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Citation: *The Journal of Chemical Physics* **114**, 10623 (2001);

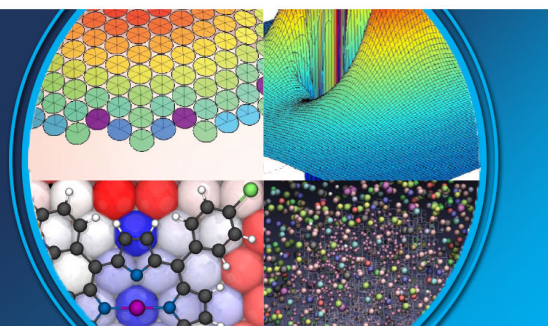
View online: <https://doi.org/10.1063/1.1374537>

View Table of Contents: <http://aip.scitation.org/toc/jcp/114/24>

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AIP | The Journal of
Chemical Physics

PERSPECTIVES



Quantum dissipative master equations: Some exact results

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(Received 18 August 2000; accepted 4 April 2001)

With the help of quantum characteristic functions some exact results are found for two quantum dissipative master equations which contain dissipative Liouvillian operators of the Lindblad-type corresponding to two forms of dissipation mechanisms. Population relaxation is used to demonstrate how dynamic information can be retrieved from these solutions. © 2001 American Institute of Physics. [DOI: 10.1063/1.1374537]

I. INTRODUCTION

Quantum dissipation is a subject of widespread interest in many fields of physics and chemistry.^{1–11} Central to the problem of quantum dissipation is the question of compatibility between quantum mechanics and Markovian motion, which is manifested by the Lindblad exclusion principle of positivity, translational invariance, and approach to canonical equilibrium. While dissipation is well described in classical mechanics by the Langevin or the Fokker–Planck equations, a quantum description of dissipation has remained a challenging task. The difficulty lies in the failure of a Hamiltonian description of those systems, and in the elusiveness of a quantization procedure that ensues.

Among various theoretical techniques applied to quantum dissipation, we mention especially the Redfield approach, the influence functional method, and the semigroup formalism. Popular in nuclear magnetic resonance and optical spectroscopy, the Redfield approach¹² treats the system-bath coupling to second order in perturbation theory. It uses the energy eigenstate representation with no promise of complete positivity. For all but the simplest cases, no analytical solutions can be found for the formidable Redfield equations. The Feynman–Vernon influence functional formulation of quantum dynamics is based on Feynman path integrals.^{13–15} It offers significant advantages when dealing with large-dimensional problems.¹⁶ While an intrinsically quantum mechanical quantity unreachable by classical methods, the influence functional can only be obtained explicitly for very restrictive cases. The semigroup formalism,^{17,18} which is positivity preserving by design, has recently attracted much attention.^{5–9,11} Under the assumption of Markovian dynamics and initial decoupling of system and bath, the semigroup approach adds dissipative dynamics to quantum master equations by means of the Lindblad dissipation operators, which are in fact operators in the system Hilbert space responsible for couplings with the bath. Semigroup methods can treat simultaneously several distinct kinds of system–bath interactions including dissipation, dephasing, collisions and energy transfer. In this paper we confine ourselves to quantum master equations with dissipative Liouvillian operators derived from the semigroup formalism.

Quantum computing is one of the fields in which quantum dissipation finds the most recent applications. An important issue in quantum computer design is to reduce decoherence in the system of interest due to interactions with its environment. Semigroup theories are used to construct for an arbitrary Hamiltonian decoherence-free subspaces within which logical qubits are encoded *not* to decohere.⁶ The Lindblad dissipation operators serves as error generators in quantum computing theories identified with generators of a Lie algebra. Different error generators correspond to different decoherence processes. Quantum computing is of course not the only field where the Lindblad-type master equation applies. In physical chemistry, for example, semigroup theories have been utilized to model dynamics of ultrafast predissociation in a condensed-phase or cluster environment,⁹ and electronic quenching due to coupling of the adsorbate negative ion resonance to the metal electrons in the desorption of neutral molecules on metal surfaces.¹⁰ In nuclear physics, the semigroup formalism is applied to model giant resonances in the nuclear spectra above the neutron emission threshold.⁷ It is therefore of great interest to systematically study the dissipative dynamics within the Lindblad formalism. In this paper we set out to solve analytically two Markovian master equations which contain dissipative Liouvillian operators of the Lindblad type corresponding to two different forms of dissipative mechanisms.

The paper is organized as follows. In Sec. II we introduce quantum master equations and the Lindblad formalism. In Sec. III we demonstrate the methodology by rederiving solutions to the traditional Agarwal master equations (2.5). In Sec. IV we apply the same approach to the Lindblad-type equation (2.16). In Sec. V the equation of motion with pure dephasing Eq. (2.7) is solved exactly up to second moments. We demonstrate in Sec. VI the usefulness of the solutions obtained in previous sections by calculating time-dependent level populations. In Sec. VII we provide some connections between the master equations and the zero-temperature evolution of a coherent state. Discussions are presented in Sec. VIII.

II. QUANTUM MASTER EQUATIONS

In absence of bath memory effects the displacement of a classical oscillator in the Brownian motion $q^c(t)$ follows the equation of motion

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$$\frac{d^2 q^c(t)}{dt^2} + 2\gamma \frac{dq^c(t)}{dt} + \omega_0^2 q^c(t) = f(t), \quad (2.1)$$

where ω_0 is the frequency of the oscillator, 2γ is the phenomenological damping coefficient, and $f(t)$ represents a random perturbation which is a δ -correlated Gaussian process with a zero mean. Equation (2.1) may be replaced by two first-order differential equations¹⁹ which are the Langevin equations for the Brownian motion. Here we are concerned with the Brownian motion of a quantum oscillator; for simplicity, we start with a model Hamiltonian describing only one primary oscillator of frequency ω_0 and mass m coupled to a bath of secondary oscillators of frequency ω_k and mass m_k ($k \neq 0$)

$$\hat{H}_A = \hbar \omega_0 a^\dagger a + \sum_k \hbar \omega_k b_k^\dagger b_k + q \sum_k g_k q_k^b, \quad (2.2)$$

where q and q_k^b are the coordinate observable for the system and the bath oscillators, respectively, which are related to the corresponding boson operators by

$$q = \left(\frac{\hbar}{2m\omega_0} \right)^{1/2} (a^\dagger + a), \quad q_k^b = \left(\frac{\hbar}{2m_k\omega_k} \right)^{1/2} (b_k^\dagger + b_k). \quad (2.3)$$

The Hamiltonian \hat{H}_A represents the excited manifold with one primary oscillator in the Brownian oscillator (BO) model, which is an exactly solvable model serving as a paradigm in the field of quantum dissipation (cf. Appendix A). Our discussion will also be confined to the thermal equilibration of the excited manifold while leaving out the ground state manifold in the BO model. The bath oscillators can be modes of vacuum radiation fields into which an excited atom decays via spontaneous emission. Phonon modes in solids can also be described by those bath oscillators. Adopting the rotating-wave approximation (RWA) widely used in fields such as quantum optics, our model Hamiltonian reduces to

$$\hat{H}_{\text{RWA}} = \hbar \omega_0 a^\dagger a + \hbar \sum_k \omega_k b_k^\dagger b_k + \sum_k g_k (b_k^\dagger a + b_k a^\dagger). \quad (2.4)$$

We note that the rotating-wave approximation neglects the rapidly oscillating terms of Eq. (2.2). For simplicity we shall set $\hbar = 1$ in the master equations in the rest of the paper.

If we consider the primary oscillator as the system of interest, then it is an open system which interacts with the bath oscillators. For open systems, the reduced system density matrix (with the environment degrees of freedom traced out from the full density matrix) is the focus of attention. If the dissipative environment has a fast response, the time evolution of the system density matrix is not dependent on the history (no memory effects), is therefore Markovian. Master equations are usually derived for the reduced density matrix with a damping coefficient representing the dissipative effect of the bath. Agarwal has obtained the Schrödinger-representation master equation for the reduced density operator ρ in the limit of an infinite number of bath oscillators ($\sum_k \rightarrow \int d\omega_k f(\omega_k)$)²⁰

$$\begin{aligned} \frac{\partial \rho}{\partial t} = & -i\omega[a^\dagger a, \rho] - \gamma \bar{n}[a + a^\dagger, [a + a^\dagger, \rho]] \\ & - \gamma(a[a + a^\dagger, \rho] - [a + a^\dagger, \rho]a^\dagger - 2\rho), \end{aligned} \quad (2.5)$$

where $\gamma = \pi f(\omega_0) |g_c(\omega_0)|^2$ is the damping constant, $f(\omega)$ is the density of bath oscillators, $g_c(\omega)$ is the continuum form of g_k , $\bar{n} = (e^{\hbar\omega/k_B T} - 1)^{-1}$, ω is the renormalized frequency of ω_0 ²¹

$$\omega = \omega_0 + \mathcal{P} \int_0^\infty d\omega \frac{f(\omega) |g_c(\omega)|^2}{\omega - \omega_0}, \quad (2.6)$$

and \mathcal{P} stands for the Cauchy principal part. Approximations assumed in deriving Eq. (2.5) includes the Born approximation which treats the bath effects in the lowest order and the short memory hypothesis for the bath.

In a recent attempt to unify the Redfield and Fokker-Planck formulations of quantum dissipation,²² the equation of motion (2.5) is generalized to include an additional β term

$$\begin{aligned} \frac{\partial \rho}{\partial t} = & -i\omega[a^\dagger a, \rho] - \beta[a^\dagger a, [a^\dagger a, \rho]] \\ & - \gamma \bar{n}[a + a^\dagger, [a + a^\dagger, \rho]] - \gamma(a[a + a^\dagger, \rho] \\ & - [a + a^\dagger, \rho]a^\dagger - 2\rho). \end{aligned} \quad (2.7)$$

The β term on the right hand side adds pure dephasing to the original master equation (2.5). Pure dephasing is an important mechanism in a variety of dynamical processes which deprives the system of coherence but leaves the energy conserved. Applications of pure dephasing to interacting many-body systems were implemented by means of time-dependent Hartree-Fock (TDHF).²³

Equation (2.5) has found applications in a variety of fields despite of a well known fact that it violates the positivity requirements of the reduced density matrix.^{24-26,3} Semigroup theories pioneered by Lindblad demonstrated that for harmonic oscillators density-matrix positivity, translational invariance and approach to thermal equilibrium cannot be satisfied simultaneously. In addition, Lindblad¹⁷ showed that a completely positive map can be generated by

$$L_D \rho = \sum_m [V_m, \rho V_m^\dagger] + [V_m \rho, V_m^\dagger] \quad (2.8)$$

from which the equation of motion of the density matrix can be expressed as

$$\frac{\partial \rho}{\partial t} + i[H, \rho] = L_D \rho. \quad (2.9)$$

Here the V_m s are the Lindblad dissipation operators. The interaction Hamiltonian between the system and the bath can be written in terms of V_m and the bath operators B_m ,

$$\hat{H}_{\text{int}} = \sum_m V_m \otimes B_m. \quad (2.10)$$

The dissipation operators by themselves do not guarantee approach of thermal equilibrium. Additional constraints must be imposed to ensure detailed balance. For example, choosing a single dissipation operator⁴

$$V = \mu q + i\nu p, \quad V^\dagger = \mu q - i\nu p, \quad (2.11)$$

where μ , ν are c -number constants which are determined by the quantum fluctuation–dissipation theorem, and the operators q and p are defined as

$$q = a + a^\dagger, \quad (2.12)$$

$$ip = a - a^\dagger, \quad (2.13)$$

one obtains

$$\begin{aligned} \frac{\partial \rho}{\partial t} = & -i[H - 2\mu\nu pq, \rho] - \mu^2[q, [q, \rho]] \\ & - 2i\mu\nu[q, [p, \rho]_+] - \nu^2[p, [p, \rho]]. \end{aligned} \quad (2.14)$$

If two sets of Lindblad dissipation operators are chosen:²⁷

$$V_i = a_i q + b_i p, \quad i = 1, 2 \quad (2.15)$$

one obtains the equation of motion^{27,28}

$$\begin{aligned} \frac{\partial \rho}{\partial t} = & -i[\omega a^\dagger a + \kappa[q, p]_+, \rho] - D_1[q, [q, \rho]] \\ & - D_2[p, [p, \rho]] - D([q, [p, \rho]] + [p, [q, \rho]]) \\ & - i\Lambda([q, [p, \rho]_+] - [p, [q, \rho]_+]), \end{aligned} \quad (2.16)$$

where D_1 , D_2 , and D are the c -number diffusion coefficients, Λ is the friction constant, and κ introduces an additional term to the original system Hamiltonian $\omega a^\dagger a$. Variants of Eq. (2.16) can also be derived from generalized Weyl and Wigner transformations.²⁸ Due to the Schwartz inequality and the way Eq. (2.16) is constructed from the dissipation operators V_1 and V_2 , Eq. (2.16) is a master equation of the Lindblad form if the following conditions are satisfied:

$$D_1, D_2 > 0, \quad (2.17)$$

$$D_1 D_2 - D^2 \geq \Lambda/4. \quad (2.18)$$

Equation (2.16) can be used to describe, for instance, an electromagnetic field mode interacting with an equilibrium bath of bosons in quantum optics, or dynamics of open systems in heavy ion collisions.²⁷

The Lindblad dissipation operators can also take forms other than linear in q and p [as in Eq. (2.15)]. In fact the added β term responsible for pure dephasing in Eq. (2.7) can be derived from the semigroup formalism and by taking the system Hamiltonian ($a^\dagger a$) as the Lindblad dissipation operator. Similar applications of the semigroup formalism can be found in quantum optics (phase-damped oscillators) corresponding to a nondestructive measurement of photon number.^{29–31}

In quantum optics, atomic physics and chemical physics, master equations often need to be solved by numerical approaches which may involve continuous evolution of the density matrix as well as jumps at random instances.³² Here we adopt an analytical approach and solve both Eqs. (2.16) and (2.7) exactly by introducing the quantum characteristic function $\chi(\lambda, \lambda^*)$ ³³

$$\chi(\lambda, \lambda^*) = \text{Tr}(\rho e^{\lambda a^\dagger} e^{-\lambda^* a}), \quad (2.19)$$

where the trace is taken over the system. Instead of dealing directly with the density matrix, we derive an equation of motion for the characteristic function $\chi(\lambda, \lambda^*)$ which is then solved by method of characteristics. Details follow in Sec. III.

III. THE TRADITIONAL EQUATIONS AND THEIR SOLUTIONS

To illustrate our methodology we reproduce in this section existing solutions to Eq. (2.5). Following the master equation (2.5) one readily obtains an equation for $\chi(\lambda, \lambda^*)$

$$\begin{aligned} \frac{\partial \chi}{\partial t} + [-i\omega\lambda + \gamma(\lambda + \lambda^*)] \frac{\partial \chi}{\partial \lambda} + [i\omega\lambda^* + \gamma(\lambda + \lambda^*)] \frac{\partial \chi}{\partial \lambda^*} \\ = -\gamma \bar{n}(\lambda + \lambda^*)^2 \chi. \end{aligned} \quad (3.1)$$

Equation (3.1) was solved by the method of characteristics.³⁴ Here we give a brief derivation. Assuming the characteristic function

$$\chi(\lambda, \lambda^*) = \exp\left[\sum_{mn} C_{mn}(t) \lambda^m (-\lambda^*)^n\right], \quad (3.2)$$

where $C_{mn}(t)$ are the coefficients to be determined, one arrives at the set of differential equations for C_{mn} :

$$\dot{C}_{10} = (i\omega - \gamma)C_{10} + \gamma C_{01}, \quad (3.3)$$

$$\dot{C}_{01} = (-i\omega - \gamma)C_{01} + \gamma C_{10}, \quad (3.4)$$

$$\dot{C}_{20} = 2(i\omega - \gamma)C_{20} - \gamma(n - C_{11}), \quad (3.5)$$

$$\dot{C}_{11} = 2\gamma(n - C_{11}) + 2\gamma(C_{02} + C_{20}), \quad (3.6)$$

$$\dot{C}_{02} = -2(i\omega + \gamma)C_{02} - \gamma(n - C_{11}). \quad (3.7)$$

The analytical solutions to the above equations of the coefficients C_{mn} for arbitrary initial conditions are explicitly given below for later comparisons. The first order coefficients control the means of the density matrix Gaussians:

$$\begin{aligned} C_{10} = & \frac{A_1}{\gamma} e^{-(\gamma + \sqrt{\gamma^2 - \omega^2})t} (i\omega - \sqrt{\gamma^2 - \omega^2}) \\ & + \frac{A_2}{\gamma} e^{-(\gamma - \sqrt{\gamma^2 - \omega^2})t} (i\omega + \sqrt{\gamma^2 - \omega^2}), \end{aligned} \quad (3.8)$$

$$C_{01} = A_1 e^{-(\gamma + \sqrt{\gamma^2 - \omega^2})t} + A_2 e^{-(\gamma - \sqrt{\gamma^2 - \omega^2})t}. \quad (3.9)$$

The second order coefficients are responsible for the Gaussian widths:

$$\begin{aligned} C_{11} = & \bar{n} + \frac{e^{-2\gamma t}}{\gamma^2 - \omega^2} [-B_1 \omega^2 + B_1 \gamma^2 \cosh(2t\sqrt{\gamma^2 - \omega^2}) \\ & + 2i(B_2 - B_3) \gamma \omega \sinh^2(t\sqrt{\gamma^2 - \omega^2}) \\ & + (B_2 + B_3) \gamma \sqrt{\gamma^2 - \omega^2} \sinh(2t\sqrt{\gamma^2 - \omega^2})], \end{aligned} \quad (3.10)$$

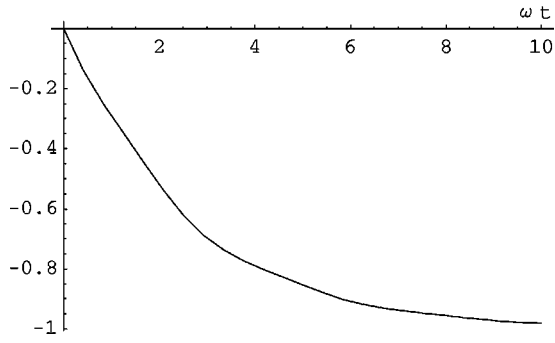


FIG. 1. $|u|^2 + v^2 - 1$ for an underdamped case: $\gamma/\omega = 0.2$. The initial state is a coherent state.

$$C_{02} = \frac{e^{-2\gamma t}}{2(\gamma^2 - \omega^2)} [\cosh(2t\sqrt{\gamma^2 - \omega^2})(-iB_1\gamma\omega + B_2\gamma^2 + B_3\gamma^2 - 2\omega^2B_3) + \sqrt{\gamma^2 - \omega^2} \sinh(2t\sqrt{\gamma^2 - \omega^2}) \times (\gamma B_1 - 2i\omega B_3) + iB_1\gamma\omega - \gamma^2(B_2 - B_3)], \quad (3.11)$$

$$C_{20} = \frac{e^{-2\gamma t}}{2(\gamma^2 - \omega^2)} [\cosh(2t\sqrt{\gamma^2 - \omega^2})(iB_1\gamma\omega + B_2\gamma^2 + B_3\gamma^2 - 2\omega^2B_2) + \sqrt{\gamma^2 - \omega^2} \sinh(2t\sqrt{\gamma^2 - \omega^2}) \times (\gamma B_1 + 2i\omega B_2) - iB_1\gamma\omega + \gamma^2(B_2 - B_3)]. \quad (3.12)$$

Here A s and B s are the constants to be determined by the initial conditions.

If initially the primary oscillator is in a coherent state with a displacement α ,³⁵ i.e.,

$$\rho(t=0) = |\alpha\rangle\langle\alpha|, \quad (3.13)$$

then

$$B_1 = -\bar{n}, \quad B_2 = B_3 = 0. \quad (3.14)$$

This is the case of interest explored by a number of authors.³⁶ For example, substituting the initial conditions in the set of solutions (3.8)–(3.12) gives the finite temperature solution for $\chi(\lambda, \lambda^*, t)$ for an initial coherent-state density matrix first derived by Savage and Walls³⁶ (in their notations)

$$\chi(\lambda, \lambda^*, t) = \exp[\lambda(u\alpha^* - v\alpha) - \lambda^*(u^*\alpha - v\alpha^*) - \bar{n}(\lambda^2 uv + \lambda^{*2} u^* v) - \bar{n}|\lambda|^2(|u|^2 + v^2 - 1)], \quad (3.15)$$

where u and v are given by

$$u = \frac{e^{-\mu_- t} + e^{-\mu_+ t}}{2} - i\omega \frac{e^{-\mu_- t} - e^{-\mu_+ t}}{\mu_- - \mu_+}, \quad (3.16)$$

$$v = \gamma \frac{e^{-\mu_- t} - e^{-\mu_+ t}}{\mu_- - \mu_+} \quad (3.17)$$

with

$$\mu_{\pm} = \gamma \pm \sqrt{\gamma^2 - \omega^2}. \quad (3.18)$$

The initial coherent state gives zero second order moments (C_{20} , C_{02} , and C_{11}) at $t=0$. At thermal equilibrium, C_{11}

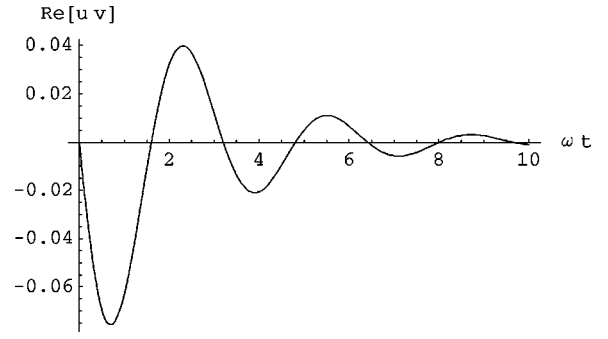


FIG. 2. $\text{Re}(uv)$ for an underdamped case: $\gamma/\omega = 0.2$. The initial state is a coherent state.

reaches \bar{n} while C_{20} and C_{02} again vanish. In Figs. 1–3, we show $|u|^2 + v^2 - 1$, and the real and imaginary parts of uv , respectively, for an underdamped case: $\gamma/\omega = 0.2$. The second order moments can be written as

$$C_{11} = -\bar{n}(|u|^2 + v^2 - 1), \quad C_{20} = C_{02}^* = -\bar{n}uv. \quad (3.19)$$

From Fig. 1, C_{11} goes monotonically from zero to \bar{n} for $\gamma/\omega = 0.2$. The real parts of C_{20} and C_{02} exhibit oscillations with a decaying amplitude, and the imaginary parts vanish at the end of each period (cf. Figs. 2 and 3). The average system energy in unit of $\hbar\omega$ can be calculated from

$$\begin{aligned} \frac{E}{\hbar\omega} &= \langle a^\dagger a \rangle = \bar{n}(1 - |u|^2 - v^2) + (u\alpha^* - v\alpha)(u^*\alpha - v\alpha^*) \\ &= \bar{n} + (|\alpha|^2 - \bar{n})(|u|^2 + v^2) - 2v\text{Re}(u^*\alpha^2). \end{aligned} \quad (3.20)$$

In Fig. 4 we display the average energy for an initial $\alpha = 0.5$ and $k_B T = \hbar\omega$. Since the initial energy $|\alpha|^2 \hbar\omega$ is less than $\bar{n}\hbar\omega \approx 0.582\hbar\omega$, the oscillator gains energy from the bath during the relaxation process.

If the initial state is a squeezed state (instead of a coherent state)

$$|\xi\rangle = S(\xi)|0\rangle, \quad (3.21)$$

where $\xi = re^{i\theta}$, and

$$S(\xi) = \exp\left(\frac{1}{2}\xi^* a^2 - \frac{1}{2}\xi a^{\dagger 2}\right), \quad (3.22)$$

the characteristic function at $t=0$ can be calculated from

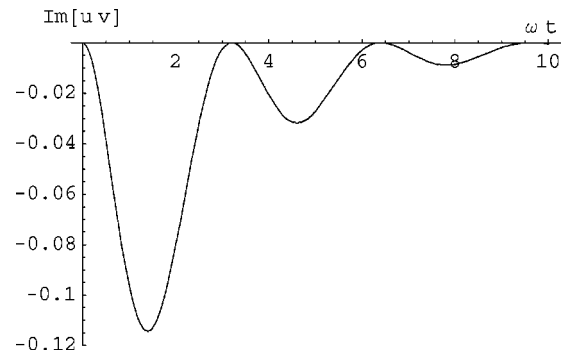


FIG. 3. $\text{Im}(uv)$ for an underdamped case: $\gamma/\omega = 0.2$. The initial state is a coherent state.

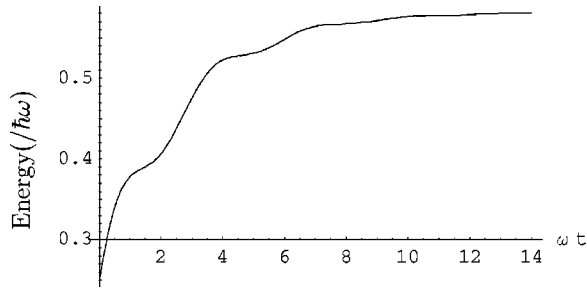


FIG. 4. The average energy in unit of $\hbar\omega$ for an underdamped case: $\gamma/\omega=0.2$, $k_B T=\hbar\omega$. The initial state is a coherent state with $\alpha=0.5$.

$$\begin{aligned} \chi(\lambda, \lambda^*, t=0) &= \text{Tr}(|\xi\rangle\langle\xi| e^{\lambda a^\dagger} e^{-\lambda^* a}) \\ &= \langle 0|S^\dagger(\xi) e^{\lambda a^\dagger} e^{-\lambda^* a} S(\xi)|0\rangle. \end{aligned} \quad (3.23)$$

Taking advantage of the fact that $S(\xi)S^\dagger(\xi)=1$, a pair of $S(\xi)$ and $S^\dagger(\xi)$ can be inserted between $e^{\lambda a^\dagger}$ and $e^{-\lambda^* a}$:

$$\chi(\lambda, \lambda^*, t=0) = \langle 0|S^\dagger(\xi) e^{\lambda a^\dagger} S(\xi) S^\dagger(\xi) e^{-\lambda^* a} S(\xi)|0\rangle. \quad (3.24)$$

The right hand side of (3.24) can be evaluated from

$$S^\dagger(\xi) e^{\lambda a^\dagger} S(\xi) = \exp(\lambda a^\dagger \cosh r - \lambda a e^{-i\theta} \sinh r), \quad (3.25)$$

$$S^\dagger(\xi) e^{-\lambda^* a} S(\xi) = \exp(-\lambda^* a \cosh r + \lambda^* a^\dagger e^{i\theta} \sinh r), \quad (3.26)$$

which follow from

$$S^\dagger(\xi) a S(\xi) = a \cosh r - a^\dagger e^{i\theta} \sinh r, \quad (3.27)$$

$$S^\dagger(\xi) a^\dagger S(\xi) = a^\dagger \cosh r - a e^{-i\theta} \sinh r. \quad (3.28)$$

Therefore one obtains

$$\begin{aligned} \chi(\lambda, \lambda^*, t=0) &= \exp(-|\lambda|^2 \sinh^2 r - \frac{1}{4} \lambda^2 e^{-i\theta} \sinh 2r \\ &\quad - \frac{1}{4} \lambda^*{}^2 e^{i\theta} \sinh 2r) \end{aligned} \quad (3.29)$$

by making use of the Baker–Hausdorff formula:

$$e^{A+B} = e^A e^B e^{-\frac{1}{2}[A,B]} \quad (3.30)$$

for any two operators A and B such that

$$[A, [A, B]] = [B, [A, B]] = 0. \quad (3.31)$$

From Eq. (3.2), the second order coefficients at $t=0$ are

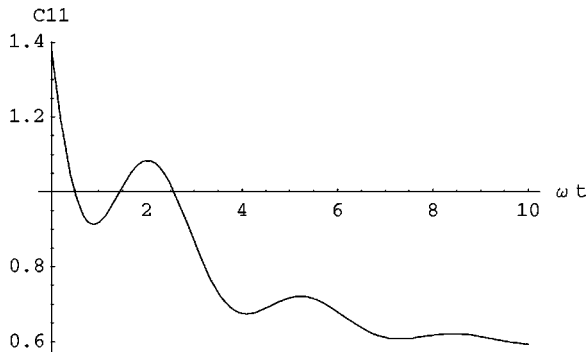


FIG. 5. C_{11} for an underdamped case: $\gamma/\omega=0.2$, $k_B T=\hbar\omega$. The initial state is a squeezed state with $\xi=1$.

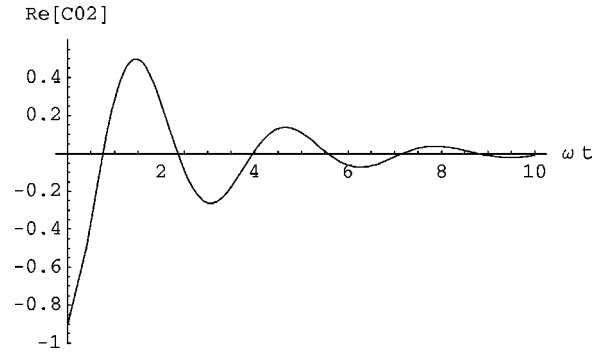


FIG. 6. $\text{Re}(C_{20})$ for an underdamped case: $\gamma/\omega=0.2$, $k_B T=\hbar\omega$. The initial state is a squeezed state with $\xi=1$.

$$C_{11}(t=0) = \bar{n} + B_1 = \sinh^2 r, \quad (3.32)$$

$$C_{20}(t=0) = B_2 = -\frac{1}{4} e^{-i\theta} \sinh 2r, \quad (3.33)$$

$$C_{02}(t=0) = B_3 = -\frac{1}{4} e^{i\theta} \sinh 2r. \quad (3.34)$$

In Fig. 5 we display the second moment $C_{11}(t)$ for $k_B T=\hbar\omega$, $\gamma/\omega=0.2$, $r=1$, $\theta=0$. For the initial squeezed state the first moments are zero all the time. Therefore the system energy $\hbar\omega\langle a^\dagger a \rangle$ can be written as $\hbar\omega C_{11}(t)$ [cf. Eq. (5.14)]. Because of the squeezing effect the initial second moment $C_{11}(0)$ equals $\sinh^2(r)$ which is then relaxed to $\bar{n} \approx 0.582$, at the thermal equilibrium, as shown in Fig. 5. Oscillations in the average system energy appear in the relaxation process although the system overall loses energy to the bath. The real and imaginary parts of the second moment C_{02} are plotted in Figs. 6 and 7, respectively. For the initial squeezed state $C_{02}(t)$ has a value of $-\sinh(2r)/4$ at $t=0$ in contrast to the case of an initial coherent state (cf. Fig. 2). The amplitudes of oscillations in $C_{02}(t)$ are more pronounced compared with a coherent-state start. The real part of $C_{02}(t)$ in fact represents the difference in variance between two canonically conjugate quadrature components [cf. Eqs. (3.37) and (3.38)]. Details will be presented later.

While the coherent state is generated by linear terms in a and a^\dagger in the exponent, the squeezed state requires quadratic terms. Squeezed states have reduced fluctuations in one quadrature component at the expense of enhanced fluctuations in the canonically conjugate quadrature component in

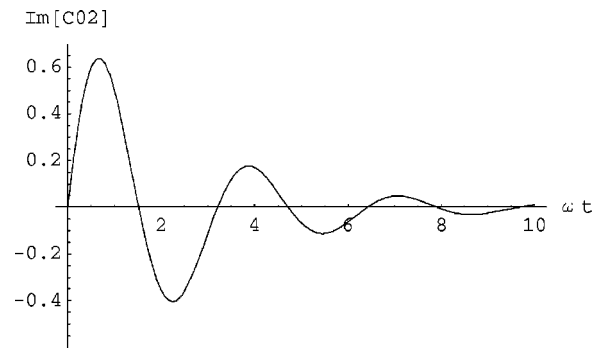


FIG. 7. $\text{Im}(C_{20})$ for an underdamped case: $\gamma/\omega=0.2$, $k_B T=\hbar\omega$. The initial state is a squeezed state with $\xi=1$.

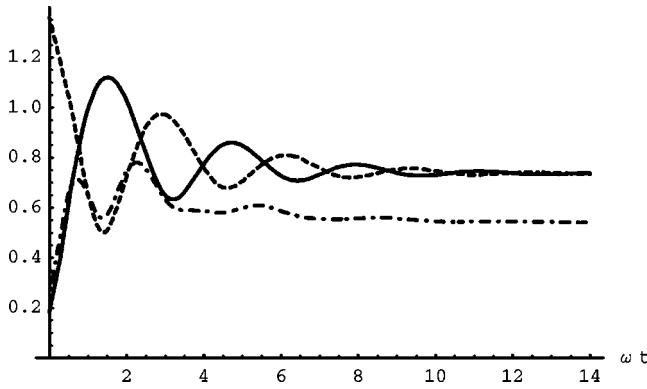


FIG. 8. $\sqrt{(\Delta Y_1)^2}$ (solid line), $\sqrt{(\Delta Y_2)^2}$ (dashed line), and their product $\sqrt{(\Delta Y_1)^2(\Delta Y_2)^2}$ (dotted-dashed line) as functions of time for an underdamped case: $\gamma/\omega=0.2$, $k_B T = \hbar\omega$. The initial state is a squeezed state with $\xi=1$.

order to comply with the uncertainty principle. To put it in more precise terms, we introduce Hermitian operators

$$Y_1 = \frac{1}{2}(a + a^\dagger), \quad Y_2 = \frac{1}{2i}(a - a^\dagger). \quad (3.35)$$

The uncertainty relation for the two operators is

$$\sqrt{(\Delta Y_1)^2(\Delta Y_2)^2} \geq \frac{1}{4}. \quad (3.36)$$

If the initial state is a squeezed state with $\xi=r$ ($\theta=0$),

$$(\Delta Y_1)^2 = \frac{1}{4} + \frac{1}{2}(C_{11} + C_{02} + C_{20}), \quad (3.37)$$

$$(\Delta Y_2)^2 = \frac{1}{4} + \frac{1}{2}(C_{11} - C_{02} - C_{20}). \quad (3.38)$$

In Fig. 8 we show $\sqrt{(\Delta Y_1)^2}$ (solid line), $\sqrt{(\Delta Y_2)^2}$ (dashed line), and their product $\sqrt{(\Delta Y_1)^2(\Delta Y_2)^2}$ (dot-dashed line) as functions of time. At $t=0$, the error ellipse is elongated along the Y_2 direction with $\sqrt{(\Delta Y_1)^2(\Delta Y_2)^2}=1/4$. At approximately $\omega t \approx 0.75$, the increasing $\sqrt{(\Delta Y_1)^2}$ catches up with the decreasing $\sqrt{(\Delta Y_2)^2}$ as the ellipse evolves. Such crossovers occur a few more times, each corresponding to a node of $\text{Re}(C_{02})$ in Fig. 6, before the error ellipse finally settles into a circle with $\sqrt{(\Delta Y_1)^2} = \sqrt{(\Delta Y_2)^2} = \sqrt{2\bar{n} + 1/2}$ at the thermal equilibrium.

The quantum characteristic function $\chi(\lambda, \lambda^*)$ is the Fourier transform of the phase space distribution function $P(z, z^*)$ of the density matrix ρ ,

$$\chi(\lambda, \lambda^*) = \int d^2z \exp(\lambda z^* - \lambda^* z) P(z, z^*). \quad (3.39)$$

The phase space distribution function $P(z, z^*)$ is also called Glauber–Sudarshan P representation of the density matrix ρ which plays the role of a quasiprobability:

$$\rho = \int d^2z P(z, z^*) |z\rangle\langle z|. \quad (3.40)$$

The corresponding equation of motion for $P(z, z^*)$ in the Schrödinger representation has the form

$$\begin{aligned} \frac{\partial P(z, z^*)}{\partial t} = & i\omega \left\{ \frac{\partial [zP(z, z^*)]}{\partial z} - \frac{\partial [z^*P(z, z^*)]}{\partial z^*} \right\} \\ & + \gamma \left\{ \frac{\partial [(z-z^*)P(z, z^*)]}{\partial z} + \text{c.c.} \right\} \\ & - \gamma \bar{n} \left(\frac{\partial}{\partial z} - \frac{\partial}{\partial z^*} \right)^2 P(z, z^*). \end{aligned} \quad (3.41)$$

Here c.c. stands for complex conjugate. In applications it is preferable to employ the master equations for the phase-space distribution functions which correspond to density matrix in the interaction picture:

$$\tilde{\rho} = \int d^2z \tilde{P}(z, z^*) |z\rangle\langle z|, \quad (3.42)$$

where

$$\tilde{\rho} = e^{i\omega a^\dagger a t} \rho e^{-i\omega a^\dagger a t}. \quad (3.43)$$

In the interaction representation and under a RWA which neglects aa and $a^\dagger a^\dagger$ terms, the equation of motion for $\tilde{P}(z, z^*)$ takes the form

$$\begin{aligned} \frac{1}{\gamma} \frac{\partial \tilde{P}(z, z^*)}{\partial t} = & 2\bar{n} \frac{\partial^2 \tilde{P}(z, z^*)}{\partial z \partial z^*} + \frac{\partial [z\tilde{P}(z, z^*)]}{\partial z} + \frac{\partial [z^*\tilde{P}(z, z^*)]}{\partial z^*}. \end{aligned} \quad (3.44)$$

We shall make use of Eq. (3.44) later in Sec. V.

IV. THE LINDBLAD-TYPE EQUATIONS

The generality of the Lindblad-type master equation, Eq. (2.16), allows descriptions of various physical processes if a proper combination of parameters Λ , κ , D_1 , D_2 and D is chosen. For example, Eq. (2.16) can be used to model the correlated-emission laser that uses atoms prepared in a coherent superposition of the states between which the laser emission takes place.³⁷ The density matrix for the field mode a (a is the annihilation operator) follows

$$\begin{aligned} \dot{\rho} = & \Lambda_1(a^\dagger \rho a - \rho a a^\dagger) + \Lambda_2(a \rho a^\dagger \rho a - a^\dagger a \rho) \\ & + \Lambda_3(\rho a^{\dagger 2} - a^\dagger \rho a^\dagger) + \Lambda_4(a^{\dagger 2} \rho - a^\dagger \rho a^\dagger) \\ & + f[a^\dagger, \rho] + \text{H.c.}, \end{aligned} \quad (4.1)$$

where the c -number parameters Λ_i can be easily expressed in terms of Λ , κ , D_1 , D_2 and D . Another example is a single mode of electromagnetic field coupled with a squeezed bath³⁸ for which the density matrix master equation takes the form

$$\begin{aligned} \dot{\rho} = & \gamma(N+1)(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a) + \gamma N(2a^\dagger \rho a \\ & - a a^\dagger \rho - \rho a a^\dagger) - \gamma M(2a^\dagger \rho a^\dagger - a^\dagger a^\dagger - \rho a^\dagger a^\dagger) \\ & - \gamma M^*(2a\rho a - a\rho a - \rho a a). \end{aligned} \quad (4.2)$$

Again the master equation is a special case of Eq. (2.16).

In this section we solve Eq. (2.16) borrowing the technique of the previous section. The equation for the characteristic function follows from Eq. (2.16):

$$\begin{aligned} \frac{\partial \chi}{\partial t} = & -i\omega \left(\lambda^* \frac{\partial}{\partial \lambda^*} - \lambda \frac{\partial}{\partial \lambda} \right) \chi + 4\Lambda \lambda^* \lambda \chi \\ & - 4\Lambda \left(\lambda^* \frac{\partial}{\partial \lambda^*} + \lambda \frac{\partial}{\partial \lambda} \right) \chi + 2\kappa(\lambda^*{}^2 + \lambda^2) \chi \\ & - 4\kappa \left(\lambda \frac{\partial}{\partial \lambda^*} + \lambda^* \frac{\partial}{\partial \lambda} \right) \chi + D_2(\lambda^* - \lambda)^2 \chi \\ & - D_1(\lambda + \lambda^*)^2 \chi + 2iD(\lambda^*{}^2 - \lambda^2) \chi \end{aligned} \quad (4.3)$$

from which one then arrives at the set of differential equations for C_{mn} :

$$\dot{C}_{10} = (i\omega - 4\Lambda)C_{10} + 4\kappa C_{01}, \quad (4.4)$$

$$\dot{C}_{01} = (-i\omega - 4\Lambda)C_{01} + 4\kappa C_{10}, \quad (4.5)$$

$$\dot{C}_{20} = 2(i\omega - 4\Lambda)C_{20} + 4\kappa C_{11} + 2\kappa + D_2 - D_1 - 2iD, \quad (4.6)$$

$$\dot{C}_{11} = -8\Lambda C_{11} + 8\kappa(C_{02} + C_{20}) - 4\Lambda + 2D_2 + 2D_1, \quad (4.7)$$

$$\dot{C}_{02} = -2(i\omega + 4\Lambda)C_{02} + 4\kappa C_{11} + 2\kappa + D_2 - D_1 + 2iD. \quad (4.8)$$

The solutions to the above equations for arbitrary initial conditions are given below. The first order coefficients are

$$C_{10} = e^{-4\Lambda t} [A_1 \cosh(\Omega t) + \Omega^{-1}(i\omega A_1 + 4\kappa A_2) \sinh(\Omega t)], \quad (4.9)$$

$$C_{01} = e^{-4\Lambda t} [A_2 \cosh(\Omega t) + \Omega^{-1}(4\kappa A_1 - i\omega A_2) \sinh(\Omega t)], \quad (4.10)$$

where A_1 and A_2 are determined by initial conditions, and

$$\Omega = \sqrt{16\kappa^2 - \omega^2}. \quad (4.11)$$

The second order coefficients are

$$\begin{aligned} C_{11} = & \frac{16\Lambda [D_1(\Lambda - \kappa) + (D_2 - 2\Lambda + 2\kappa)(\Lambda + \kappa)] + 8D\kappa\omega + (D_1 + D_2 - 2\Lambda)\omega^2}{4\Lambda(16\Lambda^2 - \Omega^2)} + 4(B_2 + B_3)\kappa\Omega^{-1}e^{-8\Lambda t} \sinh(2\Omega t) \\ & + \Omega^{-2}e^{-8\Lambda t} [-B_1\omega^2 - 4i\kappa\omega(B_2 - B_3) + (16\kappa^2 B_1 + 4i\kappa\omega B_2 - 4i\kappa\omega B_3) \cosh(2\Omega t)], \end{aligned} \quad (4.12)$$

$$\begin{aligned} C_{02} = & \frac{[D_1(\Lambda - \kappa) - D_2(\Lambda + \kappa)](4\Lambda - i\omega) - 2iD(4\Lambda^2 - 4\kappa^2 - i\Lambda\omega)}{2\Lambda(16\Lambda^2 - \Omega^2)} - \frac{1}{\Lambda\Omega} e^{-8\Lambda t} [\cosh(2\Omega t)(-2i\kappa\omega B_1 + 8\kappa^2 B_2 \\ & + 8\kappa^2 B_3 - \omega^2 B_3) + \Omega \sinh(2\Omega t)(2\kappa B_1 - i\omega B_3) + 2\kappa(i\omega B_1 - 4\kappa B_2 + 4\kappa B_3)], \end{aligned} \quad (4.13)$$

$$\begin{aligned} C_{20} = & \frac{[D_1(\Lambda - \kappa) - D_2(\Lambda + \kappa)](4\Lambda + i\omega) - 2iD(4\kappa^2 - 4\Lambda^2 - i\Lambda\omega)}{2\Lambda(16\Lambda^2 - \Omega^2)} - \frac{1}{\Lambda\Omega} e^{-8\Lambda t} [\cosh(2\Omega t)(2i\kappa\omega B_1 + 8\kappa^2 B_2 + 8\kappa^2 B_3 \\ & - \omega^2 B_2) + \Omega \sinh(\Omega t)(2\kappa B_1 + i\omega B_2) + 2\kappa(-i\omega B_1 + 4\kappa B_2 - 4\kappa B_3)]. \end{aligned} \quad (4.14)$$

Here B_s are the constants to be determined by initial conditions. The long time asymptotic value for the sum of three second order coefficients has a simple form:

$$(C_{20} + C_{11} + C_{02})|_{t=\infty} = \frac{32\Lambda(\Lambda - \kappa)(D_1 - \Lambda - \kappa) - 8D\omega(\Lambda - \kappa) + (D_1 + D_2 - 2\Lambda)\omega^2}{4\Lambda(16\Lambda^2 - \Omega^2)}. \quad (4.15)$$

One can readily show that C_{20} , C_{02} , and C_{11} in the previous section can be recovered by setting

$$\Lambda = \kappa = \frac{\gamma}{4}, \quad D_1 = \gamma \left(\bar{n} + \frac{1}{2} \right), \quad D_2 = D = 0. \quad (4.16)$$

By setting $D_2=0$, however, complete positivity is no longer preserved, and Eq. (2.5) is known to exhibit pathological behavior of the density matrix. It is obvious from the first two (linear) equations for C_{mn} that translational invariance is retained only if $\kappa=\Lambda$. This will be discussed in detail in Sec. VIII.

Our general solutions for the second moments Eqs. (4.12)–(4.14) can accommodate various forms of initial density matrices. If the initial state is a squeezed state, $|\xi\rangle$

$=S(\xi)|0\rangle$ with $\xi=re^{i\theta}$, the first moments are zero for all times. The operator average $\langle a^{\dagger 2} \rangle + \langle a^2 \rangle$ which equals to $C_{02} + C_{20}$ has a simple time dependence

$$\begin{aligned} C_{02} + C_{20} = & -\frac{\Omega}{\Lambda} e^{-8\Lambda t} (B_2 + B_3) \cosh(2\Omega t) \\ & - \frac{4\kappa}{\Lambda} e^{-8\Lambda t} B_1 \sinh(2\Omega t) + \text{const.} \end{aligned} \quad (4.17)$$

The constant term above is in fact the asymptotic value of $C_{02} + C_{20}$ at long times:

$$(C_{02} + C_{20})|_{t=\infty} = \frac{D_1(\Lambda - \kappa) - D_2(\Lambda + \kappa) + 2D\omega}{16\Lambda^2 - 16\kappa^2 + \omega^2}. \quad (4.18)$$

This term vanishes when the Lindblad-type equation (2.16) is reduced to Agarwal's equation giving the final thermalized state an error circle ($\sqrt{(\Delta Y_1)^2} = \sqrt{(\Delta Y_2)^2}$). However, it is not generally true for the Lindblad-type master equation (2.16). From Eqs. (3.37) and (3.38),

$$(\Delta Y_1)^2 - (\Delta Y_2)^2 = C_{02} + C_{20} \quad (4.19)$$

the solution of Eq. (2.16) will start from a squeezed state with $\xi=r$ and $\theta=0$, and ends up in a state with an error ellipse with $(\Delta Y_1)^2 - (\Delta Y_2)^2$ given by Eq. (4.18). Furthermore the asymptotic value of $\sqrt{(\Delta Y_1)^2}$ at long times can be calculated from Eqs. (3.37) and (4.15).

The semigroup formalism does not guarantee approach of thermal equilibrium without extra constraints. This can be easily demonstrated as follows. Under a RWA which neglects aa and $a^\dagger a^\dagger$ terms, the D term in Eq. (2.16)

$$-D([q,[p,\rho]] + [p,[q,\rho]]) \quad (4.20)$$

vanishes. Provided that

$$D_1 = \gamma \left(\bar{n} + \frac{1}{2} \right), \quad \Lambda + \kappa = \frac{\gamma}{2}, \quad \Lambda - \kappa = D_2, \quad (4.21)$$

the Lindblad-type equation under RWA takes the form

$$\begin{aligned} \dot{\rho} = & -i\omega[a^\dagger a, \rho] + [\gamma(\bar{n}+1) + 2D_2](2apa^\dagger - a^\dagger a\rho - \rho a^\dagger a) \\ & + \gamma\bar{n}(2a^\dagger \rho a - aa^\dagger \rho - \rho aa^\dagger). \end{aligned} \quad (4.22)$$

We note that the above master equation has an explicit Lindblad form

$$\dot{\rho} + i\omega[a^\dagger a, \rho] = L'_D \rho \equiv \sum_m (2V_m \rho V_m^\dagger - V_m^\dagger V_m \rho - \rho V_m^\dagger V_m), \quad (4.23)$$

where we have defined the Liouvillian operator L'_D , and the dissipation operators V_m are such that $[V_m, V_m^\dagger]$ are c -number constants. The two Lindblad dissipative operators V_m ($m=1, 2$) here are proportional to a and a^\dagger , respectively, and L'_D and L_D differ only by a constant c number. Complete positivity is therefore guaranteed for Eq. (4.22). Assuming Λ greater than κ , or equivalently, $D_2 > 0$, the downward transition rates (from number states $n+1$ to n) are increased by an amount $2D_2$ in Eq. (4.22). On the other hand, if we assume that

$$D_1 = \gamma \left(\bar{n} + \frac{1}{2} \right), \quad \Lambda + \kappa = \frac{\gamma}{2}, \quad \kappa - \Lambda = D_2 > 0, \quad (4.24)$$

i.e., κ is greater than Λ while keeping D_2 positive, the equation of motion under RWA will be changed to

$$\begin{aligned} \dot{\rho} = & -i\omega[a^\dagger a, \rho] + \gamma(\bar{n}+1)(2apa^\dagger - a^\dagger a\rho - \rho a^\dagger a) \\ & + [\gamma\bar{n} + 2D_2](2a^\dagger \rho a - aa^\dagger \rho - \rho aa^\dagger). \end{aligned} \quad (4.25)$$

Thus the upward transition rates (from number states n to $n+1$) are increased by an amount $2D_2$. This shows that the Lindblad-type equation does not provide approach to thermal equilibrium because detailed balance is easily violated with the added D_2 term in both cases.

Detailed balance, however, can be imposed onto Eq. (2.16) in the forms of parameter constraints to ensure final

approach to equilibrium in the framework of semigroup theories. Since the final thermalized state has an error circle, setting Eq. (4.18) equal to zero puts a necessary condition on the c -number parameters in Eq. (2.16). Efforts along this direction for semigroup master equations have been a matter of much recent interest.^{2,4,30} For instance, Gao has proposed a master equation constructed from one single Lindblad dissipation operator which is linear in both a and a^\dagger with the proper proportionality coefficients to sustain detailed balance.⁴ The positivity requirement in Gao's approach is marginally satisfied with $D_1 D_2 = \Lambda^2/4$. Gao's construction of L_D via a single Lindblad dissipation operator has recently found support from a first-principle derivation of master equations for collision-driven dissipative evolution.²

V. ADDITION OF PURE DEPHASING

In the previous section the two Lindblad operators responsible for positivity-preserving dissipation are linear in a^\dagger and a . If the Lindblad dissipation operator is chosen as the system Hamiltonian $a^\dagger a$, dissipation is added to the system dynamics in forms of pure dephasing. The resulting master equation with the pure-dephasing β term, Eq. (2.7), which has recently been proposed,²² can be solved exactly up to second moments by similar means as in the previous sections. Compared with Eq. (3.1), the β term in Eq. (2.7) adds

$$-\beta \left(\lambda \frac{\partial}{\partial \lambda} - \lambda^* \frac{\partial}{\partial \lambda^*} \right)^2 \chi(\lambda, \lambda^*) \quad (5.1)$$

to the equation of motion for the characteristic function $\chi(\lambda, \lambda^*)$:

$$\begin{aligned} \frac{\partial \chi}{\partial t} + [-i\omega\lambda + \gamma(\lambda + \lambda^*)] \frac{\partial \chi}{\partial \lambda} + [i\omega\lambda^* + \gamma(\lambda + \lambda^*)] \frac{\partial \chi}{\partial \lambda^*} \\ = -\gamma\bar{n}(\lambda + \lambda^*)^2 \chi - \beta \left(\lambda \frac{\partial}{\partial \lambda} - \lambda^* \frac{\partial}{\partial \lambda^*} \right)^2 \chi. \end{aligned} \quad (5.2)$$

We proceed to derive the differential equations for C_{mn} . The first order equations differ little from those in Sec. III except for an added β term:

$$\dot{C}_{10} = (i\omega - \gamma)C_{10} + \gamma C_{01} - \beta C_{10}, \quad (5.3)$$

$$\dot{C}_{01} = (-i\omega - \gamma)C_{01} + \gamma C_{10} - \beta C_{01}. \quad (5.4)$$

This β term, however, breaks the translation invariance as will be addressed later. The solution to the above equations is straightforward:

$$\begin{aligned} C_{10} = e^{-(\beta+\gamma)t} \left[A_1 \cosh(t\sqrt{\gamma^2 - \omega^2}) \right. \\ \left. + \frac{i\omega A_1 + \gamma A_2}{\sqrt{\gamma^2 - \omega^2}} \sinh(t\sqrt{\gamma^2 - \omega^2}) \right], \end{aligned} \quad (5.5)$$

$$C_{01} = e^{-(\beta + \gamma)t} \left[A_2 \cosh(t\sqrt{\gamma^2 - \omega^2}) + \frac{\gamma A_1 - i\omega A_2}{\sqrt{\gamma^2 - \omega^2}} \sinh(t\sqrt{\gamma^2 - \omega^2}) \right], \quad (5.6)$$

where A_1 and A_2 are determined by initial conditions.

The second order equations appear more complicated than the first order ones:

$$\dot{C}_{20} = 2(i\omega - \gamma)C_{20} - \gamma(n - C_{11}) - \beta(4C_{20} + C_{10}^2), \quad (5.7)$$

$$\dot{C}_{11} = 2\gamma(n - C_{11}) + 2\gamma(C_{02} + C_{20}) + 2\beta C_{01}C_{10}, \quad (5.8)$$

$$\dot{C}_{02} = -2(i\omega + \gamma)C_{02} - \gamma(n - C_{11}) - \beta(4C_{02} + C_{01}^2). \quad (5.9)$$

It proves fruitful to relate C_{mn} to the following operator averages:

$$\langle a \rangle = C_{01}, \quad (5.10)$$

$$\langle a^\dagger \rangle = C_{10}, \quad (5.11)$$

$$\langle a^2 \rangle = 2C_{02} + C_{01}^2, \quad (5.12)$$

$$\langle a^{\dagger 2} \rangle = 2C_{20} + C_{10}^2, \quad (5.13)$$

$$\langle a^\dagger a \rangle = C_{11} + C_{10}C_{01}. \quad (5.14)$$

Therefore one may derive equations of motion for those operator averages from the second order equations:

$$\frac{d}{dt} \langle a^\dagger a \rangle = 2\gamma(\bar{n} - \langle a^\dagger a \rangle) + \gamma(\langle a^{\dagger 2} \rangle + \langle a^2 \rangle), \quad (5.15)$$

$$\frac{1}{2} \frac{d}{dt} \langle a^2 \rangle = \gamma(\langle a^\dagger a \rangle - \bar{n}) - (i\omega + r + 2\beta) \langle a^2 \rangle, \quad (5.16)$$

$$\frac{1}{2} \frac{d}{dt} \langle a^{\dagger 2} \rangle = \gamma(\langle a^\dagger a \rangle - \bar{n} + (i\omega - r - 2\beta) \langle a^{\dagger 2} \rangle). \quad (5.17)$$

Since the β term introduces pure dephasing, β does not enter Eq. (5.15) which describes the evolution of the average system energy. Below we give the solutions for $\langle a^2 \rangle$, $\langle a^{\dagger 2} \rangle$, and $\langle a^\dagger a \rangle$.

The set of equations for $\langle a^2 \rangle$, $\langle a^{\dagger 2} \rangle$, and $\langle a^\dagger a \rangle$, which incorporate two types of dissipation mechanisms quantified by the two parameters β and γ , can be recast in a matrix form:

$$\frac{d}{dt} \begin{pmatrix} \langle a^\dagger a \rangle - \bar{n} \\ \langle a^2 \rangle \\ \langle a^{\dagger 2} \rangle \end{pmatrix} = \begin{pmatrix} -2\gamma & \gamma & \gamma \\ 2\gamma & -2i\omega - 2\gamma - 4\beta & 0 \\ 2\gamma & 0 & 2i\omega - 2\gamma - 4\beta \end{pmatrix} \times \begin{pmatrix} \langle a^\dagger a \rangle - \bar{n} \\ \langle a^2 \rangle \\ \langle a^{\dagger 2} \rangle \end{pmatrix}. \quad (5.18)$$

Therefore, one merely needs to diagonalize the 3 by 3 matrix, and the solutions to the differential equations are then straightforward. We display the three eigenvalues, which are solutions of a third-order equation

$$x^3 + 2(3\gamma + 4\beta)x^2 + 4(\omega^2 + 2\gamma^2 + 8\beta\gamma + 4\beta^2)x + 8\gamma(\omega^2 + 2\beta\gamma + 4\beta^2) = 0, \quad (5.19)$$

labeled as x_i ($i=1,2,3$):

$$x_1 = -\frac{2}{3}(4\beta + 3\gamma) + \frac{2^{4/3}}{3c}(4\beta^2 + 3\gamma^2 - 3\omega^2) + \frac{2^{2/3}c}{3}, \quad (5.20)$$

$$x_2 = -\frac{2}{3}(4\beta + 3\gamma) - \frac{(i\sqrt{3} + 1)2^{4/3}}{3c}(4\beta^2 + 3\gamma^2 - 3\omega^2) + \frac{(i\sqrt{3} - 1)2^{2/3}c}{6}, \quad (5.21)$$

$$x_3 = -\frac{2}{3}(4\beta + 3\gamma) + \frac{(i\sqrt{3} + 1)2^{4/3}}{3c}(4\beta^2 + 3\gamma^2 - 3\omega^2) - \frac{(i\sqrt{3} - 1)2^{2/3}c}{6}, \quad (5.22)$$

where

$$c = [16\beta^3 - 12\beta^2\gamma - 15\gamma^3 - 18\gamma\omega^2 + 3(2\beta + \gamma)(5\gamma^2 + 6\omega^2) + 2\sqrt{-(4\beta^2 + 3\gamma^2 - 3\omega^2)^3 + \beta^2(8\beta^2 + 9\gamma^2 + 18\omega^2)^2}]^{1/3}. \quad (5.23)$$

The complete solutions for Eq. (5.18) can be conveniently expressed by the three roots x_i ($i=1,2,3$) in a compact form. Below we give the detailed forms of $\langle a^\dagger a \rangle$, $\langle a^2 \rangle$, and $\langle a^{\dagger 2} \rangle$ as a function of t :

$$\begin{aligned} \langle a^\dagger a \rangle &= \bar{n} + C_1 \sum_i \exp(x_i t) g(x_i) [x_i^2 + 4(\gamma + 2\beta)x_i \\ &\quad + 4(\gamma^2 + \omega^2 + 4\beta\gamma + 4\beta^2)] \\ &\quad + \gamma C_2 \sum_i \exp(x_i t) g(x_i) [x_i + 2(\gamma - i\omega + 4\beta)] \\ &\quad + \gamma C_3 \sum_i \exp(x_i t) g(x_i) [x_i + 2(\gamma + i\omega + 4\beta)], \end{aligned} \quad (5.24)$$

$$\begin{aligned} \langle a^2 \rangle &= 2\gamma^2 C_3 \sum_i \exp(x_i t) g(x_i) + 2\gamma C_1 \sum_i \exp(x_i t) g(x_i) \\ &\quad \times [x_i + 2(\gamma - i\omega + 4\beta)] + C_2 \sum_i \exp(x_i t) g(x_i) \\ &\quad \times [x_i^2 + 4(\gamma + 2\beta - 2i\omega)x_i + 2\gamma^2 + 8\beta\gamma - 4i\gamma\omega], \end{aligned} \quad (5.25)$$

$$\begin{aligned} \langle a^{\dagger 2} \rangle &= 2\gamma^2 C_2 \sum_i \exp(x_i t) g(x_i) + 2\gamma C_1 \sum_i \exp(x_i t) g(x_i) \\ &\times [x_i + 2(\gamma + i\omega + 4\beta)] + C_3 \sum_i \exp(x_i t) g(x_i) \\ &\times [x_i^2 + 4(\gamma + 2\beta + 2i\omega)x_i + 2\gamma^2 + 8\beta\gamma + 4i\gamma\omega], \end{aligned} \quad (5.26)$$

where C_1 , C_2 , and C_3 are constants determined by initial conditions, and $g(x)$ is an auxiliary function defined by

$$g(x) = [x^2 + 4(3\gamma + 4\beta)x + 4(\omega^2 + 2\gamma^2 + 8\beta\gamma + 4\beta^2)]^{-1}. \quad (5.27)$$

VI. LEVEL POPULATIONS

The solutions we have so far presented for the three quantum master equations, Eqs. (2.5), (2.16), and (2.7), contain an enormous amount of information of the underlying quantum dissipative systems, which can be subjects for many further studies. To illustrate this point we only need to look at the simplest among the three—the solutions in Sec. III. Even this set of solutions proves to be too complex for many purposes here. We therefore further limit ourselves to the case of the underdamped systems which is characterized by $\gamma \ll \omega$. We assume that the oscillator is initially in a coherent state with a displacement α ,³⁵ i.e.,

$$\rho(t=0) = |\alpha\rangle\langle\alpha|. \quad (6.1)$$

The parameter u in Sec. III reduces to $e^{-\gamma t + i\omega t}$, and v to 0. The diagonal elements of the density matrix ρ in the coherent state representation are related to the quantum characteristic function $\chi(\lambda, \lambda^*)$ by³³

$$\langle z|\rho|z\rangle = \frac{1}{\pi} \int d^2\lambda \chi(\lambda, \lambda^*) \exp(-|\lambda|^2 - \lambda z^* + \lambda^* z). \quad (6.2)$$

Because the density matrix ρ can be expanded directly in terms of its diagonal coherent state matrix elements³⁹

$$\rho = \frac{1}{\pi^2} \int \int d^2z d^2w \frac{|z+w\rangle\langle z-w|}{\langle z-w|z+w\rangle} e^{-|w|^2} \langle z|\rho|z\rangle. \quad (6.3)$$

Therefore the probability of finding n bosons can be calculated from

$$\begin{aligned} p_n(t) &\equiv \langle n|\rho|n\rangle \\ &= \frac{1}{\pi^2} \int \int d^2z d^2w \langle z|\rho|z\rangle (z+w)^n (z-w)^{*n} \\ &\times \exp[-|w|^2 - (z^* - w^*)(z+w)]. \end{aligned} \quad (6.4)$$

Making use of the identity

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dx dy e^{-a(x^2+y^2)+x(b+c)+iy(b-c)} = \frac{\pi}{a} \exp\left(\frac{bc}{a}\right), \quad (6.5)$$

we obtain from Eq. (6.2) after the simple integration over λ

$$\langle z|\rho|z\rangle = \delta \exp[-\delta|z - \alpha e^{-i\omega t - \gamma t}|^2], \quad (6.6)$$

where

$$\delta = [1 + \bar{n}(1 - e^{-2\gamma t})]^{-1}. \quad (6.7)$$

Carrying out the quadruple integral over the complex variables z and w , one arrives at (cf. Appendix B)

$$p_0(t) \equiv \langle 0|\rho|0\rangle = \delta \exp(-\delta|\alpha|^2 e^{-2\gamma t}). \quad (6.8)$$

We note that at $t=0$, p_0 equals $e^{-|\alpha|^2}$ following the Poisson distribution for the number states, and at $t=\infty$, $p_0 = (1 + \bar{n})^{-1}$ in the thermalized state.

Similarly, one can obtain (cf. Appendix B)

$$\begin{aligned} p_1(t) &\equiv \langle 1|\rho|1\rangle = [\delta|\alpha|^2 e^{-2\gamma t} + \bar{n}(1 - e^{-2\gamma t})] \delta^2 \\ &\times \exp(-\delta|\alpha|^2 e^{-2\gamma t}). \end{aligned} \quad (6.9)$$

Again at $t=0$, $p_1 = |\alpha|^2 e^{-|\alpha|^2}$ obeys the Poisson distribution. At $t=\infty$, $p_1 = \bar{n}/(1 + \bar{n})^2 = p_0 e^{-\hbar\omega/k_B T}$ reflecting thermalization.

For higher n , straightforward evaluations of $p_n(t)$ becomes cumbersome. However, one may derive a recurrence relation for $p_n(t)$. Following Eq. (3.40), the diagonal density matrix elements $\langle n|\rho|n\rangle$ are related to the phase space distribution function $P(z, z^*)$ by

$$\langle n|\rho|n\rangle = \int d^2z P(z, z^*) \frac{|z|^{2n}}{n!} e^{-|z|^2}. \quad (6.10)$$

Therefore it follows from Eq. (3.44) that

$$\begin{aligned} \dot{p}_n(t) &= 2\gamma \bar{n} n p_{n-1}(t) + 2\gamma(1 + \bar{n})(n+1)p_{n+1}(t) \\ &\quad - 2\gamma[\bar{n}(2n+1) + n]p_n(t). \end{aligned} \quad (6.11)$$

Equation (6.11) allows one to recursively obtain all $p_n(t)$ from $p_0(t)$, $p_1(t)$. For example, $p_2(t)$ is obtained from $p_0(t)$, $p_1(t)$ and $dp_1(t)/dt$ as follows. First, a straightforward calculation gives

$$\begin{aligned} \dot{p}_1(t) &= \delta^3 \exp(-\delta|\alpha|^2 e^{-2\gamma t}) [\delta^2 |\alpha|^4 e^{-4\gamma t} (1 + \bar{n}) \\ &\quad + \delta|\alpha|^2 e^{-2\gamma t} (\bar{n}^2 - 1 - \bar{n}(\bar{n} + 3)) e^{-2\gamma t} \\ &\quad + \bar{n} e^{-2\gamma t} - \bar{n}^2 e^{-2\gamma t} (1 - e^{-2\gamma t})]. \end{aligned} \quad (6.12)$$

Then from Eqs. (6.8), (6.9), and (6.12), it follows that

$$\begin{aligned} p_2(t) &= \frac{\dot{p}_1(t) - \bar{n}p_0(t) + (3\bar{n} + 1)p_1(t)}{2(1 + \bar{n})} \\ &= p_0(t) [\delta(\delta^{-1} - 1)^{-1}]^2 L_2[x(t)], \end{aligned} \quad (6.13)$$

where $L_n[x(t)]$ are the Laguerre polynomials, and

$$x(t) = -\delta(\delta^{-1} - 1)^{-1} |\alpha|^2 e^{-2\gamma t}. \quad (6.14)$$

For arbitrary n , taking into account the recurrence relations of the Laguerre polynomials:

$$xL_n'(x) = nL_n(x) - nL_{n-1}(x), \quad (6.15)$$

$$(n+1)L_{n+1}(x) = (2n+1-x)L_n(x) - nL_{n-1}(x). \quad (6.16)$$

It can be easily shown (cf. Appendix C) that

$$p_n(t) = p_0(t) [\delta(\delta^{-1} - 1)^{-1}]^n L_n[x(t)] \quad (6.17)$$

satisfy Eq. (6.11). We would like to point out that the Boltzmann distribution is ensured at long times by the fact that

$$\lim_{t \rightarrow \infty} L_n[x(t)] = 1, \quad (6.18)$$

and, therefore,

$$\lim_{t \rightarrow \infty} \frac{p_n(t)}{p_{n-1}(t)} = e^{-\hbar\omega/k_B T}. \quad (6.19)$$

In particular, when p_n/p_{n-1} is independent of n , a population temperature which does not depend on n may be defined by

$$T_p(t) \equiv - \left[\frac{k_B}{\hbar\omega} \ln \left(\frac{p_n}{p_{n-1}} \right) \right]^{-1}. \quad (6.20)$$

A Boltzmann-type distribution is necessary in order to define T_p . The Taylor series of the Laguerre polynomials are given by

$$L_n(x) = \sum_{s=0}^n (-1)^{n-s} \frac{n! x^{n-s}}{(n-s)!(n-s)!s!}. \quad (6.21)$$

One can examine the near-equilibrium properties of the vibrational manifold from the asymptotic behavior of the Laguerre polynomials for small x :

$$L_n(x) = 1 - nx + O(x^2). \quad (6.22)$$

At a certain time scale t_B , the absolute value of $x(t)$ is sufficiently small, i.e.,

$$|x(t)| = \frac{|\alpha|^2 e^{-2\gamma t}}{\bar{n}(1 - e^{-2\gamma t})[1 + \bar{n}(1 - e^{-2\gamma t})]} \ll 1, \quad (6.23)$$

the population temperature of the vibrational manifold which establishes a Boltzmann-type distribution follows

$$T_p(t) = \left[\frac{1}{T} - \frac{k_B |x(t)|}{\hbar\omega} \right]^{-1}. \quad (6.24)$$

The time scale t_B to satisfy Eq. (6.23) can be much smaller than the time scale $1/2\gamma$ at which the system reaches full thermal equilibrium with the dissipative bath. This happens, for example, when the bath temperature is sufficiently high such that

$$\left(\frac{\hbar\omega}{k_B T} \right)^2 |\alpha|^2 (1 - e^{-2\gamma t_B})^{-2} e^{-2\gamma t_B} \sim 1. \quad (6.25)$$

If $|\alpha|$ is of order 1, one has

$$t_B \sim \left(\frac{\hbar\omega}{k_B T} \right) \frac{1}{2\gamma} \ll \frac{1}{2\gamma}. \quad (6.26)$$

For the same temperatures, t_B is reached earlier for smaller initial displacements $|\alpha|$. This means that it takes longer for the thermal field to wipe out the initial Poisson distribution for oscillators with larger initial displacements. In general, the vibrational manifold is able to achieve a Boltzmann-type distribution at a time scale

$$t_B \sim |\alpha| \left(\frac{\hbar\omega}{k_B T} \right) \frac{1}{2\gamma} \quad (6.27)$$

before it comes into full thermal equilibrium with the bath if the temperature of the bath satisfies

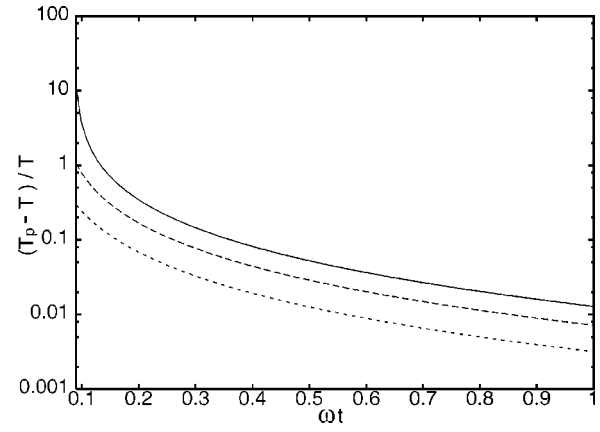


FIG. 9. The relative difference between the population temperature T_p and the bath temperature T : $(T_p - T)/T$, $\gamma = \omega/2$. Initial displacements $\alpha = 0.4$ (solid line), $\alpha = 0.3$ (dashed line), and $\alpha = 0.2$ (dotted line). $k_B T = 10\hbar\omega$.

$$T \gg \frac{\hbar\omega|\alpha|}{k_B}. \quad (6.28)$$

Figure 9 illustrates the process of approaching thermal equilibrium for an underdamped oscillator ($\gamma = \omega/2$) with initial displacements $\alpha = 0.4$ (solid line), $\alpha = 0.3$ (dashed line), and $\alpha = 0.2$ (dotted line). The bath temperature is high compared with the oscillator frequency ($k_B T = 10\hbar\omega$) so that $\bar{n} \approx k_B T / \hbar\omega = 10$. Plotted in log scale in Fig. 9 is the relative difference between the population temperature T_p and the bath temperature T : $(T_p - T)/T$. The oscillator reaches full thermal equilibrium with the bath at a time scale of $(2\gamma)^{-1} = \omega^{-1}$. However, a Boltzmann-type distribution is formed with a population temperature well defined at a time scale of $0.1\omega^{-1}$ which is ten times smaller than ω^{-1} . The larger the initial displacement, the slower such a distribution comes in place, and the higher the population temperature when compared at the same t (cf. Fig. 9).

The concept of the population temperature here bears close resemblance to a spectral temperature defined in the context of the Kennard–Stepanov relation.^{40,41} This spectral temperature approaches the ambient temperature (the bath temperature) upon thermal equilibration of the emitting electronic manifold. Experimentally such a spectral temperature is usually well defined from the steady-state absorption and fluorescence spectra, but its deviations from the ambient temperature in many cases remain quite a puzzle.⁴⁰ The time dependence of the spectral temperature was recently studied in the framework of the Brownian oscillator model.⁴¹

To conclude, we have studied the evolution of level populations for an oscillator initially in a coherent state with displacement α in the underdamped regime. We found that the vibrational manifold establishes a Boltzmann-type distribution characterized by a population temperature $T_p(t)$ before it reaches full thermal equilibrium if the bath temperature is sufficiently high.

VII. ZERO TEMPERATURE COHERENT STATES

In this section we look into the zero-temperature evolution of a coherent state $|\alpha\rangle$ defined by $a|\alpha\rangle = \alpha|\alpha\rangle$.³⁵ The

coherent state, regarded as a quantum mechanical state which approaches a classical state, is labeled by the complex number α . The real part of α represents the average position of the oscillator, and the imaginary part, the average momentum. We confine ourselves to zero temperature here, and seek to provide connections between the master equations and the zero-temperature evolution of a coherent state.

At zero temperature, the form of a coherent state is preserved in the time evolution.^{36,42} The time evolution of the coordinate and momentum expectation values can be evaluated in a functional integral approach from¹⁵

$$\langle q \rangle_t = \langle q \rangle_0 \dot{G}_+(t) + \frac{\langle p \rangle_0}{m} G_+(t), \quad (7.1)$$

$$\langle p \rangle_t = m \langle q \rangle_0 \ddot{G}_+(t) + \langle p \rangle_0 \dot{G}_+(t), \quad (7.2)$$

where $\langle q \rangle_0$ and $\langle p \rangle_0$ are the expectation values at $t=0$, $G_+(t)$ is the inverse Laplace transform of

$$\bar{G}_+(s) = \frac{1}{s^2 + 2s\gamma + \omega^2} \quad (7.3)$$

for frequency-independent friction. $G_+(t)$ therefore has the form

$$G_+(t) = \frac{e^{-\gamma t} (e^{t\sqrt{\gamma^2 - \omega^2}} - e^{-t\sqrt{\gamma^2 - \omega^2}})}{2\sqrt{\gamma^2 - \omega^2}}. \quad (7.4)$$

The complex time-dependent displacement parameter can then be calculated from $G_+(t)$ as follows:

$$\alpha(t) = \sqrt{\frac{m\omega}{2\hbar}} \langle q \rangle_t + i \sqrt{\frac{1}{2\hbar m\omega}} \langle p \rangle_t. \quad (7.5)$$

Since the coherent state is related to the number state $|n\rangle$ via

$$|\alpha\rangle = e^{-1/2|\alpha|^2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle, \quad (7.6)$$

one can obtain the probability $p_n(t)$ of residing in the number state $|n\rangle$

$$p_n(t) = e^{-|\alpha(t)|^2} \frac{|\alpha(t)|^{2n}}{n!}. \quad (7.7)$$

From $p_n(t)$, one may try to derive relaxation rates between vibrational levels. First we assume that only nearest levels exchange populations

$$\dot{p}_n = -p_n \kappa_{n-1,n} + p_{n-1} \kappa_{n,n-1} + p_{n+1} \kappa_{n,n+1} - p_n \kappa_{n+1,n}, \quad n \neq 0; \quad (7.8)$$

$$\dot{p}_0 = p_1 \kappa_{0,1} - p_0 \kappa_{1,0}, \quad (7.9)$$

where $\kappa_{m,n}$ denotes the rate from level n to level m . Next we require that detailed balance apply

$$\kappa_{n,m} = e^{\beta(m-n)\omega} \kappa_{m,n}. \quad (7.10)$$

At zero temperature, this means that all upward rates are zero. One may start from $\kappa_{0,1}$, for zero temperature,

$$\kappa_{0,1} = \frac{\dot{p}_0(t)}{p_1 - p_0 e^{-\beta\omega}} = -|\alpha|^{-2} \frac{d}{dt} |\alpha|^2, \quad (7.11)$$

where for ohmic dissipation, the magnitude of the complex displacement parameter $\alpha(t)$ decreases as follows:

$$\frac{d}{dt} |\alpha|^2 = -\frac{2\gamma m}{\hbar\omega} \left[\langle q \rangle_0 \ddot{G}_+(t) + \frac{\langle p \rangle_0}{m} \dot{G}_+(t) \right]^2. \quad (7.12)$$

For the underdamped oscillator ($\gamma \ll \omega$) and an initial real displacement $\alpha(t=0)$, one finds from Eq. (7.5),

$$|\alpha(t)|^2 = \frac{m\omega}{2\hbar} e^{-2\gamma t}. \quad (7.13)$$

Therefore from Eq. (7.11), $\kappa_{0,1} = 2\gamma$, which agrees with the zero temperature form of Eq. (6.11):

$$\dot{p}_n(t) = 2\gamma(n+1)p_{n+1}(t) - 2\gamma n p_n(t). \quad (7.14)$$

For finite temperatures, however, coherent states are no longer preserved.^{36,42} For moderate to strong damping, non-nearest neighbor transitions in the vibrational manifolds may also become important.

It is interesting to compare Eq. (7.7) to results from solutions of the Agarwal master equations. For the underdamped Brownian oscillator, agreements can be found between Eq. (6.17) at zero temperature and Eq. (7.7) for $\gamma \ll \omega$. For example, if the initial average momentum $\langle p \rangle_0 = 0$, i.e., $\alpha(t=0)$ is real, from Eq. (7.5), one obtains for $\ll \omega$,

$$\alpha(t) = \sqrt{\frac{m\omega}{2\hbar}} e^{-\gamma t} e^{-i\omega t}. \quad (7.15)$$

This agrees with Eq. (6.17) at $T=0$. Similarly a conclusion can be drawn when $\langle q \rangle_0 = 0$. However, the same is not true for the overdamped oscillator.

For the overdamped oscillator, $\gamma \gg \omega$, one has

$$u = \frac{1}{2} [e^{-(\omega^2/2\gamma)t} + e^{-2\gamma t}], \quad (7.16)$$

$$v = \frac{1}{2} [e^{-(\omega^2/2\gamma)t} - e^{-2\gamma t}]. \quad (7.17)$$

The diagonal elements of the reduced density matrix ρ in the coherent state representation takes the form

$$\langle z|\rho|z\rangle = \frac{1}{\sqrt{rs}} \exp\left\{-\frac{1}{r} [\text{Im}(\alpha) e^{-2\gamma t} + \text{Im}(z)]^2\right\} \times \exp\left\{-\frac{1}{s} [\text{Re}(\alpha) e^{-(\omega^2/2\gamma)t} - \text{Re}(z)]^2\right\}, \quad (7.18)$$

where $r = 1 + \bar{n}(1 - e^{-4\gamma t})$, $s = 1 + \bar{n}[1 - e^{-(\omega^2/\gamma)t}]$. At zero temperature, one finds for real initial displacements $\alpha(t=0) = \text{Re}(\alpha) = \alpha'$ ($\langle p \rangle_0 = 0$)

$$p_1(t) = \alpha'^2 e^{-(\omega^2/\gamma)t} \exp[-\alpha'^2 e^{-(\omega^2/\gamma)t}]; \quad (7.19)$$

and for purely imaginary initial displacements $\alpha(t=0) = i \text{Im}(\alpha) = i\alpha''$ ($\langle q \rangle_0 = 0$)

$$p_1(t) = \alpha''^2 e^{-4\gamma t} \exp(-\alpha''^2 e^{-4\gamma t}). \quad (7.20)$$

However, the results from Eq. (7.7) for $\gamma \gg \omega$ and short times generate only one time scale 4γ regardless of whether the initial α is real or purely imaginary. This points to the restricted validity of the Agarwal master equation, which was derived under the Born approximation. The solutions of the

Agarwal master equations have been used by several authors for the overdamped case.³⁶ Caution needs to be taken when results from the Agarwal master equation are extended to overdamped regimes.

VIII. DISCUSSIONS

Two generalized quantum Markovian master equations, one with the Lindblad dissipation operators linear in the position and momentum operators, and the other with an added pure-dephasing term, have been solved exactly to second moments, and the solutions are now available for many applications to come. To our knowledge those explicit forms of the exact solutions have appeared for the first time.

We study a generalized Markovian master equation constructed with two sets of Lindblad dissipation operators.^{17,27} With the aid of quantum characteristic functions, we have obtained an explicit form of exact solutions to the Lindblad-type master equation (2.16). Results in the form of the quantum characteristic function $\chi(\lambda, \lambda^*)$ can be used for calculations of various dynamic quantities of interest. For example, the coordinate-space distribution function has a mean related to the first order coefficients $C_{01} + C_{10}$, and a variance determined by the second order coefficients C_{20} , C_{11} , and C_{02} . As a demonstration level populations are derived in the underdamped regime from the characteristic functions $\chi(\lambda, \lambda^*)$ in Sec. VI. It is found that the vibrational manifold can reach a Boltzmann-type distribution characterized by a population temperature $T_p(t)$ before it comes into full thermal equilibrium with the surrounding bath if the bath temperature is sufficiently high.

Equation (2.16) is designed to preserve positivity at the sacrifice of other desirable traits of the master equations. For example, if $\kappa \neq \Lambda$, the time derivative of the position operator expectation value differs from the momentum operator expectation value, and therefore, translational invariance is in general violated. This is readily shown by adding Eqs. (4.4) and (4.5) together:

$$\frac{d}{dt}\langle a + a^\dagger \rangle = -i\omega\langle a - a^\dagger \rangle - 4(\Lambda - \kappa)\langle a + a^\dagger \rangle. \quad (8.1)$$

If $\kappa \neq \Lambda$, Eq. (8.1) violates the Ehrenfest theorem which expresses a formal connection between the time dependence of expectation values of canonically conjugate variables and the Hamiltonian equations of classical mechanics. Applications of Eq. (2.16) are therefore restricted only to systems other than extended ones.

If the Lindblad dissipation operator is chosen as the system Hamiltonian $a^\dagger a$, dissipation is added to the system dynamics in forms of pure dephasing. Applications to interacting many-body systems have been implemented in the framework of TDHF.²³ Exact solutions of the first moments as well as second moments $\langle a^2 \rangle$, $\langle a^\dagger a \rangle$, and $\langle a^{\dagger 2} \rangle$ shall facilitate studies of dissipation dynamics with pure-dephasing. Equation (2.7) again breaks translational invariance for arbitrary β as is evident from adding the two first order equations (5.3) and (5.4):

$$\frac{d}{dt}\langle a + a^\dagger \rangle = -i\omega\langle a - a^\dagger \rangle - \beta\langle a + a^\dagger \rangle. \quad (8.2)$$

For nonzero β , the damping of the average oscillator displacement $\langle a + a^\dagger \rangle$ depends on $\langle a + a^\dagger \rangle$ itself, and thus the translational invariance is broken.

A master equation for the n th level population $p_n(t)$ similar to Eq. (6.11) appeared in the literature⁴³⁻⁴⁵ based on a phenomenological approach including a radiative decay term and a driving term to represent picosecond excitations:

$$\dot{p}_n(t) + \alpha_n p_n(t) = \sum_l [\gamma_{nl} p_l(t) - \gamma_{ln} p_n(t)] + k_n(t), \quad (8.3)$$

where α_n is the intrinsic (fluorescence plus nonradiative) decay rate of the n th number state, and $k_n(t)$ is the time dependent excitation pulse which drives the n th level. The rates γ_{nl} are given by the detailed balance

$$\gamma_{nl} = \kappa(n+1)\delta_{l,n+1} + \kappa e^{-\hbar\omega/k_B T_n} \delta_{l+1,n}, \quad (8.4)$$

where κ is the rate constant which characterizes the system-bath interactions. Equation (8.3) without the driving term $k_n(t)$ but containing a phenomenological decay coefficient α_n which is linear in the energy has been solved by Seshadri and Kenkre.⁴⁶ It is found that for initial Boltzmann distributions at temperatures different from that of the bath the excited state will maintain the Boltzmann distribution with a time-dependent temperature $T_p(t)$ which is defined in a similar way as the population temperature in Eq. (6.20). However, $T_p(t)$ in their theory does not asymptotically go to the bath temperature.

There are some recent developments utilizing the Markovian, Lindblad-type master equation that deserve comments. Burghardt modeled the effect of an environment on ultrafast predissociation in terms of the semigroup formalism in the coordinate representation.⁹ Frequency components in the Liouvillian spectra were found to directly correspond to those observed in pump-probe spectra in the impulsive limit. Of particular interest is the numerical comparison between the Lindblad and the Redfield forms of the dissipative master equation in which very similar results were found for the two, and slightly negative values showed up in the diagonal elements of the density matrix evolving according to the Redfield form. Yan *et al.* reformulated the Redfield theory at the operator level in an effort to bridge the theory with a class of Fokker-Planck equations.²² The new form of the Redfield equation is of the Lindblad type if operators pertaining to dissipative modes are Hermitian (such as the β term with $a^\dagger a$ as the dissipation operator), although positivity is not preserved in general.

ACKNOWLEDGMENT

The authors thank Y. J. Yan for enlightening discussions.

APPENDIX A: THE BROWNIAN OSCILLATOR MODEL

In the Brownian oscillator model, the system is taken to be a two electronic-level system with some primary nuclear coordinates coupled linearly to the electronic systems:

$$\hat{H} = |g\rangle\hat{H}_g\langle g| + |e\rangle\hat{H}_e\langle e| + \hat{H}', \quad (A1)$$

where

$$\hat{H}_g = \sum_j \left[\frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 q_j^2 \right], \quad (\text{A2})$$

$$\hat{H}_e = h\omega_{eg}^0 + \sum_j \left[\frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 (q_j + d_j)^2 \right], \quad (\text{A3})$$

$$\hat{H}' = \sum_n \left[\frac{p_n^2}{2m_n} + \frac{m_n \omega_n^2}{2} \left(x_n - \sum_j \frac{c_{nj} q_j}{m_n \omega_n^2} \right)^2 \right]. \quad (\text{A4})$$

Here p_j (p_n), q_j (x_n), and m_j (m_n) represent the momentum, the coordinate, and the mass of the j th (n th) nuclear mode of the primary (bath) oscillators, respectively. d_j is the displacement for the j th nuclear mode. H' describes the bath oscillators and their coupling to the primary oscillators with a coupling strength c_{nj} .

APPENDIX B: DERIVATION OF POPULATIONS FOR THE ZEROth AND THE FIRST NUMBER STATES

The derivation of $p_0(t)$ is straightforward. From Eq. (6.4),

$$p_0(t) = \frac{\delta}{\pi^2} \int \int d^2 z d^2 w \exp(-|z|^2 + w^* z - z^* w - \delta |z - \alpha e^{-\gamma t} e^{-i\omega t}|^2). \quad (\text{B1})$$

The integral over w produces two δ functions ($\delta(x)$ and $\delta(y)$, $z = x + iy$). Therefore one immediately arrives at Eq. (6.8).

Now we turn to $p_1(t)$. From Eq. (6.4),

$$p_1(t) = \frac{\delta}{\pi^2} \int \int d^2 z d^2 w \exp(-|z|^2 + w^* z - z^* w - \delta |z - \alpha e^{-\gamma t} e^{-i\omega t}|^2) (|z|^2 - |w|^2 + z^* w - z w^*). \quad (\text{B2})$$

The first term with $|z|^2$ vanishes. The rest of the three terms contain derivatives with respect to x and y . Making use of the identities

$$\int_{-\infty}^{\infty} e^{i2xv} dv = \pi \delta(x), \quad (\text{B3})$$

$$\int_{-\infty}^{\infty} v e^{i2xv} dv = \frac{\pi}{2} \delta'(x), \quad (\text{B4})$$

$$\int_{-\infty}^{\infty} v^2 e^{i2xv} dv = \frac{\pi}{4} \delta''(x), \quad (\text{B5})$$

one obtains Eq. (6.9). Taking derivative of Eq. (6.9) with respect to t , one finds

$$\begin{aligned} \frac{\partial p_1(t)}{\partial t} = & \delta^5 \exp(-\delta |\alpha|^2 e^{-2\gamma t}) \{ |\alpha|^4 e^{-4\gamma t} (1 + \bar{n}) \\ & + |\alpha|^2 e^{-2\gamma t} [(1 - e^{-2\gamma t})^2 \bar{n}^3 + (1 - e^{-2\gamma t}) \\ & \times (1 - 3e^{-2\gamma t}) \bar{n}^2 - (1 + 2e^{-2\gamma t}) \bar{n} - 1] \\ & + \bar{n} e^{-2\gamma t} \delta^{-2} [\bar{n} (1 - e^{-2\gamma t}) - 1] \}. \end{aligned} \quad (\text{B6})$$

APPENDIX C: DERIVATION OF POPULATIONS FOR HIGHER NUMBER STATES

In this Appendix we show that $p_n(t)$ of Eq. (6.17) satisfies the master equation (6.11). From Eq. (6.17), a straightforward calculation yields

$$\begin{aligned} \frac{\partial p_n(t)}{\partial t} = & \bar{n} e^{-2\gamma t} \delta^4 [\delta (\delta^{-1} - 1)^{-1}]^{n-2} \\ & \times \exp(-\delta |\alpha|^2 e^{-2\gamma t}) \{ \delta |\alpha|^2 [1 + \bar{n} (1 - e^{-4\gamma t})] \\ & \times L_{n-1}^1 [x(t)] + \bar{n} L_n [x(t)] (1 - e^{-2\gamma t}) \\ & \times [(1 + \bar{n}) \delta |\alpha|^2 (1 - e^{-2\gamma t}) + n - \bar{n} (1 - e^{-2\gamma t})] \}, \end{aligned} \quad (\text{C1})$$

where $L_{n-1}^k [x(t)]$ are the associated Laguerre polynomials defined by⁴⁷

$$L_n^k(x) = (-1)^k \frac{d^k}{dx^k} [L_{n+k}(x)]. \quad (\text{C2})$$

In general, for $k > -1$,

$$L_n^k(x) = \sum_0^n (-1)^m \frac{(n+k)!}{(n-m)!(k+m)!m!} x^m. \quad (\text{C3})$$

Making use of Eq. (6.15), one quickly finds that Eq. (6.11) is obeyed.

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