

On the conundrum of deriving exact solutions from approximate master equations

Roland Doll, David Zueco, Martijn Wubs, Sigmund Kohler ^{*}, Peter Hänggi

Institut für Physik, Universität Augsburg, Universitätsstraße 1, D-86135 Augsburg, Germany

1. Introduction

The coherent evolution of small quantum systems is typically influenced by its interaction with environmental degrees of freedom, which results in quantum dissipation and decoherence. These ubiquitous phenomena play a crucial role in various fields of physics and chemistry, such as quantum optics, electron transfer reactions [1,2], the electron transport through molecular wires [3,4], and in particular in quantum information processing, where we recently witnessed significant experimental progress [5–7]. The optimisation of the coherence properties of quantum devices certainly requires a good theoretical understanding of the processes that induce decoherence.

The environment of a quantum system is frequently modelled as an ensemble of harmonic oscillators that couple to the system [8–11]. If the coupling is linear in the oscil-

lator position, one can eliminate the environment to obtain a closed equation for the dissipative quantum system. Such equations are in general not easy to deal with and, accordingly, only a few exact solutions exist in dissipative quantum mechanics, e.g. for the dissipative harmonic oscillator [12–15] and its parametrically driven version [16]. Recently, an exact solution has been found also for the dissipative Landau–Zener problem at zero temperature [17]. A whole class of system–bath models that can be solved exactly are those in which the system Hamiltonian and the system–bath coupling commute [18–27]. Herein we focus on such so-called pure phase noise models.

Even though pure phase noise allows an exact solution of the reduced quantum dynamics, it is sometimes convenient to employ a perturbative master equation approach, such as the Bloch–Redfield equation [28]. On the one hand, those approaches may provide direct access to the dephasing rates avoiding tedious algebra and in particular in the limit of weak system–bath coupling, they are expected to give quantitatively good results. On the other hand, it is

^{*} Corresponding author.

E-mail address: sigmund.kohler@physik.uni-augsburg.de (S. Kohler).

possible to test their quality when an exact solution is available. We here show that for phase noise, the results of approximate master equations can even be exact, despite the fact that they are based on second-order perturbation theory.

After introducing our system–bath model in Section 2, we present in Sections 3 and 4 a master equation for weak system–bath coupling and the exact time-evolution of the reduced system density operator, respectively. In Section 5, we argue why the exact solution complies with the master equation.

2. System–bath model

We model the dissipative quantum system by coupling the central system to a quantum heat bath that consists of harmonic oscillators, so that the total system–bath Hamiltonian reads [10,11]

$$H = H_s + H_{sb} + H_b \quad (1)$$

and $H_0 = H_s + H_b$ describes the system and the bath in the absence of the coupling. While we will not specify the system Hamiltonian, we employ for the bath an ensemble of independent harmonic oscillators with the Hamiltonian

$$H_b = \sum_k \hbar\omega_k b_k^\dagger b_k. \quad (2)$$

Here, b_k^\dagger and b_k are the usual creation and annihilation operators which obey the commutation relation $[b_k, b_{k'}^\dagger] = \delta_{kk'}$. We assume that the system couples linearly to the bath with a hermitian system operator X , so that the interaction Hamiltonian reads

$$H_{sb} = \hbar X \xi \quad (3)$$

with the effective bath coordinate

$$\xi = \sum_k (g_k b_k + g_k^* b_k^\dagger). \quad (4)$$

If the coupling operator X commutes with the system Hamiltonian, $[X, H_s] = 0$, the coupling (3) does not induce transitions between system eigenstates and, thus, constitutes pure phase noise. Henceforth we shall focus on this case.

We choose an initial condition of the Feynman–Vernon type, i.e. one for which the total density operator R at time $t = 0$ can be factorised into a system and a bath contribution ρ and ρ_b , respectively, i.e.

$$R(0) = \rho(0)\rho_b(0). \quad (5)$$

The bath itself is frequently assumed to be initially at thermal equilibrium. However, if the initial expectation value of the coupling operator X does not vanish, the coupling (3) entails a force on the bath oscillators. Then the natural initial state $\rho_b = \rho_b(0)$ of the bath is rather a displaced thermal state which falls in the class of non-squeezed Gaussian states. A convenient basis for these states is provided by the coherent states $\{|\beta_k\rangle\}$ defined by the eigenvalue equation

$b_k |\beta_k\rangle = \beta_k |\beta_k\rangle$. Owing to the overcompleteness of this basis, any hermitian operator can be written in a diagonal form, which assigns to each operator a P -function [29,30]. In particular, the bath density operator can be written as

$$\rho_b = \int |\beta_1, \beta_2, \dots\rangle \langle \beta_1, \beta_2, \dots| \prod_k P_k(\beta_k, \beta_k^*) d^2\beta_k, \quad (6)$$

where $d^2\beta_k$ denotes integration over the complex plane. Henceforth, we assume that the P -function of each oscillator k is a Gauss function, such that

$$P_k(\beta_k, \beta_k^*) = \frac{1}{\pi n_k} \exp\left(\frac{-(\beta_k - \bar{\beta}_k)(\beta_k - \bar{\beta}_k)^*}{n_k}\right). \quad (7)$$

As a central property of a Gaussian state, all expectation values are fully determined by $\bar{\beta}_k = \langle b_k \rangle_b$ and $n_k = \langle b_k^\dagger b_k \rangle_b - |\langle b_k \rangle_b|^2$, where

$$\langle \dots \rangle_b = \text{tr}_b(\rho_b \dots) \quad (8)$$

denotes the expectation value with respect to the bath state ρ_b . An important particular case is the canonical ensemble of the bath at temperature T such that $\rho_b \propto \exp(-H_b/k_B T)$, which corresponds to $\langle b_k \rangle_b = 0$ and $2n_k = \coth(\hbar\omega_k/2k_B T) - 1$.

The dynamics of the system plus the bath is governed by the Liouville–von Neumann equation

$$\dot{\tilde{R}}(t) = \tilde{\mathcal{L}}\tilde{R}(t) \quad (9)$$

with the Liouvillian $\tilde{\mathcal{L}}(\dots) = -i[\tilde{H}_{sb}(t), \dots]/\hbar$. The tilde denotes the interaction-picture representation with respect to H_0 , i.e. $\tilde{A}(t) = U_0^\dagger(t) A U_0(t)$, with the free propagator $U_0(t) = \exp(-iH_0 t/\hbar)$. The interaction-picture representation of the effective bath coordinate ξ is easily obtained from $\tilde{b}_k(t) = b_k \exp(-i\omega_k t)$, while for pure phase noise, $\tilde{X}(t) = X$, owing to the relation $[X, H_s] = 0$. For the same reason, H_s and X possess a complete set of common eigenstates $\{|n\rangle\}$, for which the respective eigenvalues are denoted by E_n and X_n .

We are exclusively interested in the state of the system, so our goal is to find the reduced density operator $\tilde{\rho}(t) = \text{tr}_b \tilde{R}(t)$. In the subsequent sections, we derive explicit expressions for the reduced dynamics.

3. Master equation approach

A common and successful approach to dissipative quantum dynamics is provided by master equations, i.e. differential or integro-differential equations of motion for the reduced density operator [30–32]. There exist various formally exact quantum master equations in time-convolutionless [16,33–42] and time-non-local form [43–45] which, however, generally cannot be solved explicitly and, thus, one often has to resort to a perturbative treatment.

Here, we employ a time-convolutionless quantum master equation of the form

$$\dot{\tilde{\rho}}(t) = \mathcal{K}(t)\tilde{\rho}(t), \quad (10)$$

with a time-dependent superoperator $\mathcal{K}(t)$. Note that there arises no inhomogeneity since we are assuming a factorising initial state of the form (5), which leads to a linear equation of motion [46]. Eq. (10) is formally exact and possesses an apparently simple form, but it generally cannot be solved analytically. Thus, it is convenient to expand the generator $\mathcal{K}(t)$ in powers of the interaction, i.e. $\mathcal{K}(t) = \sum_{\ell} \mathcal{K}_{\ell}(t)$. By a direct calculation [47,48] or by using a time-convolutionless projection operator technique [39,49,50], it is possible to obtain an expression for the ℓ th order generator \mathcal{K}_{ℓ} . In doing so, we assign to a superoperator \mathcal{G} of the total system a reduced superoperator $\langle \mathcal{G} \rangle$ defined by its action on a system operator Y , that is $\langle \mathcal{G} \rangle Y = \text{tr}_{\text{b}}\{\mathcal{G}(Y\rho_{\text{b}})\}$. With this notation, the time-convolutionless generators read $\mathcal{K}_1(t) = \langle \mathcal{L}(t) \rangle$, and

$$\mathcal{K}_{\ell}(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{\ell-2}} dt_{\ell-1} \times \langle \langle \widetilde{\mathcal{L}}(t) \widetilde{\mathcal{L}}(t_1) \dots \widetilde{\mathcal{L}}(t_{\ell-1}) \rangle \rangle_{\text{oc}}, \quad (11)$$

for $\ell = 2, 3, \dots$. The symbol $\langle \langle \dots \rangle \rangle_{\text{oc}}$ denotes an ordered cumulant [31,48], i.e. a sum of certain products of reduced superoperators of the form $\langle \widetilde{\mathcal{L}}(t) \widetilde{\mathcal{L}}(t_1) \dots \rangle$.

The fact that the Liouvillians at different times generally do not commute makes it practically impossible to write down an explicit expression for the ℓ th cumulant for large ℓ . However, for weak system–bath coupling it is possible to neglect higher than second-order terms in the expansion of the generator, i.e. we may approximate $\mathcal{K}(t) \approx \mathcal{K}_1(t) + \mathcal{K}_2(t)$. Fortunately the second time-ordered cumulant takes the simple form $\langle \langle \widetilde{\mathcal{L}}(t) \widetilde{\mathcal{L}}(t_1) \rangle \rangle_{\text{oc}} = \langle \widetilde{\mathcal{L}}(t) \widetilde{\mathcal{L}}(t_1) \rangle - \langle \widetilde{\mathcal{L}}(t) \rangle \langle \widetilde{\mathcal{L}}(t_1) \rangle$. Considering now explicitly the interaction Hamiltonian (3), we obtain the ‘‘standard’’ time-local weak-coupling equation

$$\dot{\tilde{\rho}}(t) = -i\langle \tilde{\xi}(t) \rangle_{\text{b}} [X, \tilde{\rho}(t)] - \int_0^t d\tau (\mathcal{S}(t, t - \tau) [X, [X, \tilde{\rho}(t)]] + i\mathcal{A}(t, t - \tau) [X, \{X, \tilde{\rho}(t)\}]), \quad (12)$$

where $\{A, B\} = AB + BA$ denotes the anti-commutator, and where we have defined the symmetric and anti-symmetric correlation functions

$$\mathcal{S}(t, t') = \frac{1}{2} \langle \{ \Delta \tilde{\xi}(t), \Delta \tilde{\xi}(t') \} \rangle_{\text{b}}, \quad (13)$$

$$\mathcal{A}(t, t') = \frac{1}{2} \langle [\Delta \tilde{\xi}(t), \Delta \tilde{\xi}(t')] \rangle_{\text{b}} \quad (14)$$

of the operator-valued fluctuation $\Delta \tilde{\xi}(t) = \tilde{\xi}(t) - \langle \tilde{\xi}(t) \rangle_{\text{b}}$. Note that for a Gaussian initial state ρ_{b} of the bath, the mean value $\langle \tilde{\xi}(t) \rangle_{\text{b}}$ of the bath coordinate in general does not vanish. Thus, it explicitly appears in the master equation (12).

It is convenient to expand the master equation (12) into the eigenbasis of the system–bath interaction. We then obtain for a matrix element $\langle m | \tilde{\rho} | n \rangle = \tilde{\rho}_{mn}$ the differential equation

$$\dot{\tilde{\rho}}_{mn}(t) = \left[-i(X_m - X_n) \langle \tilde{\xi}(t) \rangle_{\text{b}} - (X_m - X_n)^2 \int_0^t d\tau \mathcal{S}(t, t - \tau) - i(X_m^2 - X_n^2) \int_0^t d\tau \mathcal{A}(t, t - \tau) \right] \tilde{\rho}_{mn}(t). \quad (15)$$

For the diagonal matrix elements $\tilde{\rho}_{nn}$ the right-hand side of Eq. (15) vanishes, i.e. the populations remain constant in time as one expects for a pure dephasing model. This implies that generally the system will not reach thermal equilibrium. Although no energy is exchanged and, thus, the interaction is dissipationless, the relative phases between eigenstates will be randomised. As a consequence, off-diagonal elements of the reduced density matrix may decay, which reflects the process of decoherence.

Using the coherent state representation (7) for a Gaussian bath state, we can evaluate the mean value of the bath coordinate $\tilde{\xi}$ and the correlation functions in an explicit form and find

$$\langle \tilde{\xi}(t) \rangle_{\text{b}} = \sum_k (g_k \bar{\beta}_k e^{-i\omega_k t} + g_k^* \bar{\beta}_k^* e^{i\omega_k t}), \quad (16)$$

$$\mathcal{S}(t, t - \tau) = \sum_k |g_k|^2 \cos(\omega_k \tau) (1 + 2n_k), \quad (17)$$

$$\mathcal{A}(t, t - \tau) = - \sum_k |g_k|^2 \sin(\omega_k \tau). \quad (18)$$

Note that the correlation functions $\mathcal{S}(t, t')$ and $\mathcal{A}(t, t')$ depend on the time difference $t - t'$ only, with $\mathcal{A}(t, t')$ independent of the initial state of the bath. If the correlation functions vanish sufficiently fast, it is possible to employ a Markov approximation, i.e. to extend the upper integration limits in Eq. (15) to infinity. Then, the master equation (15) reduces to the standard Bloch–Redfield equation [28] and, moreover, is of Lindblad form, so that the complete positivity of the reduced density operator is conserved even for arbitrarily short times [51,52].

4. Exact solution

The dynamics of a system subject to pure phase noise can, at least in principle, be solved analytically [18–27]. The formal solution of the Liouville–von Neumann equation (9) after tracing over the bath’s degrees of freedom reads

$$\tilde{\rho}(t) = \text{tr}_{\text{b}}\{U(t)R(0)U^\dagger(t)\} \quad (19)$$

with the propagator

$$U(t) = \mathcal{T} \exp \left\{ \frac{1}{i\hbar} \int_0^t d\tau \tilde{H}_{\text{sb}}(\tau) \right\} \quad (20)$$

and the time-ordering operator \mathcal{T} . Although the consideration of the time-ordering can often be quite cumbersome, it nevertheless can be accomplished for the model discussed here. We deferred the explicit derivation to [Appendix A](#),

where we find for the elements of the reduced density matrix the exact expression

$$\begin{aligned} \tilde{\rho}_{mn}(t) &= \tilde{\rho}_{mn}(0) e^{i(X_m^2 - X_n^2)\phi(t) - (X_m - X_n)^2 \sum_k |z_k(t)|^2/2} \\ &\times \prod_k \chi_k(z_k(t)[X_m - X_n], z_k^*(t)[X_m - X_n]), \end{aligned} \quad (21)$$

with the time-dependent phase

$$\phi(t) = \sum_k \frac{|g_k|^2}{\omega_k^2} [\omega_k t - \sin(\omega_k t)], \quad (22)$$

the quantum characteristic function [30]

$$\chi_k(\lambda_k, \lambda_k^*) = \int e^{\lambda_k \beta_k^* - \lambda_k^* \beta_k} P_k(\beta_k, \beta_k^*) d^2 \beta_k \quad (23)$$

and the time-dependent complex number $z_k(t) = g_k^* [1 - \exp(i\omega_k t)] / \omega_k$. To arrive at Eq. (21), we only used the assumption that initially the system and the bath are uncorrelated and that the initial bath state ρ_b factorises with respect to the modes k [see Eqs. (5) and (6)]. For the case of a Gaussian distribution of the bath modes, it is possible to calculate the integral in the characteristic function (23) explicitly. We finally find the exact time-evolution of the reduced matrix element

$$\begin{aligned} \tilde{\rho}_{mn}(t) &= \rho_{mn}(0) \exp\{-(X_m - X_n)^2 A(t) \\ &+ i[(X_m^2 - X_n^2)\phi(t) + (X_m - X_n)\varphi(t)]\} \end{aligned} \quad (24)$$

with the phases $\phi(t)$ defined in Eq. (22) and

$$\varphi(t) = 2 \sum_k \text{Im} \left(\frac{\tilde{\beta}_k^* g_k^*}{\omega_k} [1 - e^{i\omega_k t}] \right). \quad (25)$$

The time-dependent damping amplitude $A(t)$ does not depend on the mean values $\tilde{\beta}_k$ of the bath modes and reads

$$A(t) = \sum_k |g_k|^2 \frac{1 - \cos(\omega_k t)}{\omega_k^2} (1 + 2n_k). \quad (26)$$

Note that a similar result was obtained recently for a bath initially in a squeezed thermal state [53].

Upon computing the time-derivative of the exact solution (24) and noting that the relations

$$\dot{\phi}(t) = \langle \xi(t) \rangle_b, \quad (27)$$

$$\dot{\phi}(t) = - \int_0^t d\tau \mathcal{A}(t, t - \tau), \quad (28)$$

$$\dot{A}(t) = \int_0^t d\tau \mathcal{S}(t, t - \tau) \quad (29)$$

hold, we find the surprising fact that the exact solution obeys the quantum master equation (15)! Or put differently, for pure phase noise, the exact result can be obtained within second-order perturbation theory from the master equation (12). For large times t , this master equation becomes the standard Markovian Bloch–Redfield equation. Thus, we find that the latter contains the exact long-time limit of the rates (27)–(29).

Before discussing the relation between both approaches in more detail, we like to close this section by writing the exact solution (24) also in terms of the usual bath spectral density [10,11]

$$J(\omega) = \sum_k |g_k|^2 \delta(\omega - \omega_k). \quad (30)$$

For the important special case of a heat bath that is initially at thermal equilibrium, we find $\varphi(t) = 0$, while the phase $\phi(t)$ and the damping amplitude $A(t)$ read

$$\phi(t) = \int d\omega J(\omega) \frac{\omega t - \sin(\omega t)}{\omega^2}, \quad (31)$$

$$A(t) = \int d\omega J(\omega) \frac{1 - \cos(\omega t)}{\omega^2} \coth \left(\frac{\hbar\omega}{2k_B T} \right). \quad (32)$$

5. When second-order is exact

We have seen that the time-local master equation (10) derived within second-order perturbation theory provides the exact time evolution of the reduced density matrix, which implies that in the expansion of the Liouvillian $\mathcal{H}(t)$, all higher order contributions vanish. This on the one hand nicely simplifies practical calculations. On the other hand, it poses the question whether we face a coincidence or whether there is any profound reason for the equivalence. In order to underline the latter point of view, we now demonstrate that for phase noise, the time-ordered cumulant in the ℓ th order generator (11) is proportional to the usual classical cumulant of the initial bath state. Consequently, we can argue that for the Gaussian initial state (7), the series $\mathcal{H}(t) = \sum_\ell \mathcal{H}_\ell(t)$ terminates after $\ell = 2$, which implies that the second-order time-local master equation (12) is exact.

We start out by defining averages with respect to the P -function as

$$\langle \dots \rangle_P = \int \dots \prod_k P(\beta_k, \beta_k^*) d^2 \beta_k. \quad (33)$$

With this notation, the exact solution (21) reads

$$\tilde{\rho}_{mn}(t) = \left\langle \exp \left\{ \int_0^t d\tau f_{mn}(\tau) \right\} \right\rangle_P \tilde{\rho}_{mn}(0) \quad (34)$$

with the complex-valued function

$$\begin{aligned} f_{mn}(t) &= i(X_m^2 - X_n^2)\phi(t) - \sum_k \left\{ (X_m - X_n)^2 \frac{|z_k(t)|^2}{2} \right. \\ &\quad \left. - (X_m - X_n)[z_k(t)\beta_k^* - z_k^*(t)\beta_k] \right\}. \end{aligned} \quad (35)$$

The average in Eq. (34) is obtained from a distribution function for the c -numbers β_k . Thus, it can be formally considered as the averaged solution of a stochastic differential equation that obeys a time-local differential equation of the form (10), but with the generator \mathcal{H} now being a c -number, not an operator. Thus, we can adapt the line

of argumentation given by van Kampen for classical Gaussian stochastic processes [48]: Differentiating the Taylor expansion of Eq. (34), we find

$$\begin{aligned} \dot{\tilde{\rho}}_{mn}(t) = & \left[\langle f_{mn}(t) \rangle_P + \int_0^t dt_1 \langle f_{mn}(t) f_{mn}(t_1) \rangle_P \right. \\ & \left. + \int_0^t dt_1 \int_0^{t_1} dt_2 \langle f_{mn}(t) f_{mn}(t_1) f_{mn}(t_2) \rangle_P + \dots \right] \tilde{\rho}_{mn}(0). \end{aligned} \quad (36)$$

A time-local equation of motion for $\tilde{\rho}_{mn}(t)$ can be obtained by inserting $\tilde{\rho}_{mn}(0)$ from Eq. (34), which yields [48]

$$\begin{aligned} \dot{\tilde{\rho}}_{mn}(t) = & \sum_{\ell=1}^{\infty} \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{\ell-2}} dt_{\ell-1} \\ & \times \langle \langle f_{mn}(t) f_{mn}(t_1) \dots f_{mn}(t_{\ell-1}) \rangle \rangle_P \tilde{\rho}_{mn}(t), \end{aligned} \quad (37)$$

where $\langle \langle \dots \rangle \rangle_P$ denotes the cumulants with respect to the P -function. Note that for the cumulants of a classical process, time-ordering is not relevant [48]. Thus the only difference of this expansion and the one in Eq. (11) for the quantum master equation is that the latter contains time-ordered cumulants.

For a Gaussian P -function, all cumulants of β_k and β_k^* beyond second-order vanish [54]. Since $f_{mn}(t)$ is linear in these variables, the same is true for the cumulants in Eq. (37) and, consequently, only the terms with $\ell = 1, 2$ contribute to this expansion. Evaluating the expansion coefficients explicitly, one finds that they are identical to those of the second-order time-local master equation (15).

The equivalence of the second-order master equation and the exact solution is based on two requirements: First, the coupling operator X needs to be diagonal in the eigenbasis of the system, so that its interaction-picture representation is time-independent, $\tilde{X}(t) = X$, and, thus, it can be effectively treated as a c -number. Hence the quantum mechanical time-ordering affects only the bath coordinate $\tilde{\xi}(t)$ for which we can express multi-time expectation values as cumulants of the P -function. In that way, we can circumvent the tedious task of normal-ordering the operators b_k and b_k^\dagger . With this precondition, secondly, the Gaussian initial state of the bath ensures that the cumulant expansion terminates after the second-order and agrees with the expansion of the master equation (10). For any non-Gaussian state, infinitely many higher-order cumulants are non-zero both in the classical case [55–57] and in the quantum mechanical case [58–60]. Consequently, the expansion of the Liouvillian is of infinite order and any truncation represents an approximation.

Let us finally stress that the second-order Nakajima–Zwanzig master equation [43,44], which was not considered in this work, can be expressed in terms of cumulants (the so-called partial cumulants), as well [61,62]. Note that the ordered and the partial cumulants up to second-order coincide. However, the second-order Nakajima–Zwanzig equation is not of a time-local form and therefore cannot yield the exact result for the model discussed here [63]. Thus,

phase noise constitutes an example for which the time-local approach is more accurate than the time-non-local one when comparing their perturbation expansions up to the same order. This outcome is in agreement with some recent findings for harmonic oscillator baths [64,65] and for spin baths [66,67]. For example for a two-level system coupled via XY Heisenberg interaction to a spin bath, the differences of both approaches have been analysed quantitatively [66]. Nevertheless, we do not give a general recommendation in favour of one or the other approach because the quality of each seems to be model dependent [61,68–70].

6. Conclusion

Quantum systems under the influence of pure phase noise represent an important special case of dissipative quantum mechanics owing to the existence of an exact solution. Moreover, on short time scales, on which the coherent system dynamics cannot manifest itself, the behaviour of the phase noise model is even generic [23]. Here, we have presented the explicit exact solution for a quantum system under the influence of phase noise with a general Gaussian initial bath state. Thereby, we have demonstrated that the coherence decay is determined by the symmetric bath correlation function, while the anti-symmetric correlation function gives rise to a time-dependent phase shift. In turn, from the exact relations (27)–(29), one can obtain information about the spectral properties of the bath by comparing our results with the experimentally observed dephasing at short times.

Despite the exact solvability of the phase-noise problem, it is often convenient to study the resulting dephasing within a master equation approach based on second-order perturbation theory in the system–bath coupling. For the (time-non-local) Nakajima–Zwanzig equation, this constitutes an approximation. For the time-local version of such a master equation, by contrast, we have found that it provides the exact solution. After noticing that this facilitates practical calculations, one might wonder why and when this equivalence holds true. By mapping the initial bath density operator to a P -function, we showed that a formal expansion of the time-local master equation for phase noise is in fact an ordinary cumulant expansion. Consequently, for a Gaussian initial bath state, all terms beyond the second-order vanish, so that the master equation becomes exact. Thus for a bosonic heat bath, there are two conditions for the exact agreement: first, the system–bath coupling must commute with the system Hamiltonian constituting the case of pure phase noise and, second, the initial state of the bath must correspond to a Gaussian P -function. If one of these conditions is violated, there might still exist an exact solution, but it can no longer be obtained within second-order perturbation theory, as for example is the case for the dissipative harmonic oscillator [13,14].

The second-order time-local master equation employed in this work agrees with the exact solution at any time. In particular in the long-time limit, it becomes Markovian and identical to the standard Bloch–Redfield master equation, which for pure phase noise is of Lindblad form. This also explains the previously observed “excellent agreement” [71] between the exact dynamics and results obtained within Bloch–Redfield theory.

Acknowledgements

P.H. and S.K. like to thank Wolfgang Domcke for many scientific discussions and wish him all the best for the future. This work was supported by Deutsche Forschungsgemeinschaft through SFB 484 and SFB 631. P.H. and S.K. acknowledge funding by the DFG excellence cluster “Nanosystems Initiative Munich”.

Appendix A. Derivation of the exact solution

In this appendix, we outline the derivation of the exact reduced dynamics discussed in Section 4. As a first step, we perform a transformation to the interaction picture with respect to H_0 , so that the coupling Hamiltonian (3) becomes

$$\tilde{H}_{\text{sb}}(t) = \tilde{V}(t) + \tilde{V}^\dagger(t) \quad (\text{A.1})$$

with $\tilde{V}(t) = \hbar X \sum_k g_k b_k \exp(-i\omega_k t)$ and the hermitian system operator X . The two operators \tilde{V} and \tilde{V}^\dagger do not commute. However, their commutator is an operator in the Hilbert space of the system, while being a c -number in the bath Hilbert space. To be specific, one obtains $[\tilde{V}(t), \tilde{V}^\dagger(t')] = f(t - t')$, where

$$f(t) = \hbar^2 X^2 \sum_k |g_k|^2 e^{-i\omega_k t}. \quad (\text{A.2})$$

Hence, we can use the Baker–Campbell–Hausdorff formula (see Ref. [30]) to express the time-ordered propagator (20) as a product of two exponentials

$$\begin{aligned} U(t) = & \exp \left\{ \frac{1}{i\hbar} \int_0^t d\tau \tilde{H}_{\text{sb}}(\tau) \right\} \\ & \times \exp \left\{ -\frac{1}{\hbar^2} \int_0^t d\tau \int_0^\tau d\tau' f(\tau - \tau') \right. \\ & \left. \times [\theta(\tau - \tau') - \theta(\tau' - \tau)] \right\}. \end{aligned} \quad (\text{A.3})$$

The first exponential in Eq. (A.3) can be written as

$$\exp \left\{ \frac{1}{i\hbar} \int_0^t d\tau \tilde{H}_{\text{sb}}(\tau) \right\} = \prod_k D_k(z_k X) \quad (\text{A.4})$$

with the complex number $z_k = g_k^* [1 - \exp(i\omega_k t)] / \omega_k$ and the displacement operators $D_k(Y) = \exp(Y b_k^\dagger - Y^\dagger b_k)$. The second exponential in Eq. (A.3) provides the time-dependent phase factor $\exp\{iX^2\phi(t)\}$ with

$$\phi(t) = \int_0^t d\tau \int_0^\tau d\tau' \sum_k |g_k|^2 \sin(\omega_k \tau'). \quad (\text{A.5})$$

We will now consider the elements of the reduced density matrix in the eigenbasis $\{|n\rangle\}$ of the system Hamiltonian. Using the initial condition (5), Eq. (19) becomes

$$\begin{aligned} \tilde{\rho}_{mn}(t) &= \text{tr}_b \langle m | U(t) \rho(0) \rho_b U^\dagger(t) | n \rangle \\ &= \rho_{mn}(0) e^{i[X_m^2 - X_n^2]\phi(t)} \left\langle \prod_k D_k^\dagger(X_n z_k) \prod_{k'} D_{k'}^\dagger(X_m z_{k'}) \right\rangle_b, \end{aligned} \quad (\text{A.6})$$

where $\langle \dots \rangle_b = \text{tr}_b(\rho_b \dots)$. By virtue of the relations $D_k^\dagger(Y) = D_k^\dagger(-Y)$ and

$$D_k(Y) D_k(Z) = D_k(Y + Z) \exp\{(YZ^\dagger - Y^\dagger Z)/2\}, \quad (\text{A.8})$$

which hold for any commuting system operators Y and Z , we obtain

$$\tilde{\rho}_{mn}(t) = \rho_{mn}(0) e^{i[X_m^2 - X_n^2]\phi(t) + i\eta_{mn}} \times \left\langle \prod_k D_k(z_k [X_m - X_n]) \right\rangle_b. \quad (\text{A.9})$$

Note that an additional phase $\eta_{mn} = 2 \sum_k |z_k|^2 \text{Im}(X_m^* X_n)$ vanishes, since the system operator X is hermitian. It remains to evaluate the expectation value in the second line of Eq. (A.9). This is readily established by writing the bath state ρ_b in its P -function representation [see Eq. (6)] and noticing that expectation values of normal-ordered products of annihilation and creation operators are identical to the moments of the P -function, where b_k^\dagger and b_k have to be replaced by β_k^* and β_k , respectively [30]. Thus, by using the Baker–Campbell–Hausdorff formula

$$\exp(\lambda b_k^\dagger - \lambda^* b_k) = \exp(\lambda b_k^\dagger) \exp(\lambda^* b_k) \exp(-|\lambda|^2/2) \quad (\text{A.10})$$

for each mode k , we write the second line of Eq. (A.9) in its normal-ordered form

$$e^{-[X_m - X_n]^2 \sum_k |z_k|^2/2} \left\langle \prod_k e^{z_k [S_m - S_n] b_k^\dagger} e^{z_k^* [S_m - S_n] b_k} \right\rangle_P, \quad (\text{A.11})$$

where $\langle \dots \rangle_P$ denotes the average (33) with respect to the P -function. Using also the fact that the P -function (7) factorises with respect to the modes k , we arrive at Eq. (21).

References

- [1] A. Kuhl, W. Domcke, Chem. Phys. 259 (2000) 227.
- [2] D. Egorova, M. Thoss, W. Domcke, H. Wang, J. Chem. Phys. 119 (2003) 2761.
- [3] M. Čížek, M. Thoss, W. Domcke, Phys. Rev. B 70 (2004) 125406.
- [4] S. Kohler, J. Lehmann, P. Hänggi, Phys. Rep. 406 (2005) 379.
- [5] D. Vion, A. Aassime, A. Cottet, P. Joyez, H. Pothier, C. Urbina, D. Esteve, M.H. Devoret, Science 296 (2002) 886.
- [6] I. Chiorescu, Y. Nakamura, C.J.P. Harmans, J.E. Mooij, Science 299 (2003) 1869.
- [7] T. Yamamoto, Y.A. Pashkin, O. Astafiev, Y. Nakamura, J.S. Tsai, Nature (London) 425 (2003) 941.
- [8] V.B. Magalinskii, Zh. Eksp. Teor. Fiz. 36 (1959) 1942; V.B. Magalinskii, Sov. Phys. JETP 9 (1959) 1381.
- [9] A.O. Caldeira, A.L. Leggett, Ann. Phys. (NY) 149 (1983) 374.
- [10] A.J. Leggett, S. Chakravarty, A.T. Dorsey, M.P.A. Fisher, A. Garg, W. Zwerger, Rev. Mod. Phys. 59 (1987) 1.

- [11] P. Hänggi, P. Talkner, M. Borkovec, *Rev. Mod. Phys.* 62 (1990) 251.
- [12] H. Grabert, U. Weiss, P. Hänggi, *Phys. Rev. Lett.* 25 (1984) 2193.
- [13] H. Grabert, U. Weiss, P. Talkner, *Z. Phys. B* 55 (1984) 87.
- [14] H. Grabert, P. Schramm, G.-L. Ingold, *Phys. Rep.* 168 (1988) 115.
- [15] P. Riseborough, P. Hänggi, U. Weiss, *Phys. Rev. A* 31 (1985) 471.
- [16] C. Zerbe, P. Hänggi, *Phys. Rev. E* 52 (1995) 1533.
- [17] M. Wubs, K. Saito, S. Kohler, Y. Kayanuma, P. Hänggi, *Phys. Rev. Lett.* 97 (2006) 200404.
- [18] J. Luczka, *Physica A* 167 (1990) 919.
- [19] N.G. van Kampen, *J. Stat. Phys.* 78 (1995) 299.
- [20] G.M. Palma, K.-A. Suominen, A.K. Ekert, *Proc. R. Soc. Lond. Ser. A* 452 (1996) 567.
- [21] L.-M. Duan, G.-C. Guo, *Quantum Semiclass. Opt.* 10 (1998) 611.
- [22] D. Mozyrsky, V. Privman, *J. Stat. Phys.* 91 (1998) 787.
- [23] D. Braun, F. Haake, W.T. Strunz, *Phys. Rev. Lett.* 86 (2001) 002913.
- [24] B. Krummheuer, V.M. Axt, T. Kuhn, *Phys. Rev. B* 65 (2002) 195313.
- [25] R. Doll, M. Wubs, P. Hänggi, S. Kohler, *Europhys. Lett.* 76 (2006) 547.
- [26] R. Doll, M. Wubs, P. Hänggi, S. Kohler, *Phys. Rev. B* 76 (2007) 045317.
- [27] D. Solenov, D. Tolkunov, V. Privman, *Phys. Rev. B* 75 (2007) 035134.
- [28] K. Blum, *Density Matrix Theory and Applications*, second ed., Plenum Press, New York, 1996.
- [29] A. Perelomov, *Generalized Coherent States and Their Applications*, Theoretical and Mathematical Physics, Springer, Berlin, 1986.
- [30] C.W. Gardiner, *Quantum Noise*. Springer Series in Synergetics, vol. 56, Springer, Berlin, 1991.
- [31] N.G. van Kampen, *Stochastic Processes in Physics and Chemistry*, North-Holland, Amsterdam, 2001.
- [32] H.-P. Breuer, F. Petruccione, *The Theory of Open Quantum Systems*, Oxford University Press, Oxford, 2002.
- [33] A. Fuliński, *Phys. Lett. A* 25 (1967) 13.
- [34] A. Fuliński, W.J. Kramarczyk, *Physica* 39 (1968) 575.
- [35] M. Tokuyama, H. Mori, *Prog. Theor. Phys.* 55 (1976) 411.
- [36] N.G. van Kampen, *Physica* 74 (1974) 215.
- [37] P. Hänggi, H. Thomas, *Z. Phys. B* 26 (1977) 85.
- [38] H. Grabert, P. Talkner, P. Hänggi, *Z. Phys. B* 26 (1977) 389.
- [39] F. Shibata, Y. Takahashi, N. Hashitsume, *J. Stat. Phys.* 17 (1977) 171.
- [40] F. Shibata, N. Hashitsume, *J. Phys. Soc. Jpn.* 44 (1978) 1435.
- [41] P. Hänggi, H. Thomas, H. Grabert, P. Talkner, *J. Stat. Phys.* 18 (1978) 155.
- [42] P. Hänggi, H. Thomas, *Phys. Rep.* 88 (1982) 206.
- [43] S. Nakajima, *Prog. Theor. Phys.* 20 (1958) 948.
- [44] R. Zwanzig, *J. Chem. Phys.* 33 (1960) 1338.
- [45] F. Haake, *Quantum Statistics in Optics and Solid-State Physics*. Springer Tracts in Modern Physics, vol. 66, Springer, Berlin, 1973.
- [46] K.M. Fonseca Romero, P. Talkner, P. Hänggi, *Phys. Rev. A* 69 (2004) 052109.
- [47] R. Kubo, *J. Phys. Soc. Jpn.* 17 (1962) 1100.
- [48] N.G. van Kampen, *Physica* 74 (1974) 239.
- [49] S. Chaturvedi, F. Shibata, *Z. Physik. B* 35 (1979) 297.
- [50] H.-P. Breuer, B. Kappler, F. Petruccione, *Ann. Phys.* 291 (2001) 483.
- [51] K. Kraus, *Ann. Phys. (NY)* 64 (1970) 311.
- [52] G. Lindblad, *Commun. Math. Phys.* 48 (1976) 119.
- [53] S. Banerjee, R. Gosh, arXiv:quant-ph/0703054, 2007.
- [54] H. Risken, *The Fokker-Planck Equation*. Springer Series in Synergetics, vol. 18, Springer, Berlin, 1984.
- [55] J. Marcinkiewicz, *Math. Z.* 44 (1938) 612.
- [56] R.F. Pawula, *Phys. Rev.* 162 (1967) 186.
- [57] P. Hänggi, P. Talkner, *J. Stat. Phys.* 22 (1980) 65.
- [58] A.K. Rajagopal, E.C.G. Sudarshan, *Phys. Rev. A* 10 (1974) 1852.
- [59] U.M. Titulaer, *Phys. Rev. A* 11 (1975) 2204.
- [60] R. Schack, A. Schenzle, *Phys. Rev. A* 41 (1990) 3847.
- [61] B. Yoon, J.M. Deutch, J.H. Freed, *J. Chem. Phys.* 62 (1975) 4687.
- [62] F. Shibata, T. Arimitsu, *J. Phys. Soc. Jpn.* 49 (1980) 891.
- [63] D. Solenov, V. Privman, *Proc. SPIE* 5436 (2004) 172.
- [64] Y.J. Yan, R.X. Xu, *Annu. Rev. Phys. Chem.* 126 (2005) 114102.
- [65] A. Royer, *Phys. Lett. A* 315 (2003) 335.
- [66] H.-P. Breuer, D. Burgarth, F. Petruccione, *Phys. Rev. B* 70 (2004) 045323.
- [67] H. Krovi, O. Oreshkov, M. Ryazanov, D.A. Lidar, arXiv:0707.2096 [quant-ph], 2007.
- [68] S. Mukamel, I. Oppenheim, J. Ross, *Phys. Rev. A* 17 (1978) 1988.
- [69] D.R. Reichman, F.L.H. Brown, P. Neu, *Phys. Rev. E* 55 (1997) 2328.
- [70] M. Schröder, M. Schreiber, U. Kleinekathöfer, *J. Chem. Phys.* 126 (2007) 114102.
- [71] D.A. Lidar, Z. Bihary, K.B. Whaley, *Chem. Phys.* 268 (2001) 35.