_0(t) = \omega\int_0^t d\tau\,\xi_0(\tau)$, while the corrections arising at higher orders in the λ expansion are

$$\varepsilon_1(t) = -\omega\int_0^t d\tau\{2\xi_1(\tau) + \xi_2(\tau)\}, \qquad (84)$$

$$\varepsilon_2(t) = \omega^3\int_0^t d\tau\,\xi_0(\tau)\int_{t-\tau}^t d\tau'\int_0^{\tau'} d\tau''\,\xi_0(\tau''). \qquad (85)$$

Note that $\varepsilon_1(t)$ and $\varepsilon_2(t)$ are of different signs for all t. If $N \ll 1$ the correction is determined mostly by $\varepsilon_1(t) < 0$ and tends to decrease the damping function $\zeta_1(t)$. In contrast, if $N \gg 1$ the main correction comes from $\varepsilon_2(t) > 0$ effectively increasing the linear damping.

For a coarse-grained description on the time scale $t \gg \tau_c$ with a time resolution $\tau_c \ll \Delta t \ll \tau_p$, one can replace the damping functions $\gamma_i(t)$ in the LE (81) by their limiting values $\gamma_i(\tau_c)$.

VI. CONCLUDING REMARKS

The notion that the character of Brownian motion of a finite-sized particle may depend on parameters other than the mass ratio λ^2 dates back to Lorentz and has been examined by many authors (see Refs. [17–19], and references therein). It is known that when hydrodynamic effects are important another relevant parameter is the ratio of the mass density of the bath to that of the particle. In this paper we have demonstrated that even when hydrodynamic effects are absent, as in the extended Rayleigh model, the character of the behavior of a tagged particle may not be governed by λ^2 but by the renormalized parameter $\lambda_*^2 = N\lambda^2$, which can be interpreted as the ratio of the average total mass of particles in the interaction zone M_* to the mass M of the tagged particle. When the average number of particles in the interaction zone is large (i.e., $N \gg 1$), $\lambda_* \ll 1$ is a necessary condition for the applicability of a conventional perturbation scheme of derivation of the LE. In this case the conventional assumption of Gaussian random force is justified for any time scale. If $N < 1$, the parameter of the expansion is λ^2, and the Gaussian force approximation holds only on a time scale that is much longer than the characteristic time for the relaxation of the bath.

Although this paper focuses on the specific model of an ideal gas bath interacting with a Brownian particle through a quadratic repulsive potential, many of the results obtained are quite general. In particular, the LE (64) and expressions (66) and (67) for the damping coefficients in terms of microscopic time-correlation functions, which may be considered as the generalized version of the fluctuation-dissipation theorem, are limited neither to the specific form of interaction potential between the bath and the tagged particle nor to the ideal gas bath. The results of Sec. III concerning the cumulant expansion of the kernel $K(t) = \langle FF^\dagger(t)\rangle$ are also general and not limited to any specific model. Combined with quite general theorems about cumulant properties [20], these results may be useful for more realistic models with interacting bath particles.

The explicit expressions for the kinetic coefficients and memory functions appearing in the Langevin equations have been derived in this paper in the thermodynamic limit, so that any correlations due to finite size of the system are neglected. It should be mentioned, however, that the equations themselves, as well as the fluctuation-dissipation relations relating the kinetic coefficients to correlation functions, also

hold for a system with finite baths. For finite systems the explicit form of the kinetic coefficients may be rather complicated even for a bath composed of ideal gas particles.

In this paper we have considered the case of the totally symmetric bath when thermodynamic and microscopic properties of the gas to the left and to the right of the piston are the same. Some interesting physical implications arising in the case of an asymmetric bath will be presented elsewhere [21].

APPENDIX A

In this appendix, for the sake of completeness, we show explicitly that the function C_4 defined by Eqs. (14) is of second order in cumulants of products of G_i. C_4 can be written as the sum

$$C_4 = C_4^{(1)} + C_4^{(2)} + C_4^{(3)} + C_4^{(4)},$$

where the first constituent

$$C_4^{(1)} = \frac{2}{m^2} \{ \langle G_0 G_0(t-t_1) G_0(t-t_2) G_2(t,t_3,t_4) \rangle - \langle G_0 G_0(t-t_1) \rangle \langle G_0(t-t_2) G_2(t,t_3,t_4) \rangle \},$$

is obviously quadratic in cumulants,

$$C_4^{(1)} = \frac{2}{m^2} \{ \langle\langle G_0 G_0(t-t_2) \rangle\rangle \langle\langle G_0(t-t_1) G_2(t,t_3,t_4) \rangle\rangle + \langle\langle G_0(t-t_1) G_0(t-t_2) \rangle\rangle \langle\langle G_0 G_2(t,t_3,t_4) \rangle\rangle + \langle\langle G_0 G_0(t-t_1) G_0(t-t_2) G_2(t,t_3,t_4) \rangle\rangle \}.$$

The second term is

$$C_4^{(2)} = \frac{1}{m^2} \{ \langle G_0 G_0(t-t_1) A_1 \rangle - \langle G_0 G_0(t-t_1) \rangle \langle A_1 \rangle - \langle G_1(t,t_4) \rangle [\langle G_0 G_0(t-t_1) G_1(t-t_3,t_2-t_3) \rangle - \langle G_0 G_0(t-t_1) \rangle \langle G_1(t-t_3,t_2-t_3) \rangle] \},$$

where $A_1 = S(t-t_2) G_0(t_2-t_3) G_1(t_2,t_4)$. Noting that A_1 can be written as

$$A_1 = G_1(t-t_3,t_2-t_3) G_1(t,t_4) + G_0(t-t_3) G_2(t,t_2,t_4),$$

and recalling that due to symmetry $\langle G_2(t) \rangle$ and $\langle G_0 G_1(t) \rangle$ are zero at all times [see Eq. (21)], one can see that $C_4^{(2)}$ is also quadratic in cumulants,

$$C_4^{(2)} = \frac{1}{m^2} \{ \langle G_1(t-t_3,t_2-t_3) \rangle \langle\langle G_0 G_0(t-t_1) G_1(t,t_4) \rangle\rangle + \langle\langle G_0 G_0(t-t_3) \rangle\rangle \langle\langle G_0(t-t_1) G_2(t,t_2,t_4) \rangle\rangle + \langle\langle G_0 G_2(t,t_2,t_4) \rangle\rangle \langle\langle G_0(t-t_1) G_0(t-t_3) \rangle\rangle + \langle\langle G_0 G_0(t-t_1) G_1(t-t_3,t_2-t_3) G_1(t,t_4) \rangle\rangle + \langle\langle G_0 G_0(t-t_1) G_0(t-t_3) G_2(t,t_2,t_4) G_1(t,t_4) \rangle\rangle \}.$$

The third term is

$$C_4^{(3)} = \frac{P_*^2}{m^3} \{ \langle G_0 B_1 \rangle - \langle G_0 B_2 \rangle \langle G_1(t_2,t_4) \rangle + 2 \langle G_0 B_3 \rangle \},$$

where

$$B_1 = S(t-t_1) S(t-t_2) G_0(t_2-t_3) G_1(t_2,t_4),$$

$$B_2 = S(t-t_1) S(t-t_2) G_0(t_2-t_3),$$

$$B_3 = S(t-t_1) G_0(t_1-t_2) G_2(t_1,t_3,t_4)$$

can be expressed as

$$B_1 = G_2(t-t_3,t_1-t_3,t_2-t_3) G_1(t,t_4) + G_1(t-t_3,t_2-t_3) G_2(t,t_1,t_4) + G_1(t-t_3,t_1-t_3) G_2(t,t_2,t_4) + G_0(t-t_3) G_3(t,t_1,t_2,t_4),$$

$$B_2 = G_2(t-t_3,t_1-t_3,t_2-t_3),$$

$$B_3 = G_1(t-t_2,t_1-t_2) G_2(t,t_3,t_4) + G_0(t-t_2) G_3(t,t_1,t_3,t_4).$$

Then using the symmetry property (21) one can see that $C_4^{(3)}$ is quadratic in cumulants.

The remaining term

$$C_4^{(4)} = \left(\frac{P_*}{m} \right)^2 \langle G_0 G_4(t,t_1,t_2,t_3,t_4) \rangle + \frac{3P_*^2}{m^3} \{ \langle G_0 G_0(t-t_1) G_3(t,t_2,t_3,t_4) \rangle - \langle G_0 G_0(t-t_1) \rangle \langle G_3(t,t_2,t_3,t_4) \rangle \}$$

is clearly linear in cumulants,

$$C_4^{(4)} = \left(\frac{P_*}{m} \right)^2 \langle\langle G_0 G_4(t,t_1,t_2,t_3,t_4) \rangle\rangle + \frac{3P_*^2}{m^3} \langle\langle G_0 G_0(t-t_1) G_3(t,t_2,t_3,t_4) \rangle\rangle.$$

APPENDIX B

In this appendix the functions G_i defined by Eqs. (19) are evaluated for the extended Rayleigh model of diffusion. These functions are defined in terms of powers of alternating operators $\partial/\partial X$ and $e^{\mathcal{L}_0(t_i - t_k)}$. As mentioned in the text, representations (33) and (35) for the force on the fixed piston are unwieldy since these involve $q_i(t)$ and $v_i(t)$ which are functions of X. For the purpose of evaluating the G_i, it is convenient to express the force in terms of $N(x,v) \equiv N(x,v;t=0)$. For $t > 0$, we have

$$
F_l(t) = k_f \int_0^\infty dv \int_{-vt}^{a(t)} dq \; N(X_l + q, v) \frac{v}{\omega} \sin \omega \left(t + \frac{q}{v} \right)
$$
$$
+ \theta(\tau_c/2 - t) k_f \int_0^\infty dv \int_0^\infty dq \; N(X_l + q, v) q(t)
$$
$$
+ \theta(\tau_c/2 - t) k_f \int_{-\infty}^0 dv \int_{Q(t)}^\infty dq \; N(X_l + q, v), q(t)
$$
$$
+ \theta(t - \tau_c/2) \theta(\tau_c - t) k_f
$$
$$
\times \int_0^\infty dv \int_0^{Q(t)} dq \; N(X_l + q, v) q(t), \tag{B1}
$$

where

$$
a(t) = -v \, \theta(t - \tau_c)(t - \tau_c),
$$

$$
q(t) = q \cos \omega t + \frac{v}{\omega} \sin \omega t,
$$

$$
Q(t) = -\frac{v}{\omega} \tan \omega t.
$$

The first term in Eq. (B1) describes the contribution to the force $F_l(t)$ from the particles that were outside the interaction zone at $t=0$ (i.e., $q<0$) and are in the interaction zone at the moment t [i.e., $q(t)>0$].

The remaining terms give the contribution from particles that were in the interaction zone at $t=0$ (i.e., $q>0$) and are still there at the moment t [i.e., $q(t)>0$]. In fact, all particles in the interaction zone with positive initial velocities at $t=0$ will be still in the interaction zone at $t<\tau_c/2$ (the second term), while the particles with negative initial velocities will be in the interaction zone at time $t<\tau_c/2$ only if at $t=0$ they reside deep inside the interaction zone, namely, $q>Q(t)$ (the third term). For $\tau_c>t>\tau_c/2$ only the particles with positive initial velocity will be in the interaction zone at time t provided their initial coordinates are less than $Q(t)$ (the last term).

From expression (B1), one easily calculates $\partial F_l(t)/\partial X$ writing $\partial N(X_l + q, v)/\partial X = \partial N(X_l + q)/\partial q$ and integrating by parts to obtain

$$
\frac{\partial F_l(t)}{\partial X} = -k_f \cos \omega t N_z(t) - k_f \int_0^\infty dv
$$
$$
\times \int_{-vt}^{a(t)} dq \; N(X_l + q, v) \frac{v}{\omega} \cos \omega \left(t + \frac{q}{v} \right). \tag{B2}
$$

Here $N_z(t)$ is the number of particles which were in the interaction zone at $t=0$ and remain at $t>0$,

$$
N_z(t) = \theta(\tau_c/2 - t) \int_0^\infty dv \int_0^\infty dq \; N(X_l + q, v)
$$
$$
+ \theta(\tau_c/2 - t) \int_{-\infty}^0 dv \int_{Q(t)}^\infty dq \; N(X_l + q, v)
$$
$$
+ \theta(t - \tau_c/2) \theta(\tau_c - t) \int_0^\infty dv \int_0^{Q(t)} dq \; N(X_l + q, v). \tag{B3}
$$

$N_z(t)$ can be written in compact form in terms of the density at time $-\tau_c$ according to

$$
N_z(t) = \theta(\tau_c - t) \int_0^\infty dv \int_{-v\tau_c}^{-vt} dq \; N(X_l + q, v; -\tau_c). \tag{B4}
$$

In fact, the number of particles in the interaction zone at $t=0$ is

$$
\int_0^\infty dv \int_{-v\tau_c}^0 dq \; N(X_l + q, v; -\tau_c), \tag{B5}
$$

while at time t it is given by

$$
\int_0^\infty dv \int_{-v\tau_c}^0 dq \; N(X_l + q, v; t - \tau_c)
$$
$$
= \int_0^\infty dv \int_{-v(t+\tau_c)}^{-vt} dq \; N(X_l + q, v; -\tau_c). \tag{B6}
$$

By definition $N_z(t)$ involves the particles that contribute to both integrals (B5) and (B6), which leads to Eq. (B4). Combining Eqs. (B2) and (B4), we have

$$\frac{\partial F_l(t)}{\partial X} = -k_f \int_0^\infty dv \int_{-vt}^{a(t)} dq \, N(X_l + q, v) \cos \omega \left(t + \frac{q}{v} \right)$$

$$- k_f \theta(\tau_c - t) \cos \omega t \int_0^\infty dv$$

$$\times \int_{-v\tau_c}^{-vt} dq \, N(X_l + q, v; -\tau_c). \qquad (\text{B7})$$

For comparison, let us calculate the average derivative of the total force $F_0(t) = F_l + F_r$,

$$\left\langle \frac{\partial F_0(t)}{\partial X} \right\rangle = 2 \left\langle \frac{\partial F_l(t)}{\partial X} \right\rangle = -\frac{2k_f nS}{\omega} \int_0^\infty f_M(v) v \, dv \, \theta(\tau_c - t)$$

$$\times [\sin \omega t + (\pi - \omega t) \cos \omega t].$$

Comparing this expression with Eq. (43) for the memory kernel $K_0(t) = \langle FF_0(t) \rangle$, it is evident that $\langle \partial F_0(t)/\partial X \rangle = -\beta \langle FF_0(t) \rangle$. This is the general result used in the main text, Eq. (10), confirmed using the explicit expression for $\partial F_0(t)/\partial X$.

From Eq. (B7), we obtain the following expression for G_{l1}:

$$G_{l1}(t_1, t_2) \equiv e^{\mathcal{L}_0(t_1 - t_2)} \frac{\partial F_l(t_2)}{\partial X}$$

$$= -k_f \int_0^\infty dv \int_{-vt_2}^{a(t_2)} dq \, N(X_l + q, v; t_1 - t_2)$$

$$\times \cos \omega \left(t_2 + \frac{q}{v} \right) - k_f \theta(\tau_c - t_2) \cos \omega t_2$$

$$\times \int_0^\infty dv \int_{-v\tau_c}^{-vt_2} dq \, N(X_l + q, v; t_1 - t_2 - \tau_c),$$

$$(\text{B8})$$

where, according to Eq. (13), it is assumed that $t_1 > t_2$. Expressing this in terms of the microscopic density at time $-\tau_c$ using Eq. (37), one obtains Eq. (68) of the main text.

To evaluate G_2, one has to take the derivative of G_1 with respect to X. Let us express G_1 in terms of the density at time $t = 0$, $N(x, v)$, as done above for $F_l(t)$. The first term in the right-hand side of Eq. (B8) involves only particles located outside the interaction zone, so using property (37), the first term can be written as

$$-k_f \int_0^\infty dv \int_{-vt_2}^{a(t_2)} dq \, N(X_l + q - v(t_1 - t_2), v) \cos \omega \left(t_2 + \frac{q}{v} \right)$$

$$= -k_f \int_0^\infty dv \int_{-vt_1}^{b} dq \, N(X_l + q, v) \cos \omega \left(t_1 + \frac{q}{v} \right), \qquad (\text{B9})$$

where the upper integration limit is

$$b = -v(t_1 - t_2) \theta(\tau_c - t_2) - v(t_1 - \tau_c) \theta(t_2 - \tau_c).$$

The second term on the right-hand side of Eq. (B8) can be expressed as

$$-k_f \theta(\tau_c - t_2) \cos \omega t_2 \int_0^\infty dv$$

$$\times \int_{-v\tau_c}^{-vt_2} dq \, N(X_l + q - v(t_1 - t_2), v; -\tau_c)$$

$$= -k_f \theta(\tau_c - t_2) \cos \omega t_2 \int_0^\infty dv$$

$$\times \int_{-v(\tau_c + t_1 - t_2)}^{-vt_1} dq \, N(X_l + q, v; -\tau_c). \qquad (\text{B10})$$

If $t_1 > \tau_c$, then all particles that contribute to this integral at $t = 0$ will be outside the interaction zone in the q interval from $-v(t_1 - t_2)$ to $-v(t_1 - \tau_c)$. Therefore, for $t_1 > \tau_c$ the second term equals

$$-k_f \theta(\tau_c - t_2) \theta(t_1 - \tau_c) \cos \omega t_2 \int_0^\infty dv$$

$$\times \int_{-v(t_1 - t_2)}^{-v(t_1 - \tau_c)} dq \, N(X_l + q, v). \qquad (\text{B11})$$

If $t_1 < \tau_c$, then two sets of particles contribute to expression (B10). The first set of particles is composed of particles that at time $-\tau_c$ are in the q interval from $-v(\tau_c + t_1 - t_2)$ to $-v\tau_c$. At $t = 0$, these particles will be outside the interaction zone in the interval $(-v(t_1 - t_2), 0)$, and hence their contribution is

$$-k_f \cos \omega t_2 \int_0^\infty dv \int_{-v(t_1 - t_2)}^{0} dq \, N(X_l + q, v). \quad (\text{B12})$$

The second group of particles are those that were in the q interval from $-v\tau_c$ to $-vt_1$ at time $-\tau_c$. At $t = 0$, all these particles will be in the interaction zone. Taking into account Eq. (B4), the corresponding contribution can be written as $-k_f \cos \omega t_2 N_z(t_1)$. Using expression (B3) for $N_z(t)$, one arrives at the following representation for G_{l1} in terms of $N(x, v)$,

$$
G_{l1}(t_1,t_2) = -k_f \int_0^\infty dv \int_{-vt_1}^b dq\, N(X_l+q,v) \cos \omega \left(t_1 + \frac{q}{v} \right) - k_f \theta(\tau_c-t_2)\,\theta(t_1-\tau_c)\cos \omega t_2
$$

$$
\times \int_0^\infty dv \int_{-v(t_1-t_2)}^{-v(t_1-\tau_c)} dq\, N(X_l+q,v) - k_f \theta(\tau_c-t_2)\,\theta(\tau_c-t_1)\cos \omega t_2
$$

$$
\times \int_0^\infty dv \int_{-v(t_1-t_2)}^0 dq\, N(X_l+q,v) - k_f \theta(\tau_c-t_2)\,\theta(\tau_c/2-t_1)\cos \omega t_2
$$

$$
\times \int_0^\infty dv \int_0^\infty dq\, N(X_l+q,v) - k_f \theta(\tau_c-t_2)\,\theta(\tau_c/2-t_1)\cos \omega t_2 \int_{-\infty}^0 dv \int_{Q(t_1)}^\infty dq\, N(X_l+q,v)
$$

$$
- k_f \theta(\tau_c-t_2)\,\theta(\tau_c-t_1)\,\theta(t_1-\tau_c/2)\cos \omega t_2 \int_0^\infty dv \int_0^{Q(t_1)} dq\, N(X_l+q,v). \tag{B13}
$$

Taking derivative of this expression with respect to X gives

$$
\frac{\partial}{\partial X} G_{l1}(t_1,t_2) = k_f \int_0^\infty dv\, N(X_l-vt_1,v) + k_f [\,\theta(t_2-\tau_c) - \theta(\tau_c-t_2)\,\theta(t_1-\tau_c)\cos \omega t_2\,] \int_0^\infty dv\, N(X_l-v(t_1-\tau_c),v)
$$

$$
- k_f \int_0^\infty dv \int_{-vt_1}^b dq\, N(X_l+q,v)\frac{\omega}{v}\sin \omega \left(t_1 + \frac{q}{v} \right) + \theta(\tau_c/2-t_1)\,\theta(\tau_c-t_2)k \cos \omega t_2
$$

$$
\times \int_{-\infty}^0 dv\, N(X_l+Q(t_1),v) - \theta(\tau_c-t_1)\,\theta(t_1-\tau_c/2)\,\theta(\tau_c-t_2)k \cos \omega t_2 \int_0^\infty dv\, N(X_l+Q(t_1),v). \tag{B14}
$$

The last two terms can be written as

$$
k_f \theta(\tau_c-t_2)\,\theta(\tau_c-t_1)\frac{\cos \omega t_2}{\tan \omega t_1} \int_0^\infty dv\, N\left(X_l + \frac{v}{\omega}, -\frac{v}{\tan \omega t_1} \right)
$$

$$
= k_f \theta(\tau_c-t_2)\,\theta(\tau_c-t_1)\frac{\cos \omega t_2}{\tan \omega t_1} \int_0^\infty dv\, N\left(X_l - \frac{vt_1}{\sin \omega t_1}, \frac{v}{\sin \omega t_1}; -\tau_c \right)
$$

$$
= k_f \theta(\tau_c-t_2)\,\theta(\tau_c-t_1)\cos \omega t_1 \, \cos \omega t_2 \int_0^\infty dv\, N(X_l-vt_1,v;-\tau_c), \tag{B15}
$$

where we have used the property that if the initial coordinate and velocity of the particle in the interaction zone are

$$
q(0) = \frac{V}{\omega}, \quad v(0) = -\frac{V}{\tan \omega t} \tag{B16}
$$

with $V>0$ and $0<t<\tau_c$, then at $t=-\tau_c$

$$
q(-\tau_c) = -v(-\tau_c)t, \quad v(-\tau_c) = \frac{V}{\sin \omega t}. \tag{B17}
$$

Substituting Eq. (B15) into Eq. (B14), acting on the result by the propagator $e^{\mathcal{L}_0(t-t_1)}$, and using again the property $N(x,v;t+\tau) = N(x-v\tau,v;t)$ for the motion outside the interaction zone, we finally obtain for $t>t_1>t_2$,

$$G_{l2}(t,t_1,t_2) \equiv e^{\mathcal{L}_0(t-t_1)} \frac{\partial}{\partial X} G_1(t_1,t_2)$$

$$= k_f \int_0^\infty dv\, N(X_l - vt, v) + k_f \theta(t_1 - \tau_c)\theta(t_2 - \tau_c) \int_0^\infty dv\, N(X_l - v(t - \tau_c), v) - k_f \theta(\tau_c - t_2)\theta(t_1 - \tau_c)\cos \omega t_2$$

$$\times \int_0^\infty dv\, N(X_l - v(t - \tau_c), v) - k_f \int_0^\infty dv \int_{-vt}^b dq\, N(X_l + q, v) \frac{\omega}{v}\sin \omega\left(t + \frac{q}{v}\right)$$

$$+ k_f \theta(\tau_c - t_2)\theta(\tau_c - t_1)\cos \omega t_1 \, \cos \omega t_2 \int_0^\infty dv\, N(X_l - vt, v; -\tau_c), \tag{B18}$$

where $b = -v(t - t_2)\theta(\tau_c - t_2) - v(t - \tau_c)\theta(t_2 - \tau_c)$. Expressing the first three terms through $N(x, v; -\tau_c)$ rather than $N(x, v)$, one arrives at Eq. (71) of the main text.

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