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Exact time evolution and master equations for the damped harmonic oscillator

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Using the exact path integral solution for the damped harmonic oscillator it is shown that in general there does not exist an exact dissipative Liouville operator describing the dynamics of the oscillator for arbitrary initial bath preparations. Exact nonstationary Liouville operators can be found only for particular preparations. Three physically meaningful examples are examined. An exact master equation is derived for thermal initial conditions. Second, the Liouville operator governing the time evolution of equilibrium correlations is obtained. Third, factorizing initial conditions are studied. Additionally, one can show that there are approximate Liouville operators independent of the initial preparation describing the long-time dynamics under appropriate conditions. The general form of these approximate master equations is derived and the coefficients are determined for special cases of the bath spectral density including the Ohmic, Drude, and weak coupling cases. The connection with earlier work is discussed. [S1063-651X(97)04001-4]

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I. INTRODUCTION

Recently the problem of the reduced dynamics of a quantum system in contact with a reservoir has gained renewed interest [1–3]. While the foundations of quantum dissipative processes were layed already in the 1960s [4], this early work was mainly concerned with weakly damped systems and has relied on the Born and Markov approximations. In this conventional approach the dynamics of the dissipative quantum system is described in terms of quantum master or Langevin equations. Important progress beyond the limitations of the weak coupling approach was made in the 1980s, in particular path integral techniques were shown to be powerful means to describe quantum dissipative systems [5]. This has led to unexpected results, such as, for instance, the nonexponential decay of correlation functions in the low temperature range [6,7], not directly available within the conventional master equation approach.

The insight gained also provided a basis for profound critique [1,3,8,9] of concepts developed in the context of the quantum master equation approach such as complete positivity or the quantum regression theorem. However, the results of the path integral approach were rarely used to derive improved master equations valid in the range of strong damping and/or low temperatures. In this paper we shall address this problem for the ubiquitous quantum dissipative system, the damped harmonic oscillator. Based on available exact results [10] we derive generalized quantum master equations describing the exact relaxation of mean values and the time evolution of equilibrium correlation functions. Whenever appropriate, the results will be confronted and compared with earlier findings.

More specifically, this paper is organized as follows. In the following section the microscopic model of a harmonic oscillator coupled to a harmonic bath is introduced. The exact time evolution of the reduced density matrix of the oscillator [10] will briefly be summarized. In Sec. III we examine whether the exact time evolution of the density matrix is compatible with a generalized quantum master equation with a time-dependent Liouville operator. It is shown that in general there is no Liouville operator independent of the initial preparation. In Sec. IV we study specific types of initial preparations. Exact time-dependent Liouville operators describing the time evolution of thermal initial conditions and of correlation functions are derived. In the classical limit these Liouville operators are shown to reduce to the Adelman Fokker-Planck operator [11]. We also examine the case of factorizing initial conditions.

In Sec. V we investigate particular types of damping leading to a time evolution of the density matrix governed approximately by a time-independent Liouville operator. Earlier results by Talkner [9] and by Haake and Reibold [12] are recovered. We discuss in detail the limit of a weakly damped oscillator and derive a generalization of the Agarwal equation [13]. Additional time coarse graining is shown to lead to the quantum-optical master equation by Weidlich and Haake [14]. Finally in Sec. VI we present our conclusions.

II. MODEL HAMILTONIAN AND EXACT TIME EVOLUTION

The standard microscopic model [15–17] for the damped harmonic oscillator starts out from a Hamiltonian H composed of the oscillator part

$$H_O=\frac{p^2}{2M}+\frac{M}{2}\omega_0^2q^2, \tag{1}$$

the bath Hamiltonian

$$H_R=\sum_{n=1}^{N}\left(\frac{p_n^2}{2m_n}+\frac{m_n}{2}\omega_n^2x_n^2\right), \tag{2}$$

and an interaction part

$$H_{OR}=-q\sum_{n=1}^{N}c_nx_n+q^2\sum_{n=1}^{N}\frac{c_n^2}{2m_n\omega_n^2}. \tag{3}$$

This model describes an oscillator with the associated classical equation of motion

$$M\ddot{q}(t)+\int_0^t ds\,\gamma(t-s)\dot{q}(s)+\omega_0^2 q(t)=0, \tag{4}$$

where

$$\gamma(t)=\frac{1}{M}\sum_{n=1}^{N}\frac{c_n^2}{m_n\omega_n^2}\cos(\omega_n t) \tag{5}$$

is the damping kernel. The dynamics of the density matrix of the entire system (oscillator and bath) is given by

$$W(t)=\exp(-iHt/\hbar)W(0)\exp(iHt/\hbar), \tag{6}$$

from where the reduced density matrix of the oscillator is obtained by tracing out the bath modes

$$\rho(t)=\mathrm{Tr}_R W(t). \tag{7}$$

The path integral technique allows for a complete solution to this problem for a large class of initial states. Since the derivation is expounded in [10], we merely state the result

$$\rho(q_f,q_f',t)=\int dq_i dq_i' d\overline{q}d\overline{q'}J(q_f,q_f',t,q_i,q_i',\overline{q,q'})\times\lambda(q_i,q_i',\overline{q,q'}), \tag{8}$$

where $\lambda(q_i,q_i',\overline{q,q'})$ is the so-called preparation function defining the initial nonequilibrium state $W(0)$ as a modification of the equilibrium state $W_\beta=\exp(-\beta H)/\mathrm{Tr}\exp(-\beta H)$ in the subspace of the oscillator. This modification can be produced by operators O_j, O_j' that act upon the particle only leaving the reservoir coordinates unchanged:

$$W(0)=\sum_j O_j W_\beta O_j'. \tag{9}$$

We then have

$$\langle q|W(0)|q'\rangle=\int d\overline{q}d\overline{q'}\lambda(q,q',\overline{q,q'})\langle\overline{q}|W_\beta|\overline{q'}\rangle, \tag{10}$$

where

$$\lambda(q,q',\overline{q,q'})=\sum_j\langle q|O_j|\overline{q}\rangle\langle\overline{q'}|O_j'|q'\rangle. \tag{11}$$

Several relevant examples for $\lambda(q,q',\overline{q,q'})$ are discussed in [10] and special cases will be considered below. $J(q_f,q_f',t,q_i,q_i',\overline{q,q'})$ is the propagating function describing the time evolution of the reduced density matrix. In terms of the transformed coordinates $r=(q+q')/2$, $x=q-q'$, it reads

$$J(x_f,r_f,t,x_i,r_i,\overline{x,r})=\frac{1}{N(t)}\exp\left[\frac{i}{\hbar}\Sigma(x_f,r_f,t,x_i,r_i,\overline{x,r})\right], \tag{12}$$

where

$$\begin{aligned}\Sigma(x_f,r_f,t,x_i,r_i,\overline{x,r})={}&i\left(\frac{\hbar\overline{r^2}}{2\langle q^2\rangle}+\frac{\langle p^2\rangle}{2\hbar}\overline{x^2}\right)+M[x_f r_f f_1(t)\\&+x_i r_i f_2(t)-x_i r_f f_3(t)-x_f r_i f_4(t)]\\&+\frac{i}{2}M[x_i^2R^{++}(t)+2x_f x_i R^{+-}(t)\\&+x_f^2R^{--}(t)]+M\{\overline{r[}x_iC_1^+(t)\\&+x_fC_1^-(t)]-i\overline{x[}x_iC_2^+(t)\\&+x_fC_2^-(t)]\}\end{aligned} \tag{13}$$

and

$$N(t)=\frac{2\pi\hbar}{M|f_3(t)|}(2\pi\langle q^2\rangle)^{1/2}. \tag{14}$$

The functions $f_i(t)$, $R^{\pm\pm}(t)$, and $C_i^\pm(t)$ are given in terms of the symmetrized part $S(t)$ and the antisymmetrized part $A(t)=-(\hbar/2M)G(t)$ of the equilibrium coordinate autocorrelation function $C(t)=\langle q(t)q\rangle=S(t)+iA(t)$ in the following way:

$$f_1(t)=f_2(t)=\frac{\dot{G}(t)}{G(t)}, \tag{15}$$

$$f_3(t)=\frac{1}{G(t)}, \tag{16}$$

$$f_4(t)=-\ddot{G}(t)+\frac{\dot{G}(t)^2}{G(t)}, \tag{17}$$

$$\begin{aligned}R^{++}(t)=\frac{M}{\hbar}\Bigg\{&\frac{\langle p^2\rangle}{M^2}+\frac{f_3(t)}{\langle q^2\rangle}\{2\langle q^2\rangle\dot{S}(t)\\&+f_3(t)[\langle q^2\rangle^2-S^2(t)]\}\Bigg\},\end{aligned} \tag{18}$$

$$\begin{aligned}R^{+-}(t)=\frac{M}{\hbar}\Bigg\{&\ddot{S}(t)-f_1(t)\dot{S}(t)-\frac{f_3(t)}{\langle q^2\rangle}\{\dot{S}(t)S(t)\\&+f_1(t)[\langle q^2\rangle^2-S^2(t)]\}\Bigg\},\end{aligned} \tag{19}$$

$$R^{--}(t)=\frac{M}{\hbar}\left\{\frac{\langle p^2\rangle}{M^2}-\frac{1}{\langle q^2\rangle}[\dot{S}(t)-f_1(t)S(t)]^2+\langle q^2\rangle f_1^2(t)\right\}, \tag{20}$$

$$C_1^+(t)=f_3(t)\frac{S(t)}{\langle q^2\rangle}-f_1(t), \tag{21}$$

$$C_1^-(t)=\frac{\dot{S}(t)}{\langle q^2\rangle}-f_1(t)\frac{S(t)}{\langle q^2\rangle}+f_4(t), \tag{22}$$

$$C_2^+(t)=\frac{M}{\hbar}\left[\frac{\langle p^2\rangle}{M^2}+f_3(t)\dot{S}(t)\right], \tag{23}$$

$$C_2^-(t)=\frac{M}{\hbar}[\ddot{S}(t)-f_1(t)\dot{S}(t)]. \tag{24}$$

In the following we shall give results mainly in terms of $S(t)$ and $G(t)$, which both remain finite in the classical limit while $A(t)$ vanishes. The Laplace transforms of $G(t)$ and $S(t)$ read in terms of the Laplace transform $\hat{\gamma}(z)$ of the damping kernel [10,18]

$$\hat{G}(z)=[z^2+\omega_0^2+z\hat{\gamma}(z)]^{-1},$$

$$\hat{S}(z)=\frac{1}{M\beta}\sum_{n=-\infty}^{\infty}\frac{z}{\nu_n^2-z^2}[\hat{G}(z)-\hat{G}(|\nu_n|)], \tag{25}$$

where $\nu_n=2\pi n/\hbar\beta$. Finally,

$$\langle q^2\rangle=S(0)=\frac{1}{M\beta}\sum_{n=-\infty}^{\infty}[\omega_0^2+\nu_n^2+|\nu_n|\hat{\gamma}(|\nu_n|)]^{-1}, \tag{26}$$

$$\langle p^2\rangle=-M^2\ddot{S}(0)=\frac{M}{\beta}\sum_{n=-\infty}^{\infty}\frac{\omega_0^2+|\nu_n|\hat{\gamma}(|\nu_n|)}{\omega_0^2+\nu_n^2+|\nu_n|\hat{\gamma}(|\nu_n|)}$$

are the equilibrium variances.

III. MASTER EQUATION

We now want to investigate the conditions under which the time evolution of the reduced density matrix can be described by a master equation

$$\frac{\partial}{\partial t}\rho(x,r,t)=\mathcal{L}\left(x,r,\frac{\partial}{\partial x},\frac{\partial}{\partial r},t\right)\rho(x,r,t) \tag{27}$$

with a generally time-dependent Liouville operator $\mathcal{L}$. Since Σ is quadratic in x and r, it is sufficient to make the ansatz

$$\begin{aligned}\mathcal{L}=L_c(t)&+\frac{\hbar}{M}\left(\frac{\partial}{\partial x},\frac{\partial}{\partial r}\right)\begin{pmatrix}X_{xx}(t) & X_{xr}(t)\\ X_{rx}(t) & X_{rr}(t)\end{pmatrix}\begin{pmatrix}\dfrac{\partial}{\partial x}\\ \dfrac{\partial}{\partial r}\end{pmatrix}\\ &+(x,r)\begin{pmatrix}Y_{xx}(t) & Y_{xr}(t)\\ Y_{rx}(t) & Y_{rr}(t)\end{pmatrix}\begin{pmatrix}\dfrac{\partial}{\partial x}\\ \dfrac{\partial}{\partial r}\end{pmatrix}\\ &-\frac{M}{\hbar}(x,r)\begin{pmatrix}Z_{xx}(t) & Z_{xr}(t)\\ Z_{rx}(t) & Z_{rr}(t)\end{pmatrix}\begin{pmatrix}x\\ r\end{pmatrix}\end{aligned} \tag{28}$$

with complex parameter $L_c(t)$ and complex matrices $\mathbf{X}(t)$, $\mathbf{Y}(t)$, and $\mathbf{Z}(t)$. Without loss of generality we may put $X_{rx}(t)=X_{xr}(t)$ and $Z_{rx}(t)=Z_{xr}(t)$. Hence, there are eleven time-dependent functions in the ansatz (28). Using Eqs. (8), (12), (13), and (28) to write the left-hand and right-hand sides of Eq. (27) in explicit form, we find that a master equation exists for arbitrary preparation functions provided that 15 ordinary differential equations are fulfilled. Three of these, obtained by comparing the coefficients of $r_f^2, r_f r_i, r_i^2$ read

$$0=Z_{rr}(t)+X_{xx}(t)f_1(t)^2-iY_{rx}(t)f_1(t),$$

$$0=-iY_{rx}(t)+2X_{xx}(t)f_1(t),$$

$$0=X_{xx}(t)f_4(t)^2.$$

Inserting the solution $X_{xx}(t)=Y_{rx}(t)=Z_{rr}(t)=0$ into the 12 remaining equations, one finds that the conditions obtained by comparing the coefficients of 1 and $r_f x_i$ imply $L_c(t)=Y_{rr}(t)$. Taking this into account, we are left with the following set of 11 equations:

$$\dot{f}_1(t)=2iX_{xr}(t)f_1(t)^2+[Y_{rr}(t)+Y_{xx}(t)]f_1(t)+2iZ_{xr}(t), \tag{29}$$

$$\dot{f}_2(t)=2iX_{xr}(t)f_4(t)f_3(t), \tag{30}$$

$$\dot{f}_3(t)=[Y_{rr}(t)+2iX_{xr}(t)f_1(t)]f_3(t), \tag{31}$$

$$\dot{f}_4(t)=[Y_{xx}(t)+2iX_{xr}(t)f_1(t)]f_4(t), \tag{32}$$

$$\dot{R}^{++}(t)=-4iX_{xr}(t)f_3(t)R^{+-}(t)+2X_{rr}(t)f_3^2(t), \tag{33}$$

$$\begin{aligned}\dot{R}^{+-}(t)=[Y_{xx}(t)+2iX_{xr}(t)f_1(t)]R^{+-}(t)+[-2X_{rr}(t)f_1(t)\\ -2iX_{xr}(t)R^{--}(t)+iY_{xr}(t)]f_3(t),\end{aligned} \tag{34}$$

$$\begin{aligned}\dot{R}^{--}(t)=2[Y_{xx}(t)+2iX_{xr}(t)f_1(t)]R^{--}(t)+2Z_{xx}(t)\\ -2iY_{xr}(t)f_1(t)+2X_{rr}(t)f_1^2(t),\end{aligned} \tag{35}$$

$$\dot{C}_1^+(t)=-2iX_{xr}(t)f_3(t)C_1^-(t), \tag{36}$$

$$\dot{C}_1^-(t)=[Y_{xx}(t)+2iX_{xr}(t)f_1(t)]C_1^-(t), \tag{37}$$

$$\dot{C}_2^+(t)=-2iX_{xr}(t)f_3(t)C_2^-(t), \tag{38}$$

$$\dot{C}_2^-(t)=[Y_{xx}(t)+2iX_{xr}(t)f_1(t)]C_2^-(t). \tag{39}$$

Since only seven functions of the ansatz (28) remain to be determined, the set (29)–(39) will be seen to have no solution in general. To demonstrate this explicitly, let us first disregard Eqs. (36)–(39), which stem from comparing coefficients involving the coordinates $\bar{x}$ and $\bar{r}$ of the preparation function. The remaining set of equations (29)–(35) has a unique solution and, the resulting Liouville operator can be written in the form

$$\begin{aligned}\mathcal{L}(t)=\frac{i\hbar}{M}\frac{\partial^2}{\partial x\partial r}-\frac{iM}{\hbar}\gamma_q(t)rx-\gamma_p(t)x\frac{\partial}{\partial x}-\frac{iM}{\hbar}D_q(t)x\frac{\partial}{\partial r}\\ -\frac{M^2}{\hbar^2}D_p(t)x^2.\end{aligned} \tag{40}$$

To see this one first notes that Eqs. (30), (31), and (33) give

$$X_{rr}(t)=Y_{rr}(t)=0,\quad X_{xr}(t)=i/2. \tag{41}$$

The remaining four functions are then readily determined. For later convenience they are expressed in terms of the four functions introduced in Eq. (40), which are given by

$$\gamma_q(t) = -2iZ_{xr}(t) = \frac{\ddot{G}^2(t) - \dot{G}(t)\dddot{G}(t)}{\dot{G}^2(t) - G(t)\ddot{G}(t)},$$

$$\gamma_p(t) = -Y_{xx}(t) = \frac{G(t)\dddot{G}(t) - \dot{G}(t)\ddot{G}(t)}{\dot{G}^2(t) - G(t)\ddot{G}(t)}, \tag{42}$$

$$D_q(t) = \frac{i\hbar}{M}Y_{xr}(t) = \gamma_q(t)\langle q^2\rangle - \frac{\langle p^2\rangle}{M^2} + \frac{S(t)[\gamma_p(t)X(t) + \dot{X}(t)]}{\langle q^2\rangle G(t)} + \gamma_p(t)Y(t) + \dot{Y}(t),$$

$$D_p(t) = \frac{\hbar}{M}Z_{xx}(t) = \gamma_p(t)\frac{\langle p^2\rangle}{M^2} + \frac{\dot{S}(t)[\gamma_p(t)X(t) + \dot{X}(t)]}{\langle q^2\rangle G(t)} + \frac{\dot{G}(t)[\gamma_p(t)Y(t) + \dot{Y}(t)]}{G(t)},$$

where we have introduced

$$X(t) = \dot{G}(t)S(t) - G(t)\dot{S}(t),$$
$$Y(t) = G(t)\ddot{S}(t) - \dot{G}(t)\dot{S}(t). \tag{43}$$

Inserting $X_{xr}(t) = i/2$ and $Y_{rr}(t) = 0$ into Eqs. (36)-(39), it is readily seen that Eq. (36) and (38) are already satisfied. However, Eqs. (37) and (39) are only fulfilled provided that

$$\ddot{S}(t) + \gamma_p(t)\dot{S}(t) + \gamma_q(t)S(t) = 0, \tag{44}$$

$$\dddot{S}(t) + \gamma_p(t)\ddot{S}(t) + \gamma_q(t)\dot{S}(t) = 0. \tag{45}$$

Differentiating Eq. (44) and subtracting Eq. (45) we obtain

$$\dot{\gamma}_p(t)\dot{S}(t) + \dot{\gamma}_q(t)S(t) = 0. \tag{46}$$

Now for $\dot{\gamma}_p(t) \neq 0$ this gives

$$\frac{\dot{S}(t)}{S(t)} = -\frac{\dot{\gamma}_q(t)}{\dot{\gamma}_p(t)} = \frac{\dot{G}(t)}{G(t)}, \tag{47}$$

where the last equation follows by means of Eq. (42). This implies

$$S(t) = cG(t) \quad (t \geqslant 0), \tag{48}$$

where c is a real constant. Clearly, this condition, which is equivalent to Onsager's regression hypothesis [3], is never met exactly whatever the form of the damping kernel. To see this explicitly we note that the Taylor series of $S(t)$ and $G(t)$ start according to

$$S(t) = \langle q^2\rangle - \frac{\langle p^2\rangle}{2M^2}t^2 + O(t^4),$$
$$G(t) = t + O(t^3). \tag{49}$$

On the other hand, in the case $\dot{\gamma}_p(t) = 0$, the condition (46) is only fulfilled if $\gamma_p(t)$ and $\gamma_q(t)$ are both independent of time. Further $S(t)$ must be of the form

$$S(t) = d_1 e^{-\lambda_1 t} + d_2 e^{-\lambda_2 t} \quad (t \geqslant 0), \tag{50}$$

where d_1 and d_2 are complex constants and $\lambda_{1/2} = \gamma_p/2 \pm i\sqrt{\gamma_q - \gamma_p^2/4}$. Moreover, from Eq. (42) we see that $\gamma_p(t)$ is constant provided that $G(t)$ is of the form

$$G(t) = c_1 e^{-\lambda_1 t} + c_2 e^{-\lambda_2 t} \quad (t \geqslant 0), \tag{51}$$

where c_1 and c_2 are complex constants. It is now easily seen that Eqs. (50) and (51) never hold exactly except for $\gamma_p = 0$, which means in the absence of damping. Hence there is no *exact* master equation for the damped harmonic oscillator with a Liouville operator $\mathcal{L}$ *independent* of the preparation function.

In the remainder of this work we first consider specific initial preparations for which the time evolution is described exactly by a time-dependent Liouville operator. We then give examples for approximate Liouville operators valid for particular types of damping.

IV. LIOUVILLE OPERATORS FOR SPECIAL INITIAL PREPARATIONS

In the previous section we have shown that there is no exact Liouville operator that is independent of the preparation function. However, for certain preparations the set of equations (29)–(39), which determine the time-dependent parameters in $\mathcal{L}$ can be reduced allowing for an exact solution. In the following three types of initial states will be considered.

A. Thermal initial condition

Let us first consider a system that is initially in a state

$$\langle q|W(0)|q'\rangle = r(q,q')\langle q|W_\beta|q'\rangle, \tag{52}$$

where $r(q,q')$ is an arbitrary function of q and q'. This initial condition allows in Eq. (9) only for operators O_j and O_j' that are diagonal in position space. It can be used to describe initial states resulting from position measurements but excludes measurements of velocities or variables that couple to the position and the momentum. Following Hakim and Ambegaokar [19] we call Eq. (52) a *thermal initial condition*. Inspection of Eq. (10) shows that the corresponding preparation function is given by

$$\lambda(x_i, r_i, \overline{x,r}) = r(x_i, r_i)\,\delta(\overline{x} - x_i)\,\delta(\overline{r} - r_i). \tag{53}$$

This form of the preparation function has the consequence that only the difference of Eqs. (37) and (32) and likewise the difference of Eqs. (39) and (34) must be fulfilled. Since Eqs. (36) and (38) are again satisfied as a consequence of Eq. (41), there is indeed an exact solution

$$\gamma_q(t) = \frac{\dot{G}(t)\ddot{S}(t) - \ddot{G}(t)\dot{S}(t)}{\dot{G}(t)S(t) - G(t)\dot{S}(t)}, \tag{54}$$

$$\gamma_p(t) = \frac{G(t)\ddot{S}(t) - \ddot{G}(t)S(t)}{\dot{G}(t)S(t) - G(t)\dot{S}(t)},$$

$$D_q(t)=\gamma_q(t)\langle q^2\rangle-\frac{\langle p^2\rangle}{M^2},$$

$$D_p(t)=\frac{\langle p^2\rangle}{M^2}\gamma_p(t).$$

When these coefficients are inserted into Eq. (40), we obtain a time-dependent Liouville operator valid for a large class of initial states. Using the relations

$$x\rightarrow[q,\cdot], \quad r\rightarrow\frac{1}{2}\{q,\cdot\},$$

$$\frac{\partial}{\partial x}\rightarrow\frac{i}{2\hbar}\{p,\cdot\}, \quad \frac{\partial}{\partial r}\rightarrow\frac{i}{\hbar}[p,\cdot] \tag{55}$$

the resulting exact master equation can be written in the form

$$\begin{aligned}\dot{\rho}(t)=&-\frac{iM}{\hbar}\gamma_q(t)\left[q,\frac{1}{2}\{q,\rho(t)\}+\frac{i}{\hbar}\langle q^2\rangle[p,\rho(t)]\right]\\&-\frac{i}{\hbar}\gamma_p(t)\left[q,\frac{1}{2}\{p,\rho(t)\}-\frac{i}{\hbar}\langle p^2\rangle[q,\rho(t)]\right]\\&-\frac{i}{M\hbar}\left[p,\frac{1}{2}\{p,\rho(t)\}-\frac{i}{\hbar}\langle p^2\rangle[q,\rho(t)]\right].\end{aligned} \tag{56}$$

Here $[A,B]=AB-BA$ denotes the commutator and $\{A,B\}=AB+BA$ the anti-commutator.

To see the connection of this master equation with earlier results, we rewrite it in terms of the Wigner transform of the reduced density matrix defined by

$$w(p,q,t)=\int dx\exp\left(-\frac{i}{\hbar}xp\right)\rho(x,q,t). \tag{57}$$

Using the rules

$$[q,\cdot]\rightarrow-\frac{\hbar}{i}\frac{\partial}{\partial p}, \quad \{q,\cdot\}\rightarrow 2q, \tag{58}$$

$$[p,\cdot]\rightarrow\frac{\hbar}{i}\frac{\partial}{\partial q}, \quad \{p,\cdot\}\rightarrow 2p, \tag{59}$$

we find from Eq. (56)

$$\begin{aligned}\dot{w}(p,q,t)=&\left\{\frac{\partial}{\partial p}M\gamma_q(t)\left[q+\frac{\partial}{\partial q}\langle q^2\rangle\right]\right.\\&\left.+\left[\frac{\partial}{\partial p}\gamma_p(t)-\frac{\partial}{\partial q}\frac{1}{M}\right]\left[p+\frac{\partial}{\partial p}\langle p^2\rangle\right]\right\}\\&\times w(p,q,t).\end{aligned} \tag{60}$$

This is of the form of a generalized Fokker-Planck equation. For the classical harmonic oscillator an equation of similar form was found by Adelman [11] based on the generalized classical Langevin equation

$$\ddot{q}(t)+\omega_0^2q(t)+\int_0^t ds\,\gamma(t-s)\dot{q}(s)=\frac{1}{M}\xi(t), \tag{61}$$

with a noise force $\xi(t)$ satisfying

$$\langle\xi(t)\rangle=0 \tag{62}$$

and

$$\langle\xi(t)\xi(0)\rangle=Mk_BT\gamma(t). \tag{63}$$

The time-dependent coefficients of the Adelman equation are given by

$$\gamma_q(t)=\frac{\ddot{C}_{\rm cl}^2(t)-\dot{C}_{\rm cl}(t)\dddot{C}_{\rm cl}(t)}{\dot{C}_{\rm cl}^2(t)-C_{\rm cl}(t)\ddot{C}_{\rm cl}(t)},$$

$$\gamma_p(t)=\frac{C_{\rm cl}(t)\dddot{C}_{\rm cl}(t)-\dot{C}_{\rm cl}(t)\ddot{C}_{\rm cl}(t)}{\dot{C}_{\rm cl}^2(t)-C_{\rm cl}(t)\ddot{C}_{\rm cl}(t)}. \tag{64}$$

Since in the classical limit $S(t)$ reduces to $C_{\rm cl}(t)$, and

$$G(t)=-\frac{M}{k_BT}\dot{C}_{\rm cl}(t) \tag{65}$$

the coefficients (54) reduce to (64). This shows that we have derived an exact quantum mechanical generalization of the Adelman equation.

B. Liouville operator for the time evolution of equilibrium correlations

In this section we investigate the time evolution of equilibrium correlation functions

$$\langle A(t)B\rangle={\rm Tr}(Ae^{-iHt/\hbar}BW_\beta e^{iHt/\hbar}), \tag{66}$$

where A and B are variables of the oscillator and Tr denotes the trace over the Hilbert space of the entire system (oscillator and bath). Inserting three partitions of unity and using

$${\rm Tr}=\int dq{\rm Tr}_R\langle q|\cdot|q\rangle, \tag{67}$$

where ${\rm Tr}_R$ is the trace over the Hilbert space of the bath, Eq. (67) can be written as a fourfold integral:

$$\begin{aligned}\langle A(t)B\rangle=&\int dq_1dq_2dq_3dq_4A(q_2,q_1)B(q_3,q_4)\\&\times P(q_1,q_2,t,q_3,q_4),\end{aligned} \tag{68}$$

where $A(q,q')=\langle q|A|q'\rangle$ and

$$P(q_1,q_2,t,q_3,q_4)={\rm Tr}_R(\langle q_1|e^{-iHt/\hbar}|q_3\rangle\langle q_4|W_\beta e^{iHt/\hbar}|q_2\rangle). \tag{69}$$

This function contains complete information about equilibrium correlation functions.

$P(q_1,q_2,t,q_3,q_4)$ satisfies an exact master equation. To see this we first note that an equilibrium correlation function may be calculated in the following way [10]. One propagates the initial reduced ''density matrix'' $\rho_B(0)=B\rho_\beta$ and takes the expectation value of A after time t. Hence,

$$\langle A(t)B\rangle = \mathrm{tr}[A\rho_B(t)] = \int dq_f dq_f' A(q_f',q_f)\rho_B(q_f,q_f',t), \tag{70}$$

where tr denotes the trace over the Hilbert space of the oscillator. Since the initial reduced ''density matrix'' $B\rho_\beta$ corresponds to the ''density matrix'' BW_β of the entire system, the preparation function reads

$$\lambda_B(q_i,q_i',\overline{q},\overline{q'}) = B(q_i,\overline{q})\,\delta(\overline{q'}-q_i'). \tag{71}$$

Now, using

$$\rho_B(q_f,q_f',t) = \int dq_i dq_i' d\overline{q}\,d\overline{q'}\,J(q_f,q_f',t,q_i,q_i',\overline{q},\overline{q'}) \times \lambda_B(q_i,q_i',\overline{q},\overline{q'}) \tag{72}$$

we arrive at

$$\langle A(t)B\rangle = \int dq_f dq_f' dq_i dq_i' d\overline{q}\,d\overline{q'}\,A(q_f,q_f')B(q_i,\overline{q}) \times J(q_f,q_f',t,q_i,q_i',\overline{q},\overline{q'})\,\delta(\overline{q'}-q_i'), \tag{73}$$

which yields by comparison with Eq. (68)

$$P(q_1,q_2,t,q_3,q_4) = \int dy J(q_1,q_2,t,q_3,y,q_4,y). \tag{74}$$

Inserting the explicit form of the propagating function and after the Gaussian integration over y, we are left with an expression containing $S(t)$ and $G(t)$ only in the combination $S(t)-i\hbar G(t)/2M = C(t)$. One finds

$$P(x_f,r_f,t,x_i,r_i) = \frac{1}{\sqrt{2\pi N(t)}}\exp\left\{ \frac{iM}{2\hbar}\left[\frac{\dot N(t)}{N(t)}(x_f r_f - x_i r_i) + \frac{\dot N(t)\langle q^2\rangle}{N(t)C(t)}(x_i r_f - x_f r_i) - \frac{iM}{\hbar}\left[-\frac{\langle p^2\rangle}{M^2} + \frac{\dot N(t)^2\langle q^2\rangle}{4C(t)^2 N(t)}\right](x_f^2+x_i^2) - \frac{iM}{\hbar}\left(\frac{\ddot N(t)}{C(t)} + \frac{\dot N^2(t)\langle q^2\rangle}{2N(t)C^3(t)}\right) x_f x_i + \frac{i\hbar}{M}\frac{\langle q^2\rangle}{N(t)}(r_f^2+r_i^2) - 2\frac{i\hbar}{M}\frac{C(t)}{N(t)} r_i r_f \right]\right\}, \tag{75}$$

where $r_f = (q_1+q_2)/2$, $x_f = q_1 - q_2$, $r_i = (q_3+q_4)/2$, and $x_i = q_4 - q_3$. Further, we have introduced $N(t) = \langle q^2\rangle^2 - C(t)^2$. In view of the δ function in Eq. (71) there are again fewer conditions that must be satisfied by the time-dependent coefficients of the Liouville operator. In fact, it is easily seen that

$$\dot P(x_f,r_f,t,x_i,r_i) = \mathcal{L}\left(x_f, r_f, \frac{\partial}{\partial x_f}, \frac{\partial}{\partial r_f}, t\right) P(x_f,r_f,t,x_i,r_i), \tag{76}$$

with a Liouville operator $\mathcal{L}$ of the form Eq. (40) with the coefficients

$$\gamma_q(t) = \frac{\ddot C^2(t) - \dot C(t)\dddot C(t)}{\dot C^2(t) - C(t)\ddot C(t)},$$

$$\gamma_p(t) = \frac{C(t)\dddot C(t) - \dot C(t)\ddot C(t)}{\dot C^2(t) - C(t)\ddot C(t)}, \tag{77}$$

$$D_q(t) = \gamma_q(t)\langle q^2\rangle - \frac{\langle p^2\rangle}{M^2},$$

$$D_p(t) = \frac{\langle p^2\rangle}{M^2}\gamma_p(t).$$

Hence $P(x_f,r_f,t,x_i,r_i)$ satisfies the exact evolution equation

$$\dot P(x_f,r_f,t,x_i,r_i) = \left[-\frac{iM}{\hbar}\gamma_q(t)x\left(r + \langle q^2\rangle\frac{\partial}{\partial r}\right) - \left(\gamma_p(t)x + \frac{\hbar}{iM}\frac{\partial}{\partial r}\right)\left(\frac{\partial}{\partial x} + \frac{\langle p^2\rangle}{\hbar^2}x\right)\right] \times P(x_f,r_f,t,x_i,r_i). \tag{78}$$

To illuminate the virtue of this equation we note that it can be used to calculate correlation functions in a quasiclassical manner. Introducing the double Wigner transform

$$\widetilde{P}(p_f,q_f,t,p_i,q_i) = \frac{1}{(2\pi\hbar)^2}\int dx_f dx_i P(x_f,q_f,t,x_i,q_i) \times \exp\left[\frac{i}{\hbar}(-x_f p_f + x_i p_i)\right]. \tag{79}$$

and the Wigner-Moyal transforms of $A(x_f,r_f)$ and $B(x_i,r_i)$ according to

$$\widetilde{A}(p_f,q_f) = \int dx_f \exp\left(-\frac{i}{\hbar}x_f p_f\right) A(x_f,q_f), \tag{80}$$

equilibrium correlations may be written as a double phase-space integral

$$\langle A(t)B\rangle = \int dp_f dq_f dp_i dq_i \widetilde{A}(p_f,q_f)\widetilde{B}(p_i,q_i) \times \widetilde{P}(p_f,q_f,t,p_i,q_i). \tag{81}$$

This means that we can view $\widetilde{P}(p_f,q_f,t,p_i,q_i)$ as a quantum mechanical generalization of the classical joint probability.

The Wigner form of the master equation (78), which is again a generalized Fokker-Planck equation, was first derived by Schramm, Jung, and Grabert [20] on the basis of phenomenological reasoning. Note that the time-dependent coefficients are complex. However, in the classical limit they become real, since the imaginary part of $C(t)$ vanishes, and the generalized Fokker-Planck equation reduces again to the Adelman equation.

C. Factorizing initial preparation

In earlier work it has been frequently assumed [12,17,21,22] that the initial density matrix W_0 of the entire system factorizes according to

$$W_0 = \rho_0 W_R, \tag{82}$$

where ρ_0 is the density matrix of the oscillator, while $W_R = Z_R \exp(-\beta H_R)$ is the canonical density matrix of the unperturbed heat bath. Within our approach this situation cannot be described by a special form of the preparation function $\lambda(q_i, q_i', \overline{q, q'})$, however, factorizing initial states are easily gained by disregarding contributions coming from the imaginary time path integral (see [10] for details). The time evolution of the density matrix is then given by

$$\rho(x_f, r_f, t) = \int dr_i dx_i J_{\mathrm{FV}}(x_f, r_f, t, x_i, r_i) \rho(x_i, r_i, 0), \tag{83}$$

where

$$J_{\mathrm{FV}}(x_f, r_r, t, x_i, r_i) = \frac{M|f_3(t)|}{2\pi\hbar} \exp\left[\frac{i}{\hbar} \Sigma_{\mathrm{FV}}(x_f, r_r, t, x_i, r_i)\right] \tag{84}$$

and

$$\begin{aligned}\Sigma_{\mathrm{FV}}(x_f, r_f, t, x_i, r_i) = {} & M[x_f r_f f_1(t) + x_i r_i f_2(t) - x_i r_f f_3(t) \\ & - x_f r_i f_4(t)] + \frac{i}{2} M[x_i^2 R_{\mathrm{FV}}^{++}(t) \\ & + 2 x_f x_i R_{\mathrm{FV}}^{+-}(t) + x_f^2 R_{\mathrm{FV}}^{--}(t)].\end{aligned} \tag{85}$$

The index FV refers to Feynman and Vernon [21]. Note that the definitions of $R^{\pm\pm}(t)$ are now modified:

$$\begin{aligned}R_{\mathrm{FV}}^{++}(t) &= f_3^2(t) K_q(t), \\ R_{\mathrm{FV}}^{+-}(t) &= f_3(t)[\tfrac{1}{2}\dot{K}_q(t) - f_1(t) K_q(t)], \\ R_{\mathrm{FV}}^{--}(t) &= K_p(t) - f_1(t)\dot{K}_q(t) + f_1^2(t) K_q(t),\end{aligned} \tag{86}$$

where the $f_i(t)$ are given by Eqs. (15)–(17) and

$$\begin{aligned}K_q(t) &= \frac{1}{M}\int_0^t ds \int_0^t du K'(s-u) G(s) G(u), \\ K_p(t) &= \frac{1}{M}\int_0^t ds \int_0^t du K'(s-u) \dot{G}(s) \dot{G}(u).\end{aligned} \tag{87}$$

The function $K'(t)$ is the real part of the real-time influence kernel. Its Laplace transform is related to the Laplace transform of the damping kernel $\hat{\gamma}(z)$ by [10]

$$\hat{K}'(z) = \frac{M}{\hbar\beta} \sum_{n=-\infty}^{\infty} \frac{z}{z^2 - \nu_n^2} [z\hat{\gamma}(z) - |\nu_n| \hat{\gamma}(|\nu_n|)]. \tag{88}$$

The integrals in Eq. (87) cannot be expressed in terms of $S(t)$ and $G(t)$ as is the case for $R^{\pm\pm}(t)$. The quadratic ansatz (28) now leads to Eqs. (29)–(35) with the functions $R^{\pm\pm}(t)$ replaced by $R_{\mathrm{FV}}^{\pm\pm}(t)$. There are no equations replacing (36)–(39) since oscillator and bath are uncorrelated in the initial state. This set allows for a solution by the time-dependent parameters

$$\begin{aligned}\gamma_q(t) &= \frac{\ddot{G}^2(t) - \dot{G}(t)\dddot{G}(t)}{\dot{G}^2(t) - G(t)\ddot{G}(t)} \\ \gamma_p(t) &= \frac{G(t)\dddot{G}(t) - \dot{G}(t)\ddot{G}(t)}{\dot{G}^2(t) - G(t)\ddot{G}(t)}, \\ D_q(t) &= \frac{\hbar}{M}\left[\frac{1}{2}\ddot{K}_q(t) - K_p(t) + \gamma_q(t) K_q(t) + \frac{\gamma_p(t)}{2}\dot{K}_q(t)\right], \\ D_p(t) &= \frac{\hbar}{M}\left[\frac{1}{2}\dot{K}_p(t) + \frac{\gamma_q(t)}{2}\dot{K}_q(t) + \gamma_p(t) K_p(t)\right].\end{aligned} \tag{89}$$

The resulting master equation is equivalent to the result by Haake and Reibold [12,23] who derived it directly from microscopic dynamics. Their formulas for $K_q(t)$ and $K_p(t)$ contain frequency integrals that may be evaluated to obtain Eq. (87). Later, this equation was rederived by Hu, Paz, and Zhang [22] from the path integral representation. The equivalence can most easily be seen using the simplified derivation given by Paz [24].

We mention that for factorizing initial conditions the classical limit does not yield the Adelman equation [25]. The generalized Fokker-Planck operator differs by terms that persist over times of the order of the relaxation time. This means that switching on the interaction with the bath at $t=0$ pathologically affects also the long-time behavior of the system. Usually, the oscillator and the bath are integral parts of the same system and the factorization assumption is not appropriate.

V. LIOUVILLE OPERATORS FOR PARTICULAR TYPES OF DAMPING

So far we have searched for exact master equations. Let us now turn to the question whether for particular types of damping the dynamics may be described in terms of approximate Liouville operators valid for arbitrary preparation functions. Thus, we have to find circumstances under which $S(t)$ and $G(t)$ take the forms of Eqs. (50) and (51). Using Eq. (42) we see that in this case the Liouville operator (40) is time independent and the coefficients $D_p(t) = D_p$ and $D_q(t) = D_q$ read

$$D_q = \gamma_q \langle q^2 \rangle - \frac{\langle p^2 \rangle}{M^2},$$

$$D_p = \gamma_p \frac{\langle p^2 \rangle}{M^2}. \tag{90}$$

Note that this result is independent of the coefficients c_1, c_2, d_1, d_2 in Eqs. (50) and (51). The Wigner transform of the density matrix then obeys Eq. (60) with time-independent coefficients γ_q and γ_p. This result is in accordance with the findings of Talkner [9] on the most general form of a Liouville operator in Wigner form compatible with the correct equilibrium expectation values.

To see explicitly when conditions (50) and (51) hold, we first investigate the consequences of Eq. (51). By virtue of Ehrenfest's theorem $G(t)$ is purely classical [6], since it is related to the response function

$$\chi(t) = \theta(t) \frac{1}{M} G(t), \tag{91}$$

describing the mean nonequilibrium displacement in response to an applied force. Hence Eq. (51) implies that the classical equation of motion is solved by a sum of two exponentials. This means essentially Ohmic damping.

A. Ohmic damping

For strictly Ohmic damping $\hat{\gamma}(z) = \gamma$ and condition (51) holds exactly. We have

$$G(t) = c_1 e^{-\lambda_1 t} + c_2 e^{-\lambda_2 t}, \tag{92}$$

where $\lambda_{1/2} = \gamma/2 \pm i\sqrt{\omega_0^2 - \gamma^2/4}$ and $c_{1/2} = \mp(\lambda_1 - \lambda_2)^{-1}$. Thus $\gamma_p = \gamma$ and $\gamma_q = \omega_0^2$. We still have to examine whether $S(t)$ fulfills Eq. (50). To this purpose it is useful to note that Eq. (25) gives for the Fourier transforms

$$\tilde{S}(\omega) = \frac{i\hbar}{2M} \coth\left(\frac{\omega\pi}{\nu}\right) \tilde{G}(\omega), \tag{93}$$

where

$$\tilde{S}(\omega) = \hat{S}(-i\omega) + \hat{S}(i\omega),$$
$$\tilde{G}(\omega) = \hat{G}(-i\omega) - \hat{G}(i\omega). \tag{94}$$

The latter relations follow from the fact that $S(t)$ is symmetric and $G(t)$ antisymmetric. Of course, Eq. (93) is just the familiar fluctuation-dissipation theorem. Now, performing the inverse Fourier transform we find [6]

$$S(t) = d_1 e^{-\lambda_1 t} + d_2 e^{-\lambda_2 t} - \Gamma(t), \tag{95}$$

where

$$d_{1/2} = c_{1/2} \frac{\hbar}{2M} \cot\left(\frac{\pi\lambda_{1/2}}{\nu}\right),$$

$$\Gamma(t) = \frac{2\gamma}{M\beta} \sum_{n=1}^{\infty} \frac{\nu_n \exp(-\nu_n t)}{(\omega_0^2 + \nu_n^2)^2 - \gamma^2 \nu_n^2}. \tag{96}$$

This means that we have to find conditions under which $\Gamma(t)$ may be disregarded. Now $\Gamma(t)$ decays at least as $\exp(-\nu t)$, where $\nu \equiv \nu_1 = 2\pi k_B T/\hbar$. Therefore, for temperatures $T \gg \hbar\gamma/4\pi k_B$, we have $\nu \gg \mathrm{Re}(\lambda_1)$, $\mathrm{Re}(\lambda_2)$, and $\Gamma(t)$ decays faster than the first two terms in Eq. (95). Hence for $t \gg \nu^{-1}$ the last term in Eq. (95) may be disregarded and $S(t)$ is of the form of Eq. (51). However, in the strictly Ohmic case we do not have a well-defined Liouville operator since the sum (26) for $\langle p^2 \rangle$ is logarithmically divergent leading to associated divergences of the coefficients D_q and D_p in Eq. (90). To avoid this divergence we have to take the high frequency behavior of the damping coefficient into account, which implies in realistic cases

$$\lim_{z \to \infty} \hat{\gamma}(z) = 0. \tag{97}$$

B. Drude regularization

A more realistic behavior of the damping coefficient is modeled by

$$\hat{\gamma}(z) = \frac{\gamma\omega_D}{z + \omega_D}, \tag{98}$$

often referred to as Drude damping. Using Eq. (25) we then find

$$G(t) = c_1 e^{-\lambda_1 t} + c_2 e^{-\lambda_1 t} + c_3 e^{-\lambda_3 t}, \tag{99}$$

where

$$\lambda_{1/2} = \alpha \pm i\eta, \quad \lambda_3 = \delta \tag{100}$$

are the solutions of

$$z^3 - \omega_D z^2 + (\omega_0^2 + \gamma\omega_D) z - \omega_0^2 \omega_D = 0 \tag{101}$$

and

$$c_1 = -\frac{i}{2\eta} \frac{\alpha - i\eta + \delta}{\alpha + i\eta - \delta},$$

$$c_2 = \frac{i}{2\eta} \frac{\alpha + i\eta + \delta}{\alpha - i\eta - \delta}, \tag{102}$$

$$c_3 = \frac{2\alpha}{(\alpha - i\eta - \delta)(\alpha + i\eta - \delta)}.$$

This result holds also in the overdamped case where η becomes imaginary. To calculate $S(t)$ we have to evaluate the inverse Fourier transform of Eq. (93) by contour integration. Using Eq. (99) one finds [26]

$$S(t) = d_1 e^{-\lambda_1 t} + d_2 e^{-\lambda_1 t} + d_3 e^{-\lambda_3 t} - \Gamma(t), \tag{103}$$

where

$$d_i = c_i \frac{\hbar}{2M} \cot\left(\frac{\pi\lambda_i}{\nu}\right), \quad i = 1,2,3, \tag{104}$$

$$\Gamma(t) = \frac{2\gamma}{M\beta} \sum_{n=1}^{\infty} \frac{\omega_D^2 \nu_n e^{-\nu_n t}}{(\lambda_1^2 - \nu_n^2)(\lambda_2^2 - \nu_n^2)(\lambda_3^2 - \nu_n^2)}. \tag{105}$$

From these results we see that $S(t)$ and $G(t)$ are of the forms (50) and (51), respectively, if the exponentials $\exp(-\lambda_3 t)$ and $\exp(-\nu_n t)$ decay much faster than $\exp(-\lambda_{1/2}t)$. This is the case for

$$\mathrm{Re}(\lambda_1), \quad \mathrm{Re}(\lambda_2) \ll \delta, \nu. \tag{106}$$

Using the Vieta relations

$$\begin{aligned} 2\alpha + \delta &= \omega_D, \\ \alpha^2 + \eta^2 &= \omega_0^2 \omega_D / \delta, \\ \alpha^2 + \eta^2 + 2\alpha\delta &= \omega_0^2 + \gamma\omega_D, \end{aligned} \tag{107}$$

we find that $\mathrm{Re}(\lambda_{1/2}) \ll \delta$ implies

$$\alpha \ll \delta. \tag{108}$$

Further, the relations (107) yield

$$\frac{\gamma}{\omega_D} = 2\frac{\alpha}{\delta}\frac{1+(\omega_0/\delta)^2}{(1+2\alpha/\delta)^2}. \tag{109}$$

In view of Eq. (108) this gives [27]

$$\gamma/\omega_D = 2\alpha/\delta - 8(\alpha/\delta)^2 + O((\alpha/\delta)^3). \tag{110}$$

Hence, it is natural to use γ/ω_D as a small parameter to determine the roots of Eq. (101). Up to first order in γ/ω_D one obtains from Eqs. (100) and (101)

$$\begin{aligned} \alpha &= \frac{\gamma}{2}\frac{\omega_D^2}{\omega_D^2 + \omega_0^2}, \\ \eta &= \sqrt{\omega_0^2 + 2\alpha\omega_0^2/\omega_D - \alpha^2}, \\ \delta &= \omega_D - 2\alpha. \end{aligned} \tag{111}$$

These relations are valid for arbitrary ratios of γ and ω_0. Also ω_D/ω_0 is not necessarily large. As a consequence of this analysis we find that only under the conditions

$$\gamma \ll \omega_D, \nu \tag{112}$$

and

$$t \gg \omega_D^{-1}, \nu^{-1}. \tag{113}$$

$S(t)$ may be approximated by the first two terms in Eq. (103). Hence, for sufficiently large Drude cutoff and sufficiently high temperatures, $k_B T \gg \hbar\gamma$, the oscillator dynamics can be described by an approximate Liouville operator with the coefficients

$$\begin{aligned} \gamma_q &= \alpha^2 + \eta^2 \\ \gamma_p &= 2\alpha. \end{aligned} \tag{114}$$

This combines with Eq. (90) to yield the approximate master equation

$$\begin{aligned} \dot{\rho}(t) = &-\frac{iM}{\hbar}(\alpha^2+\eta^2)\left[q, \frac{1}{2}\{q,\rho(t)\} + \frac{i}{\hbar}\langle q^2\rangle[p,\rho(t)]\right] \\ &-\frac{2i}{\hbar}\alpha\left[q, \frac{1}{2}\{p,\rho(t)\} - \frac{i}{\hbar}\langle p^2\rangle[q,\rho(t)]\right] \\ &-\frac{i}{M\hbar}\left[p, \frac{1}{2}\{p,\rho(t)\} - \frac{i}{\hbar}\langle p^2\rangle[q,\rho(t)]\right], \end{aligned} \tag{115}$$

first derived by Haake and Reibold [12].

The equilibrium variances can be calculated analytically [6]. In the strictly Ohmic limit $\omega_D \to \infty$, $\langle q^2\rangle$ is a regular expression but $\langle p^2\rangle$ diverges logarithmically. If we disregard terms of the order ω_0/ω_D, γ/ω_D, ν/ω_D, the divergent part of $\langle p^2\rangle$ is given by

$$\frac{M\hbar\gamma}{\pi}\ln\left(\frac{\omega_D}{\nu}\right).$$

Hence, for the classical limit it is not sufficient to have $\nu \gg \omega_0$, that is $k_B T \gg \hbar\omega_0$, rather we also need $\gamma\ln(\omega_D/\nu) \ll \nu$. Thus the strictly Ohmic limit $\omega_D \to \infty$ can only be taken after the high temperature limit. With this sequence of limits we obtain the classical coefficients

$$\gamma_q = \omega_0^2, \quad \gamma_p = \gamma, \quad D_q = 0, \quad D_p = \frac{\gamma k_B T}{M}. \tag{116}$$

The associated Liouville operator is equivalent to the classical Fokker-Planck operator of the Kramers equation [28]. We stress again that the results in this section remain valid for strong damping provided Eq. (112) is satisfied.

C. Weak damping with arbitrary frequency dependence

In this section we show that $S(t)$ and $G(t)$ always take the forms of Eqs. (50) and (51) in the limit of weak damping. Let us assume that the damping kernel $\gamma(t)$ has a high frequency cutoff ω_c and that its Laplace transform is an analytic function in the vicinity of $-i\omega_0$. We introduce a typical damping strength by

$$\gamma_c = \int_0^\infty ds\, \gamma(s)\cos(\omega_0 s). \tag{117}$$

Then the weak damping condition is

$$\gamma_c \ll \omega_0, \omega_c, \nu. \tag{118}$$

Apart from this $\gamma(t)$ is not assumed to have additional properties.

In the limit considered one can determine the poles of $\hat{G}(z)$ from Eq. (25) perturbatively. To first order in γ_c the poles are

$$\lambda_{1/2} = \frac{\gamma_c}{2} \pm i\left(\omega_0 + \frac{\gamma_s}{2}\right), \tag{119}$$

where

$$\gamma_c + i\gamma_s = \hat{\gamma}(-i\omega_0). \tag{120}$$

Performing the inverse Laplace transform of Eq. (25) we find

$$G(t)=\frac{i}{2\omega_0}\left(e^{-\lambda_1 t}-e^{-\lambda_2 t}\right) \tag{121}$$

with residues in zeroth order. Higher-order corrections would depend on the specific form of $\hat{\gamma}(z)$ but need not be determined because the diffusion constants (90) are independent of the residues. Thus, in the weak damping limit $G(t)$ is of the form of Eq. (51). From Eq. (93) we see that $\tilde{S}(\omega)$ has the same poles as $\tilde{G}(\omega)$ and, in addition, poles at $i\nu_n$ (n integer). As shown above, we can disregard the terms coming from the poles at $i\nu_n$ for times greater than the thermal relaxation time ν^{-1}. Thus, $S(t)$ is effectively of the form of Eq. (50). Inserting $\lambda_{1/2}$ into Eq. (90), we find to leading order in the damping strength

$$\begin{aligned}D_q&=\frac{1}{M\beta}\sum_{n=-\infty}^{\infty}\frac{\gamma_s\omega_0-|\nu_n|\hat{\gamma}(|\nu_n|)}{\omega_0^2+\nu_n^2},\\ D_p&=\frac{1}{M\beta}\sum_{n=-\infty}^{\infty}\frac{\gamma_c\omega_0^2}{\omega_0^2+\nu_n^2}=\gamma_c\frac{\hbar\omega_0}{2M}\coth\left(\frac{\omega_0\pi}{\nu}\right).\end{aligned} \tag{122}$$

This result can be expressed in terms of the Laplace transform (88) of the real part of the influence kernel

$$D_p+i\omega_0 D_q=\frac{\hbar}{M^2}\hat{K}'(-i\omega_0). \tag{123}$$

Thus, D_q and D_p are essentially given by the sine and cosine moments of $K'(t)$. With $K_c+iK_s=\hat{K}'(-i\omega_0)$ the master equation takes the form

$$\begin{aligned}\dot{\rho}(t)=&-\frac{i}{\hbar}\left[\frac{p^2}{2M}+\frac{M(\omega_0^2+\omega_0\gamma_s)}{2}q^2,\rho(t)\right]\\ &-\frac{i\gamma_c}{2\hbar}[q,\{p,\rho(t)\}]-\frac{K_s}{M\hbar\omega_0}[p,[q,\rho(t)]]\\ &-\frac{K_c}{\hbar}[q,[q,\rho(t)]].\end{aligned} \tag{124}$$

This general weak coupling master equation is given in terms of four dissipation coefficients. γ_s leads to a frequency shift and may be absorbed by renormalizing ω_0. γ_c is the classical damping coefficient. The coefficients K_c and K_s depend on the temperature. While K_c equals $M\gamma_c/\hbar$ times the average energy of a quantum oscillator of frequency ω_0, K_s depends on the specific form of $\hat{\gamma}(z)$ and can be calculated analytically only in certain cases. One of these is the Drude model. Then the moments $\gamma_{c/s}$ and $K_{c/s}$ are readily evaluated to read

$$\begin{aligned}\gamma_c&=\frac{\gamma\omega_D^2}{\omega_D^2+\omega_0^2},\\ \gamma_s&=\frac{\gamma\omega_0\omega_D}{\omega_D^2+\omega_0^2},\\ K_c&=\frac{\gamma_c\omega_0 M}{2}\coth\left(\frac{\omega_0\pi}{\nu}\right),\\ K_s&=\frac{\omega_0 M}{\pi}\left\{\frac{\nu\gamma_s}{2\omega_0}+\gamma_c\mathrm{Re}\left[\psi\left(1+i\frac{\omega_0}{\nu}\right)-\psi\left(1+\frac{\omega_D}{\nu}\right)\right]\right\}.\end{aligned} \tag{125}$$

In the strictly Ohmic limit $\omega_D\to\infty$ we have $\gamma_c=\gamma$, $\gamma_s=0$ but

$$K_s=-\frac{\gamma\omega_0 M}{\pi}\ln\left(\frac{\omega_D}{\nu}\right) \tag{126}$$

is logarithmically divergent. The master equation (124) with the coefficients (125) can be compared with the well-known Agarwal equation [13]

$$\begin{aligned}\dot{\rho}(t)=&-\frac{i}{\hbar}\left[\frac{p^2}{2M}+\frac{M\omega_0^2}{2}q^2,\rho(t)\right]-\frac{i\kappa}{\hbar}[q,\{p,\rho(t)\}]\\ &-\kappa\frac{M\omega_0}{\hbar}\coth\left(\frac{\omega_0\pi}{\nu}\right)[q,[q,\rho(t)]],\end{aligned} \tag{127}$$

which was derived with the help of projection operator techniques from the same microscopic model using the Born approximation in conjunction with a short-memory approximation. As a main difference, we see that in Agarwal's equation the K_s term is absent. This term is only negligible if

$$\omega_0\ll\omega_D\ll\nu \tag{128}$$

Hence, the master equation (124) is a generalization of the Agarwal equation.

D. Connection to Lindblad theory

The approximate time-independent Liouville operators studied above describe the dynamics after the decay of fast transients. Markovian Liouville operators such as these are often discussed in the context of Lindblad theory [29]. This theory establishes the most general form of generators $\mathcal{L}$ of dissipative quantum dynamics $\dot{\rho}(t)=\mathcal{L}\rho(t)$ preserving the positivity of density operators. The Lindblad master equation reads

$$\dot{\rho}(t)=-\frac{i}{\hbar}[\tilde{H},\rho(t)]+\frac{1}{2\hbar}\sum_i [L_i\rho(t),L_i^{+}]+[L_i,\rho(t)L_i^{+}], \tag{129}$$

where L_l are arbitrary operators and $\tilde{H}$ is a Hermitian operator. Using results by Sandulescu and Scutaru [30] it is easily seen that all the above-derived time-independent Liouville operators are not of Lindblad form. This is not too astonishing since the master equations derived hold only for times $t>t_0$ where t_0 is larger than an inverse cutoff frequency and ν^{-1}. The short time dynamics for $t\lesssim t_0$ reduces the density matrix to a subspace where the fast components have decayed. The Markovian master equation holds within this subspace only, while Lindblad theory requires validity for *any* reduced density matrix. This is of course not necessary as has been emphasized again recently [1,31].

However, we will show that in the weak coupling limit further coarse graining will result in a Lindblad operator. To this aim we first write the weak-coupling master equation (124) in the form

$$\dot{\rho}(t)=\mathcal{L}\rho(t)=\mathcal{L}_0\rho(t)+\gamma\mathcal{L}_1\rho(t). \tag{130}$$

In terms of the usual creation and annihilation operators $a^\dagger$, a we have

$$\mathcal{L}_0=-i\omega_0[a^\dagger a,\cdot]. \tag{131}$$

Using the operators

$$\mathcal{P}_n=\sum_k |k\rangle\langle k+n|, \tag{132}$$

where $|k\rangle$ are the eigenstates of $a^\dagger a$, $\mathcal{L}_0$ may be written as

$$\mathcal{L}_0=\sum_n i\omega_0 n\mathcal{P}_n. \tag{133}$$

Further

$$\begin{aligned}\gamma\mathcal{L}_1={}&-i\frac{\gamma_s}{2}[a^\dagger a,\cdot]+\frac{\gamma_c-i\gamma_s}{4}[a^{\dagger 2},\cdot]-\frac{\gamma_c+i\gamma_s}{4}[a^2,\cdot]\\&+\gamma_\downarrow([a\cdot,a^\dagger]+[a,\cdot a^\dagger])+\gamma_\uparrow([a^\dagger\cdot,a]+[a^\dagger,\cdot a])\\&+\frac{K_c+iK_s}{2M\omega_0}([a^\dagger\cdot,a^\dagger]+[a^\dagger,\cdot a^\dagger])\\&+\frac{K_c-iK_s}{2M\omega_0}([a\cdot,a]+[a,\cdot a]),\end{aligned} \tag{134}$$

where we have introduced

$$\gamma_{\downarrow\uparrow}=\frac{K_c}{2M\omega_0}\pm\frac{\gamma_c}{4}=\frac{\gamma_c}{4}\left[\coth\left(\frac{\omega_0\pi}{\nu}\right)\pm 1\right]. \tag{135}$$

The time evolution is formally given by

$$\rho(t)=e^{\mathcal{L}t}\rho(0). \tag{136}$$

Now we rewrite this by a well-known operator identity

$$\rho(t)=e^{\mathcal{L}_0 t}\rho(0)+\gamma\int_0^t ds e^{\mathcal{L}_0(t-s)}\mathcal{L}_1 e^{\mathcal{L}s}\rho(0). \tag{137}$$

For weak damping and times $t\ll\gamma^{-1}$ the operator $e^{\mathcal{L}s}$ in the integrand may be replaced by $e^{\mathcal{L}_0 s}$. Inserting then Eq. (133) into Eq. (137) we find

$$\rho(t)=e^{\mathcal{L}_0 t}\left[1+\gamma\sum_{n,m}\int_0^t ds e^{i\omega_0(n-m)s}\mathcal{P}_n\mathcal{L}_1\mathcal{P}_m\right]\rho(0). \tag{138}$$

Further coarse graining is achieved by demanding

$$t\gg\omega_0^{-1}. \tag{139}$$

Then, by performing the time integral, the off-diagonal terms $(n\neq m)$ are seen to be smaller than the diagonal terms by a factor $(\omega_0 t)^{-1}$. This means that in the time window $\omega_0^{-1}\ll t\ll\gamma^{-1}$

$$\rho(t)=e^{\mathcal{L}_0 t}[1+\gamma t\widetilde{\mathcal{L}}_1]\rho(0), \tag{140}$$

where we have introduced the *effective* dissipative Liouville operator

$$\widetilde{\mathcal{L}}_1=\sum_n \mathcal{P}_n\mathcal{L}_1\mathcal{P}_n. \tag{141}$$

The density matrix (140) coincides with the solution of the master equation

$$\dot{\rho}(t)=(\mathcal{L}_0+\gamma\widetilde{\mathcal{L}}_1)\rho(t) \tag{142}$$

for

$$t\ll\gamma^{-1}. \tag{143}$$

Thus, within the time window $\omega_0^{-1}\ll t\ll\gamma^{-1}$ the two operators $\mathcal{L}_0+\gamma\mathcal{L}_1$ and $\mathcal{L}_0+\gamma\widetilde{\mathcal{L}}_1$ give the same dynamics. The operator $\widetilde{\mathcal{L}}_1$ may be evaluated further. It is seen that only ''non-rotating'' terms containing equal numbers of creation and annihilation operators survive the coarse graining in time. The resulting master equation

$$\begin{aligned}\dot{\rho}(t)={}&-i\left(\omega_0+\frac{\gamma_s}{2}\right)[a^+a,\rho(t)]+\gamma_\uparrow([a^+\rho(t),a]\\&+[a^+,\rho(t)a])+\gamma_\downarrow([a\rho(t),a^+]+[a,\rho(t)a^+])\end{aligned} \tag{144}$$

was first derived by Weidlich and Haake [14] from a microscopic model for the damped motion of a single mode of the electromagnetic field in a cavity. The generator defined by this master equation is of Lindblad form. However, the resulting mean value equations violate Ehrenfest's theorem, in particular

$$\frac{\partial}{\partial t}\langle q(t)\rangle\neq\langle p(t)\rangle/M. \tag{145}$$

This is due to the fact that on the coarse-grained time scale $\Delta t\gg\omega_0^{-1}$ the variables $p(t)/M\omega_0$ and $q(t)$ exchange identity frequently and only a time-averaged version of the mean value equations must be obeyed.

VI. CONCLUSIONS

Based on results of the path integral technique we have examined quantum master equations for the damped harmonic oscillator. An exact generalized master equation describing the relaxation of initial thermal conditions was derived. This equation was shown to be a quantum mechanical generalization of Adelman's Fokker-Planck equation. We also have given an exact Liouville operator describing the time evolution of equilibrium correlation functions, which likewise reduces to the Adelman Fokker-Planck operator in the classical limit. The fact that two different quantum generalizations of the Adelman operator must be used for the relaxation of expectation values and the regression of fluctuations is intimately connected with the failure of the Onsager regression hypothesis in the quantum regime. Indeed, the two Liouville operators (56) and (76) are only identical if $S(t)$ is proportional to $G(t)$.

Apart from these exact results we have studied in detail the range of parameters leading to quantum master equations with time-independent generator. In the case of strong damping a time-independent Liouville operator is obtained approximately for essentially frequency-independent damping. However, strictly Ohmic damping is ill behaved in the quantum case, and the appropriate generalization of the classical Fokker-Planck process is given by a low frequency Ohmic model with high frequency cutoff such as the Drude model. This is not too amazing since already in the classical limit the Adelman operator becomes time independent only for frequency-independent damping.

On the other hand, in the case of weak damping the detailed frequency dependence of the damping coefficient is unimportant. We have derived a generalized master equation valid for arbitrary weak damping as long as the sine and cosine moments of the damping kernel exist. The new weak-coupling master equation is more general than the well-known Agarwal equation. We have explained why the Liouville operator is not of Lindblad form. However, time coarse graining leads to a generator of Lindblad form. The resulting coarse grained master equation was found to be the Weidlich-Haake equation also known as the quantum optical master equation. Due to the time coarse graining only a time-averaged version of the mean value equation is obeyed leading to an apparent contradiction with the Ehrenfest theorem.

In summary, we have derived several generalized master equations for the damped quantum oscillator for various cases of interest. In view of the results earlier findings were put in proper perspective.

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