

Bath-induced coherence and the secular approximation

P. R. Eastham,¹ P. Kirton,² H. M. Cammack,² B. W. Lovett,² and J. Keeling²

¹*School of Physics and CRANN, Trinity College Dublin, Dublin 2, Ireland*

²*SUPA, School of Physics and Astronomy, University of St Andrews, St. Andrews KY16 9SS, United Kingdom*

(Received 21 October 2015; revised manuscript received 18 March 2016; published 19 July 2016)

Finding efficient descriptions of how an environment affects a collection of discrete quantum systems would lead to new insights into many areas of modern physics. Markovian, or time-local, methods work well for individual systems, but for groups a question arises: Does system-bath or intersystem coupling dominate the dissipative dynamics? The answer has profound consequences for the long-time quantum correlations within the system. We consider two bosonic modes coupled to a bath. By comparing an exact solution against different Markovian master equations, we find that a smooth crossover of the equations of motion between dominant intersystem and system-bath coupling exists—but it requires a nonsecular master equation. We predict singular behavior of the dynamics and show that the ultimate failure of nonsecular equations of motion is essentially a failure of the Markov approximation. Our findings support the use of time-local theories throughout the crossover between system-bath-dominated and intersystem-coupling-dominated dynamics.

DOI: [10.1103/PhysRevA.94.012110](https://doi.org/10.1103/PhysRevA.94.012110)

I. INTRODUCTION

A Markovian system is one in which the future time evolution depends only on the current state, and not on its history [1]. In the context of open quantum systems, Markovianity generally implies that the reduced density operator obeys a first-order differential equation. This class of theory has been developed for many years, is applied to a vast range of systems, and provides our understanding of quantum damping and decoherence [2]. Recent work, however, presents it with challenges. The development of solid-state quantum emitters, such as single and coupled quantum dots [3,4] and superconducting-qubit cavities [5,6], demands theories capable of treating driven or coupled systems damped by complex structured baths [7–11]. Such theories reveal, among other effects, the possibility of engineering the reservoirs to control quantum coherence [12,13]. They show that under appropriate conditions both quantum coherence [14] and entanglement [15–17] can survive indefinitely, even for high-temperature baths [18,19].

These problems do not necessarily elude treatment by a time-local theory, i.e., a (Markovian) quantum master equation. Such theories accurately reproduce the intensity-dependent damping of quantum dots in a structured reservoir [4,20,21], for example, and provide recent predictions of bath-induced coherence [14] and entanglement [16,17]. However, there are several master equations consistent with, and derivable from, the assumption of weak coupling [22,23]. Furthermore, master equations are often postulated phenomenologically, by the choice of the jump operators in the Lindblad form. For problems with multiple oscillators and structured baths this choice is not straightforward, with different choices plausible in different limits. Nor is it innocent: different forms of master equation lead to different behaviors [24]. Thus it is important to establish which, if any, of the various time-local theories is correct.

In this paper we address this question by studying an exactly solvable model and comparing the exact solution against various time-local theories. We consider a model of two bosonic modes, $\hat{\psi}_{a,b}$, with frequencies $\omega_{a,b}$, coupled to a

thermally occupied bath with spectral density $J(\nu)$. This model has a nontrivial Hamiltonian, multiple degrees of freedom, and frequency-dependent damping yet is exactly solvable. We consider the general case where the natural frequencies $\omega_{a,b}$ differ and the bath couples to a superposition of modes, $\varphi_a^* \hat{\psi}_a + \varphi_b^* \hat{\psi}_b$, and calculate the evolution of the coherence, $\langle \hat{\psi}_a^\dagger \hat{\psi}_b \rangle$. We find a complex behavior with multiple regimes, shown in Fig. 2, reflecting the competing effects of the system Hamiltonian and the coupling to the bath. We compare the exact solution with time-local theories and, so, identify those which correctly capture such physics. This allows us to establish their validity in a generic problem and avoids the difficulty inherent in studying only approximate theories.

A physical issue we address is the appropriate form of dissipator for systems with multiple components. Two different forms are expected on physical grounds [25]. In the case of the two-oscillator model it is clear that at resonance, $\omega_a = \omega_b$, the damping can depend only on the pattern of coupling to the baths. Thus we expect collective decay, described by a Lindblad form, $L_c = \Gamma^\downarrow \mathcal{L}[\varphi_a^* \hat{\psi}_a + \varphi_b^* \hat{\psi}_b] + \Gamma^\uparrow \mathcal{L}[\varphi_a \hat{\psi}_a^\dagger + \varphi_b \hat{\psi}_b^\dagger]$, where $\mathcal{L}[\hat{\psi}]$ is the standard dissipator with jump operator $\hat{\psi}$ [2]. Far off-resonance, however, we expect individual decay terms, $L_i = \sum_{i=a,b} \Gamma_i^\downarrow \mathcal{L}[\hat{\psi}_i] + \sum_i \Gamma_i^\uparrow \mathcal{L}[\hat{\psi}_i^\dagger]$. The first form predicts a nonzero steady-state coherence, while the second predicts that this vanishes. We show that neither of these forms is, in general, correct, and both make misleading predictions outside of limiting cases. Nonetheless, we find that the general behavior can be accurately treated by a time-local theory, specifically a Bloch-Redfield (BR) equation. While one might naïvely have expected some smooth crossover between the limits captured by L_c and L_i , the real answer is more subtle: a smooth interpolation exists for the equations of motion, but the steady state is *singular* at degeneracy. This allows mutually exclusive behavior in different regimes and implies that some useful effects—specifically, the protection of coherence against the bath—are critically sensitive to microscopic parameters. Moreover, while the crossover can be treated by a time-local theory, this theory is not a Lindblad form with the required positive rates. The use of such forms is

the subject of ongoing debate, since they are not completely positive maps [26]. This means that they can lead to unphysical density operators, with negative eigenvalues.

A methodological issue in this debate is the procedure of secularization. This amounts to removing from the equations of motion those terms which are time dependent in the interaction picture. It was used in some of the earliest work on quantum damping by Bloch and Wangsness [27] but was then argued to be unnecessary by Redfield [28] as well as Bloch [29]. That position was challenged by the subsequent Lindblad theorem [30]: as argued by Dümcke and Spohn [23], secularization is required to reach a description where Lindblad’s theorem ensures positivity of the density operator. Indeed, Lindblad’s theorem guarantees that the density operator will remain positive even when there is entanglement with an auxiliary system, a criterion known as complete positivity [2]. Secularization, which leads to a completely positive theory, is clearly appropriate when the interaction-picture time dependence is fast, since off-diagonal terms then rapidly average to 0. In our case, this is far off-resonance, and secularization indeed leads from the BR equation to the form L_i . However, for a tunable system it may occur that the time dependence in the interaction picture becomes slow in certain regimes, i.e., approaching resonance, so that secularization becomes inappropriate. An interesting improved version of the secularization procedure is studied in Ref. [26].

Recently, the necessity of secularization has been questioned [31,32]: Simulations indicate that for time evolution following an initially prepared separable state, secularization (and even a Lindblad form for the equation of motion) can be unnecessary for positivity [33] and even complete positivity [34], in particular, for time-convolutionless [2] and Nakajima-Zwanzig [35,36] approaches. Stronger statements to this effect have also been made by Hell *et al.* [37], noting that a conservation law [38] is violated by secularized theories; we discuss this sum rule in detail further below. The question of how the operator forms of time-local and non-Markovian approaches are related is reviewed by Karlewski and Marthaler [39]. Our focus in this paper is, however, on cases where a time-local description is sufficient. This will enable us to explore the entire parameter space of a model and identify the regions where BR equations predict physical behavior. We show that, although the damping is not of Lindblad form, the anticipated unstable behavior does not occur within the domain of applicability of the theory—specifically, as long as the bath remains Markovian.

The exact results we present are restricted to only a subset of possible initial density matrices. We take as initial conditions a thermal state of the bath and the ground state of the two oscillators. The reduced density matrix is then Gaussian at all times and, so, completely characterized by its second moments. Thus we are able to establish whether the BR equations are accurate and physical from the dynamics of those moments alone. This does not, however, rule out inaccurate or even unphysical behavior for arbitrary (non-Gaussian) initial density matrices.

Within the scope of coupled open quantum systems, a particular motivation for our work comes from the timely theory of “weak lasing” [40] introduced in the context of polariton

condensates. The idea presented is that for modes which are close to resonance, (dissipative) radiative coupling can select which linear combination of modes lases (condenses) first. These works started from a phenomenological description of radiative coupling, in which collective dissipation terms are introduced by hand. In the following we see, however, that the effects of collective dissipation terms are strongly dependent on whether or not the individual modes are degenerate. Our work does not consider the general problem with both drive and dissipation, but the results we present for coupling to a single bath suggest that there may be a need to re-examine how weak lasing evolves where radiative coupling selects superpositions of nondegenerate modes.

The remainder of this paper is structured as follows. In Sec. II we describe the model. In Sec. III we present the exact solution and discuss its behavior. In Sec. IV we discuss the comparison with the BR equation and the naïve Lindblad forms mentioned above. We also identify the parameter regimes where the BR equation gives physical behavior. In Sec. V we develop an alternative to the BR equation and show it to be an improvement both numerically and analytically. In Sec. VI we report the generalization of our work to the case of multiple baths. Finally, in Sec. VII, we give our conclusions.

II. MODEL

The two bosons and the common bosonic bath are represented by the Hamiltonian $\hat{H} = \hat{H}_S + \hat{H}_{SB} + \hat{H}_B$. The system Hamiltonian is $\hat{H}_S = \omega_a \hat{\psi}_a^\dagger \hat{\psi}_a + \omega_b \hat{\psi}_b^\dagger \hat{\psi}_b$, in terms of bosonic annihilation operators $\hat{\psi}_i$. The bath Hamiltonian is $\hat{H}_B = \sum_i \omega_i \hat{c}_i^\dagger \hat{c}_i$, where c_i annihilates a boson in mode i . The system-bath coupling takes the form

$$\hat{H}_{SB} = (\varphi_a^* \hat{\psi}_a^\dagger + \varphi_b^* \hat{\psi}_b^\dagger) \sum_i g_i \hat{c}_i + \text{H.c.} \quad (1)$$

The complex coefficients φ_i determine which pattern of system operators the bath couples to, and g_i captures the overall coupling to mode i . We assume that the bath has a continuous density of states parameterized by the spectral density $J(\nu) = \sum_i g_i^2 \delta(\nu - \omega_i)$.

Since this model is a linear system of coupled harmonic oscillators it is exactly solvable. The exact solution for a single harmonic oscillator coupled to a bath [41] is well known [42]. The extension to the case of two identical oscillators coupled symmetrically to a bath can be found in Ref. [43] and has been used to test master equation approaches [31]. In this special case the normal modes exactly match the pattern of bath coupling. The antisymmetric mode then decouples from the bath, immediately reducing the problem to one damped oscillator and one undamped one. The dynamics of entanglement in this case was studied by Paz and Roncaglia [15], who showed that the undamped mode allows entanglement to persist indefinitely. We consider a more general problem, including detuning $\Delta = (\omega_a - \omega_b)/2 \neq 0$, which prevents such a decoupling and leads to finite lifetimes.

The existence of finite lifetimes at nonzero Δ can be understood by observing that the model above is equivalent to a system of two coupled oscillators, one of which is coupled to a bath, i.e., the Hamiltonian $\hat{H}_S = \omega_c \hat{\psi}_c^\dagger \hat{\psi}_c + \omega_d \hat{\psi}_d^\dagger \hat{\psi}_d +$

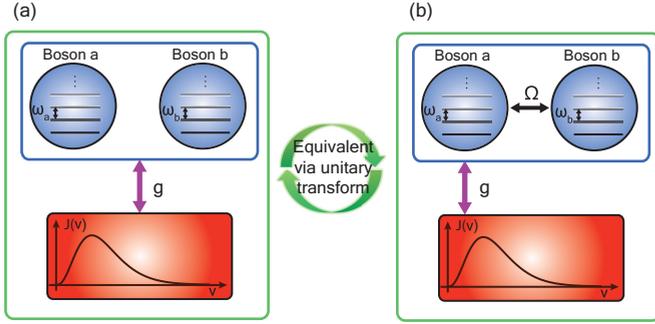


FIG. 1. Cartoon of the system we consider. (a) Two bosonic modes of frequencies ω_a and ω_b coupled collectively to a single bath. As illustrated, we take a superohmic bath with an exponential cutoff when an explicit form is required. (b) The equivalent problem of two coupled modes with a bath coupling to only one of the modes.

$\Omega \hat{\psi}_c^\dagger \hat{\psi}_d + \text{H.c.}$, with $\hat{H}_{SB} = \hat{\psi}_c^\dagger \sum_i g_i \hat{c}_i + \text{H.c.}$ This mapping follows upon transforming the latter problem to a basis in which \hat{H}_S is diagonal. These two equivalent problems are illustrated schematically in Fig. 1. We use the basis of Eq. (1) in the following; the results of the other problem can be simply extracted by the appropriate rotations.

In what follows, we consider the time evolution of the observables $F_{ij}(t) \equiv \langle \hat{\psi}_i^\dagger \hat{\psi}_j \rangle$, focusing, in particular, on the coherence $F_{ab}(t)$, which, as mentioned earlier, distinguishes collective from individual decay. Furthermore, these observables fully characterize the density matrix for the initial conditions we consider; it is Gaussian, so that higher moments are related to the F_{ij} by Wick's theorem. We first present the exact solution and discuss its observed properties, before considering the (nonsecularized) Bloch-Redfield equation of motion. We show both analytically and numerically that this approach reproduces the exact solution, while either of the naïve Lindblad master equations fails to reproduce the exact results.

III. EXACT SOLUTION

The exact time evolution can be readily found by using a Laplace transform to write the system operators in terms of the $t = 0$ bath operators and then evaluating $F_{ij}(t)$ using thermal correlations for the bath operators at $t = 0$. With the oscillators in the ground state at $t = 0$ we find

$$F_{ij}(t) = \int d\nu J(\nu) n_B(\nu) W_i^*(\nu, t) W_j(\nu, t), \quad (2)$$

$$W_i(\nu, t) = \varphi_i \int \frac{d\zeta}{2\pi} \frac{(\omega_i - \zeta) e^{-i\zeta t}}{(\nu - \zeta - i0) d(\zeta + i0)}, \quad (3)$$

where $\omega_{\bar{a}} = \omega_b$, and vice versa, $n_B(\nu)$ is the Bose-Einstein distribution function, and $d(\zeta) = -(\omega_a - \zeta)(\omega_b - \zeta) + iK^*(\zeta)[|\varphi_a|^2(\omega_b - \zeta) + |\varphi_b|^2(\omega_a - \zeta)]$ is the denominator of the retarded Green's function. Here we have introduced $K(\zeta)$, the analytic continuation of the damping rate to the lower half-plane $K(\zeta) = i \int dx J(x)/(x - \zeta + i0)$. For real ζ the real part of $K(\zeta)$ is proportional to the spectral density, while the imaginary part follows from a Kramers-Kronig relation. In the numerical results which follow we use the form of spectral

density illustrated in Fig. 1. For numerical evaluation, it is computationally more efficient to write this as a convolution:

$$F_{ij}(t) = \int_0^t d\tau \int_0^t d\sigma D_i(t - \tau)^* D_j(t - \sigma) \alpha(\sigma - \tau), \quad (4)$$

$$D_i(t) = \varphi_i \int \frac{d\zeta}{2\pi} \frac{(\omega_i - \zeta) e^{-i\zeta t}}{d(\zeta + i0)}, \quad (5)$$

$$\alpha(\tau) = \int d\nu J(\nu) n_B(\nu) e^{-i\nu\tau}. \quad (6)$$

One may readily check that this is equivalent to Eqs. (2) and (3).

A. Behavior near degeneracy

Using the above expressions, one may directly find how the coherence evolves with time, as the detuning $\Delta = (\omega_a - \omega_b)/2$ changes; this is shown in Fig. 2. As is clear in this figure, the behavior at degeneracy ($\Delta = 0$) and away from degeneracy is different. Near degeneracy there is strong, long-lived coherence, corresponding to the noise-induced coherence recently analyzed for few-level models [14,17]. What is not immediately clear, however, is that the degenerate limit is in fact singular: the form of $F_{ab}(\infty)$ is discontinuous as a function of frequency. We next discuss how and why this singular behavior occurs.

The origin of the discontinuity at degeneracy is the emergence of a slow mode, whose lifetime diverges as $\Delta \rightarrow 0$. The decay rates of oscillations can be extracted from the poles ζ^0 of the Keldysh Green's function, i.e., solutions of $d(\zeta^0) = 0$.

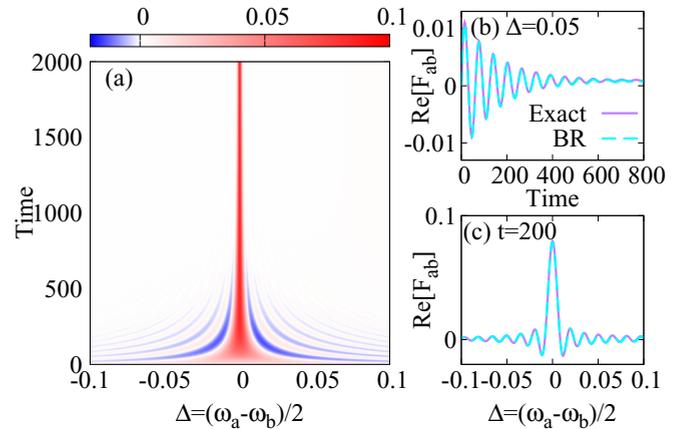


FIG. 2. (a) Evolution of coherence F_{ab} with time (vertical) and detuning (horizontal), found from the exact solution. We use a superohmic density of states with an exponential cutoff, $J(\omega) = J_0 e^{z-\omega/\omega_0} (\omega/\omega_0)^z$. This form is written such that its peak value is at $\omega = \omega_0$, and $J(\omega_0) = J_0$. We choose $z = 3$, and we measure all energies and times in units such that $\omega_a = 1$. In these units $\omega_0 = 0.9$, $J_0 = 0.001$, and the thermal occupation is controlled by $k_B T = 0.52$. The horizontal axis is the detuning, found by varying ω_b for a fixed ω_a . (b) Vertical slice at $\omega_b = 0.9$ corresponding to $\Delta = 0.05$, showing a comparison between the exact and the Bloch-Redfield theories. (c) Horizontal slice at $t = 200$. In the secularized theory the coherence $F_{ab} = 0$ for all times.

Writing $\omega_{a,b} = \Omega \pm \Delta$ leads to the expression

$$0 = \Delta^2 + \Delta(|\varphi_a|^2 - |\varphi_b|^2) i K^*(\zeta^0) - (\Omega - \zeta^0)[\Omega - \zeta^0 - i K^*(\zeta^0)(|\varphi_a|^2 + |\varphi_b|^2)]. \quad (7)$$

At $\Delta = 0$, the first line vanishes so it is clear that there is a pole $\zeta^0 = \Omega$, which is real and so entirely undamped. This has a simple physical interpretation: at $\Delta = 0$, there is no coupling between the combination $\sum_i \varphi_i \hat{\psi}_i$ and the orthogonal combination of fields. As such, the orthogonal combination is entirely undamped and maintains its original state. With nonzero detuning, beating between the modes $\hat{\psi}_{i=a,b}$ means that the orthogonal combination evolves into $\sum_i \varphi_i \hat{\psi}_i$ with time and is, thus, damped. Considering small Δ perturbatively gives

$$\text{Im}[\zeta^0] = -\frac{4\Delta^2 |\varphi_a|^2 |\varphi_b|^2}{(|\varphi_a|^2 + |\varphi_b|^2)^3} \frac{K'(\Omega)}{|K(\Omega)|^2} + O(\Delta^3). \quad (8)$$

This explains the Gaussian form of the singular response shown in Fig. 2: we expect $F_{ab}(\Delta, t \rightarrow \infty) \sim \bar{F}_{ab}(\Delta) + C \exp(-\alpha \Delta^2 t)$ at large t , where C , α are constant factors and $\bar{F}_{ab}(\Delta)$ is a smooth function.

B. Late-time asymptotes

The long-time asymptotes of the observables can be obtained from the pole structure of the Laplace-transform solution [44]. For late times, $W_i(v, t)$ simplifies significantly because the pole at $\zeta = v - i0$ lies on the real axis and so has a vanishing decay rate, while the poles of $d(\zeta + i0)$ are generically off axis and so have decayed at late times. Thus $W_i(v, t \rightarrow \infty) = [-i e^{-ivt}] \varphi_i (\omega_{\bar{i}} - v) / d(v + i0)$. This gives a simplified expression:

$$F_{ij}(\infty) = \varphi_i^* \varphi_j \int d\nu J(\nu) n_B(\nu) \frac{(\omega_{\bar{i}} - \nu)(\omega_{\bar{j}} - \nu)}{|d(\nu + i0)|^2}. \quad (9)$$

As can just be seen in Fig. 2, away from the resonance point, the off-diagonal coherence decays at late times to a small value but not strictly to 0. However, if the bath density of states and occupation are strictly flat, i.e., if $J(\nu) = J_0, n_B(\nu) = n_0$, then one may show that the asymptotic value $F_{ij}(\infty)$ vanishes for $\Delta \neq 0$. In this case Eq. (9) simplifies considerably, as $K(\nu) = \pi J_0$ for a flat bath, so $d(\nu + i0)$ becomes a simple polynomial. This integral then has only four simple poles, and one may readily check that it exactly vanishes—except at $\omega_a = \omega_b$, where two of the poles coincide and cancel with the zeros of the numerator. The small residual coherence that exists away from resonance in Fig. 2 is thus due to the frequency dependence of $n_B(\nu)$, $J(\nu)$.

IV. BLOCH-REDFIELD APPROACH

So far, we have seen that the exact solution of the bosonic problem does show a crossover between strong coherence at degeneracy and weak coherence, due to a frequency-dependent spectral density, away from degeneracy. However, this crossover occurs as a function of time, with coherence surviving over the range $\alpha t \simeq \Delta^{-2}$. A similar quadratically diverging time scale is found in the V-type system [14]. We now consider whether the behavior of the coherence, and

other observables, can be reproduced by a time-local master equation.

We may first note that neither of the naïve forms (individual or collective damping) discussed in Sec. I reproduces the correct behavior. Separate decay predicts that the coherence vanishes, for all times and detunings. The collective decay model does predict a strong long-lived coherence close to resonance and, indeed, a quadratically diverging lifetime. After this time, however, the coherence decays to 0, rather than the nonzero value predicted by the exact solution. More significantly, however, the collective form fails to reproduce the behavior once the detuning becomes significant. This may be seen from the steady-state populations: for large detuning or weak coupling these correspond to equilibrium with the bath, so $F_{aa} = n_B(\omega_a)$, $F_{bb} = n_B(\omega_b)$, whereas the Lindblad form gives equal populations. As pointed out by Cresser for the Jaynes-Cummings model [45], such master equations do not reach canonical equilibrium. More generally, since L_c is parameterized by one pair of forward-backward rates, it cannot account for the presence of two frequencies in the dynamics at which the bath should be sampled. As shown in Fig. 2, this occurs above a critical value of the detuning. Thus this model cannot possibly be accurate in this regime, unless the bath and its occupation are flat.

Thus, neither naïve form of dissipator can give a full account of problems with multiple system frequencies and structured baths, particularly if one seeks to analyze coherence. Unfortunately many interesting problems in solid-state quantum optics fall in this class, as discussed in the Introduction (Sec. I). We now show, however, that a BR equation does reproduce the correct behavior, as long as one does not secularize the final result. Such an approach is frequently stated to be invalid, as it leads to negative rates and instabilities. We, however, show analytically that such instabilities occur in a much restricted parameter regime, and, in fact, only when the Markov approximation breaks down. The nonsecularized theory is, also, often argued to be invalid on the related grounds that it is not a completely positive map and may not even be a positive one. We, however, show analytically that, although the map is not positive, it preserves positivity for almost all Gaussian states. Furthermore, we find numerically that these states soon dominate under the time evolution, even if dangerous ones are present under the initial conditions.

Following the standard method [2] one finds that the master equation has the form

$$\begin{aligned} \partial_t \rho = & -i[\hat{H}, \rho] + \sum_{ij} L_{ij}^\downarrow \varphi_i^* \varphi_j (2\hat{\psi}_j \rho \hat{\psi}_i^\dagger - [\rho, \hat{\psi}_i^\dagger \hat{\psi}_j]_+) \\ & + \sum_{ij} L_{ij}^\uparrow \varphi_i \varphi_j^* (2\hat{\psi}_j^\dagger \rho \hat{\psi}_i - [\rho, \hat{\psi}_i \hat{\psi}_j^\dagger]_+). \end{aligned} \quad (10)$$

Here the Hamiltonian includes Lamb shifts $\hat{H} = \hat{H}_S - \sum_{ij} h_{ij} \varphi_i^* \varphi_j \hat{\psi}_i^\dagger \hat{\psi}_j$. The matrices $L^{\sigma \in \downarrow, \uparrow}, h$ can be written in a compact form,

$$L^\sigma = \begin{pmatrix} K'_{a\sigma} & \bar{K}'_\sigma \pm i\delta K''_\sigma \\ \bar{K}'_\sigma \mp i\delta K''_\sigma & K'_{b,\sigma} \end{pmatrix}, \quad (11)$$

$$h = \begin{pmatrix} K''_a & \bar{K}'' - i\delta K' \\ \bar{K}'' + i\delta K' & K''_b \end{pmatrix}, \quad (12)$$

with the upper (lower) signs in Eq. (11) for L^\downarrow (L^\uparrow). Here we have introduced several new pieces of notation. We have used the shorthand $K_i = K(\omega_i)$ in terms of the Hilbert transform (analytic continuation) defined previously and have also defined Hilbert transforms of the excitation (absorption) rate, $K_{i\uparrow} = i \int d\xi n_B(\xi) J(\xi)/(\xi - \omega_i + i0)$, and de-excitation (emission) rate, $K_{i\downarrow} = i \int d\xi (n_B(\xi) + 1) J(\xi)/(\xi - \omega_i + i0)$. Note that this means $K_i = K_{i\downarrow} - K_{i\uparrow}$. Primes signify real and imaginary parts and $\bar{X} = (X_a + X_b)/2$, $\delta X = (X_a - X_b)/2$.

While Eqs. (10)–(12) fully describe the equations of motion, it is more convenient to use the (closed) set of equations of motion for the quantities F_{ij} derived from these master equations. In order to simplify these equations, it is convenient to note that the phase of the complex coefficients φ_i can be eliminated by a phase twist of the original operators, and we thus assume that φ_i is real henceforth. We may then define the vector of real quantities $\mathbf{f} = (F_{aa}, F_{bb}, 2F'_{ab}, 2F''_{ab})^T$ and produce an equation of motion $\partial_t \mathbf{f} = -\mathbf{M}\mathbf{f} + \mathbf{f}_0$, where

$$\mathbf{M} = \begin{pmatrix} 2\varphi_a^2 K'_a & 0 & \varphi_a \varphi_b K'_b & \varphi_a \varphi_b K''_b \\ 0 & 2\varphi_b^2 K'_b & \varphi_b \varphi_a K'_a & -\varphi_b \varphi_a K''_a \\ 2\varphi_a \varphi_b K'_a & 2\varphi_b \varphi_a K'_b & \Gamma_0 & -E_0 \\ -2\varphi_a \varphi_b K''_a & 2\varphi_b \varphi_a K''_b & E_0 & \Gamma_0 \end{pmatrix}, \quad (13)$$

with $E_0 = (\omega_b - \varphi_b^2 K''_b) - (\omega_a - \varphi_a^2 K''_a)$ and $\Gamma_0 = \varphi_a^2 K'_a + \varphi_b^2 K'_b$. None of these rates depend on the bath mode occupations, however, the constant vector $\mathbf{f}_0 = 2(\varphi_a^2 K'_{a,\uparrow}, \varphi_b^2 K'_{b,\uparrow}, \varphi_a \varphi_b 2\bar{K}'_\uparrow, -\varphi_a \varphi_b 2\delta K''_\uparrow)^T$ involves the excitation rate, so that populations are proportional to the bath occupations as expected.

The result of time evolving this closed set of equations is shown in Figs. 2(b) and 2(c) and clearly compares very well to the exact solution. Moreover, we can easily see that secularizing this set of equations, as is often claimed to be a crucial step [23], could only decrease the agreement: secularization can be shown to be equivalent to setting all terms involving the product $\varphi_a \varphi_b$ to 0, thus removing the off-diagonal blocks of Eq. (13) and the last two elements of the vector \mathbf{f}_0 . This then makes the coherence $F_{ab}(t)$ identically 0. This is as expected for a secular theory: a nonzero detuning $\omega_a \neq \omega_b$ means that the master equation contains no cross terms between modes a and b and thus no coherence arises. Note that the coherence in the secular theory is identically 0, whereas that in the exact result decays to a small value after time $1/(\alpha\Delta^2)$; see Eq. (8). We can thus identify this time scale as the one controlling the secular approximation.

A. Stability of time evolution

The frequently stated reason [23] for secularizing the equation of motion is that it is required to ensure that the equation is of Lindblad form with positive rates, i.e., that the master equation takes the form $\dot{\rho} = -i[H, \rho] + \sum_i \lambda_i (2\hat{\Lambda}_i \rho \hat{\Lambda}_i^\dagger - [\hat{\Lambda}_i^\dagger \hat{\Lambda}_i, \rho]_+)$ with $\lambda_i \geq 0$. This is desired so that Lindblad's theorem can guarantee complete positivity of the density matrix. In addition, negative decay rates may lead to exponentially growing observables. Despite its near-perfect match to the exact solution, our nonsecularized equation

clearly fails these requirements. Equation (10) can be put into Lindblad form by diagonalizing the matrices $L^{\sigma \in \uparrow, \downarrow}$ in Eq. (11), however, the eigenvalues are $\lambda_\sigma = \bar{K}'_\sigma \pm S_\sigma$, where $S_\sigma^2 = (\bar{K}'_\sigma)^2 + |\delta K_\sigma|^2 \geq (\bar{K}'_\sigma)^2$. This means that except when $\delta K_\sigma = 0$, one rate is always negative. Despite this, there have been several recent works [32] which suggest that it is not established that this formal problem leads to any practical difficulties in applying such a theory.

In our problem, we are able to find precise conditions under which the negative rates in the Lindblad form cause a practical problem. Operationally, our problem is to solve the four linear coupled equation for the components F_{ij} . This method will fail if the matrix \mathbf{M} has negative eigenvalues. For a Gaussian problem such as the one we consider here this condition is in fact the only practical consideration; all higher moments factorize by Wick's theorem and so positivity of the eigenvalues of \mathbf{M} ensures that the dynamics remains bounded. Remarkably, the eigenvalues of \mathbf{M} can be found in closed form. They are $\mathbf{M}\phi_i = \mu_i \phi_i$, with

$$\mu_i = \text{Re}[\bar{K}_a + \bar{K}_b] \pm \sqrt{\text{Re}[Q] \pm |Q|}, \quad (14)$$

$$Q = 2\bar{K}_a \bar{K}_b + \frac{1}{2}[\bar{K}_a - \bar{K}_b + i(\omega_a - \omega_b)]^2,$$

where $\bar{K}_i = \varphi_i^2 K_i$. It is clear that when $\omega_a = \omega_b$, one finds $Q = [\bar{K}_a + \bar{K}_b]^2/2$, which means that $\text{Re}[Q] + |Q| = (\text{Re}[\bar{K}_a + \bar{K}_b])^2$. Thus the BR form recovers the fact that there is a zero eigenvalue at degeneracy.

From this closed form we may check that the eigenvalues μ_i remain positive (stable) as long as

$$2\Delta^2 \bar{K}'_a \bar{K}'_b + \Delta(\bar{K}'_a + \bar{K}'_b)(\bar{K}'_a \bar{K}''_b - \bar{K}'_b \bar{K}''_a) > 0. \quad (15)$$

The first term is always positive, and thus instability requires two conditions: First, it requires that $\Delta(\bar{K}'_a \bar{K}''_b - \bar{K}'_b \bar{K}''_a) < 0$, placing a constraint on the frequency dependence of $J(\nu)$; typically an instability is hard to achieve if $J(\nu)$ has only a single peak but is possible for a multi-peaked structure. Second, and more importantly, in order for the second term in Eq. (15) to dominate, $dK(\omega)/d\omega$ must be large enough; this corresponds directly to requiring that the spectral density vary significantly on a scale $J(\omega)$, i.e., that the memory time of the bath is comparable to the damping time scale. If this condition is satisfied, then the Markov approximation is *a priori* invalid.

To summarize, as long as the Markov approximation is valid *a priori*—i.e., the bath memory time is short compared to the damping time—then the eigenvalues of \mathbf{M} are positive and the solution is stable. This result shows that Markovianity is, for this problem, a sufficient condition for stability. This is despite the Lindblad matrices $L^{\uparrow, \downarrow}$ always having negative eigenvalues, except at resonance.

B. Comparison to the exact solution near degeneracy

We have already seen the numerical agreement between this BR treatment and the exact result in Fig. 2. We may note that near resonance one can compare the perturbative solution of the exact problem to a perturbative expansion of the BR eigenvalues. Starting from Eq. (14) and expanding up

to quadratic order in Δ and δK , one finds

$$\mu_0 = \frac{8\varphi_a^2\varphi_b^2\Delta^2\bar{K}'}{(\varphi_a^2+\varphi_b^2)^3|\bar{K}|^2} - \frac{8\varphi_a^2\varphi_b^2\Delta(\bar{K}'\delta K''-\delta K'\bar{K}'')}{(\varphi_a^2+\varphi_b^2)^2|\bar{K}|^2} + O(\Delta^3). \quad (16)$$

Recall that δK depends on the detuning, vanishing at least linearly as $\Delta \rightarrow 0$, so that the second term is at least second order in Δ . This eigenvalue can be compared to the exact perturbative result by referring to Eq. (8) and noting that $\mu_0^{\text{exact}} = -2\text{Im}[\zeta^0]$. The factor of 2 appearing here is because μ corresponds to the eigenvalue of the population equation, whereas the pole in Eq. (8) gives the decay of fields $\hat{\psi}_i$.

Comparing Eq. (8) to Eq. (16) one sees that the leading-order term in $K(\omega)$ is correct, but the second term in Eq. (16) is not there in the exact solution. The second term is, however, dependent on the derivative of the function K . Thus one finds again that the BR theory is correct *as long as the Markovian approximation holds*, i.e., as long as the derivative of the density of states is sufficiently small.

C. Positivity of time evolution

As we have seen, the BR time evolution is stable and has the correct steady state, as long as the Markovian approximation is justified. This rules out the most dramatic pathologies that could arise from the negative rates and suggests that the dynamics will not stray far from the correct behavior. This is consistent with the essentially perfect agreement seen numerically. We now consider a related issue: the extent to which the negative rates lead to unphysical density matrices with negative eigenvalues.

We first summarize some standard definitions [2]. An operator is positive if all its expectation values are positive, and a map is positive if it is between positive operators. Since density operators are positive the exact time-evolution superoperator, which is a map between density matrices, is positive. The secularized master equation in fact satisfies the stronger criterion of complete positivity, which corresponds to positivity in the presence of arbitrary entanglement with an auxiliary system.

The map given by Eq. (10) can be shown to be nonpositive specifically because of the negative eigenvalues of the Kosakowski matrices $L^{\uparrow,\downarrow}$. To demonstrate this we suppose that L^{\downarrow} has a negative eigenvalue and work in its diagonal basis. We denote the field operator corresponding to the unstable (stable) eigenvector by $\hat{\psi}_c$ ($\hat{\psi}_d$), so that there will be terms in Eq. (10) of the form

$$r(2\hat{\psi}_c\rho\hat{\psi}_c^\dagger - [\rho,\hat{\psi}_c^\dagger\hat{\psi}_c]_+), \quad (17)$$

with $r < 0$. In general neither h_{ij} nor L^\uparrow will be diagonal in this eigenbasis of L^\downarrow , so that H contains terms $\hat{\psi}_i^\dagger\hat{\psi}_j$, for all pairs $i, j \in c, d$. Similarly, we will have terms in Eq. (10) from L^\uparrow of the form $2\hat{\psi}_j^\dagger\rho\hat{\psi}_i - [\rho,\hat{\psi}_i\hat{\psi}_j^\dagger]_+$, for all such pairs. However, as positivity requires that all positive operators are mapped to positive operators, showing that it is violated only requires us to construct a single counterexample of a positive operator mapped to a nonpositive operator, and it is possible to do this despite the nondiagonal nature of these other terms. To construct this counterexample we suppose that ρ describes

a pure Fock state in the diagonal basis of L^\downarrow , $\rho = |n, m\rangle\langle n, m|$. This is a positive operator, which is mapped by the first term in Eq. (17) to $2rn|n-1, m\rangle\langle n-1, m|$. Furthermore, we see that no other term in the infinitesimal time-evolution superoperator $\Phi(\rho)$ generates this operator. The Hamiltonian and anticommutator terms in Eq. (10) conserve the total excitation number, while the jump terms from L^\uparrow increase it. Thus $\langle n-1, m|\Phi(\rho)|n-1, m\rangle = 2rn < 0$. Since a positive operator X obeys $\forall|\psi\rangle : \langle\psi|X|\psi\rangle > 0$, this fact proves that the map has taken a positive operator to a nonpositive operator. This proves that the map is not positive. It follows immediately that it is not completely positive. An analogous argument applies for a negative rate in L^\uparrow .

While the BR equation, (10), is not positive, it nonetheless agrees well with the exact solution. This suggests that the operators which are mapped out of the physical space, such as the one constructed above, are absent from, or at least a negligible contribution to, the density matrix. To investigate this and explore the domain of validity of the theory more generally, we consider whether the dynamics is positive for Gaussian states. We consider specifically the subset of Gaussian states relevant to the dynamics above, where the baths and initial conditions are such that $G_{ij} = \langle\hat{\psi}_i\hat{\psi}_j\rangle = 0$.

For Gaussian states the density matrix is positive if the uncertainty principle is satisfied [46], which here is equivalent to F_{ij} being positive semidefinite. This follows upon noting that for two oscillators any normalized linear combination of the operators $\hat{\psi}_a, \hat{\psi}_b$ is a lowering operator $\hat{\eta}$, with corresponding quadratures $\hat{x} = (\hat{\eta} + \hat{\eta}^\dagger)/\sqrt{2}$, $\hat{p} = -i(\hat{\eta} - \hat{\eta}^\dagger)/\sqrt{2}$ and requiring $\Delta x \Delta p \geq 1/2$ for all such quadratures. Positivity of the density matrix can thus be checked numerically by calculating the smallest eigenvalue of F_{ij} . In the BR solution corresponding to Fig. 2(a) we find that there is a brief transient period, up to $t \approx 1$, where the state violates positivity by a tiny amount. Specifically, the smallest eigenvalue of F_{ij} reaches $\lambda_m \sim -10^{-4}$ in this regime, after which it is always positive or 0, with typical values $\lambda_m \sim 0.1$. More generally, the BR λ_m agrees with the exact result to four decimal places. The error is hardly noticeable, except in that it takes the results slightly outside the physical regime at early times.

The behavior discussed above can be understood by deriving the condition under which the BR equation, (10), preserves positivity for Gaussian states. For a time increment Δt the BR equation, (10), implies a shift in the F_{ij} , $F_{ij} \rightarrow F_{ij}^{(0)} + \Delta t R_{ij}$. Since the time evolution of the density matrix is continuous it can only become unphysical if $F_{ij}^{(0)}$ has a zero eigenvalue, which becomes negative under the perturbation R_{ij} . Such an $F_{ij}^{(0)}$ must be of the form

$$\begin{pmatrix} n_a & \sqrt{n_a n_b} e^{i\phi} \\ \sqrt{n_a n_b} e^{-i\phi} & n_b \end{pmatrix}, \quad (18)$$

with $n_a, n_b \geq 0$. From the forms of \mathbf{M} and \mathbf{f}_0 we calculate the shift matrix elements R_{ij} for the state $F_{ij}^{(0)}$. We can then calculate the shift in the zero eigenvalue perturbatively and find it to be negative when

$$\begin{aligned} & \varphi_a^2 K'_{a,\uparrow} n_b + \varphi_b^2 K'_{b,\uparrow} n_a - 2\varphi_a \varphi_b \sqrt{n_a n_b} [\bar{K}'_\uparrow \cos(\phi) \\ & - \delta K''_\uparrow \sin(\phi)] < 0. \end{aligned} \quad (19)$$

This condition gives a range of $n_a - n_b$ and ϕ for which the minimum-uncertainty Gaussian state, Eq. (18), is mapped out of the space of physical states. If the rates are not too different, i.e., the Markov approximation is well satisfied, then this range is small.

In summary, the two-mode BR equation is positive for most Gaussian states. The exceptions are rare, being the subset of minimum-uncertainty states defined by Eq. (19). Since the dissipation drives the system towards safe Gaussian states these dominate the dynamics, even if the others are present under the initial conditions. Indeed, a positivity-violating state is present under the initial condition for Fig. 2, but its effects are transient and quantitatively small.

V. BEYOND THE BLOCH-REDFIELD EQUATION

From the above we may conclude that over a wide range of parameters the Bloch-Redfield theory without secularization accurately matches the exact solution, while secularization reduces the accuracy. This, however, leaves open an alternate question: Does the BR master equation, and the corresponding coupled equations of motion for $F_{ij}(t)$, represent the best possible time-local theory of this problem? In this section we show that a better set of time-local equations exists and involves a minor change to the form of matrix \mathbf{M} that appears in Eq. (13).

A. Sum-rule violation

There are two motivations to suggest that an improved equation is possible. The first is that, as noted above, the BR prediction for the slowest decay rate of coherence, Eq. (16), does not match the rate derived from the poles of the exact solution, Eq. (8). The second reason concerns sum rules as discussed in [37,38]. These state that for operators which commute with the system-bath coupling the time evolution of such operators in the full dynamics should be equal to that in the absence of system-bath coupling. For our model, the operators $\hat{X} = \varphi_b^* \hat{\psi}_a - \varphi_a^* \hat{\psi}_b$ and \hat{X}^\dagger obviously commute with Eq. (1). As such, their time derivatives should be the same as that following from \hat{H}_S alone. In terms of population equations this corresponds to the statement that

$$I \equiv \langle \hat{X}^\dagger \dot{\hat{X}} \rangle = |\varphi_b|^2 F_{aa} + |\varphi_a|^2 F_{bb} - 2\text{Re}[\varphi_a^* \varphi_b F_{ab}]$$

should obey $\partial_t I = \text{Re}[2i(\omega_a - \omega_b)\varphi_a^* \varphi_b F_{ab}]$. In the case where φ_i are real, this means that one should have

$$\begin{pmatrix} \varphi_b^2 \\ \varphi_a^2 \\ -2\varphi_a \varphi_b \\ 0 \end{pmatrix}^T \mathbf{M} = (\omega_a - \omega_b) \begin{pmatrix} 0 \\ 0 \\ 0 \\ 2\varphi_a \varphi_b \end{pmatrix}. \quad (20)$$

One may, however, immediately see that this does not hold for the solution, Eq. (13), of our time-local master equation, unless $K_a = K_b$. We next find an alternative time-local equation of motion for the observables $F_{ij}(t)$ that both satisfies this sum rule and gives the exact eigenvalues near degeneracy.

B. Schrödinger-picture Bloch-Redfield equation

The basis of the alternate approach is to consider the Born approximation for the equation of motion, before making any Markov approximation. We therefore first recall the form of the integrodifferential equation for the density matrix after the Born approximation. In the interaction picture this has the general form

$$\partial_t \rho^{(I)}(t) = \sum_{kl} \int_{kl}^t dt' \eta_{kl}(t-t') [\hat{O}_k(t), [\hat{O}_l(t'), \rho^{(I)}(t')]],$$

where $\hat{O}_k(t)$ is an operator in the interaction picture and $\eta_{kl}(\tau)$ accounts for the system-bath coupling and the integral over the bath density of states. From this one may derive the population equation

$$\partial_t F_{ij} = \sum_{kl} \int_{kl}^t dt' \eta_{kl}(t-t') \langle [[\hat{\psi}_i^\dagger(t) \hat{\psi}_j(t), \hat{O}_k(t)], \hat{O}_l(t')] \rangle_I,$$

where $\langle \dots \rangle_I = \text{Tr}[\dots \rho^{(I)}(t)]$. The BR population equation then follows by assuming that $\rho^{(I)}(t')$ has a slow time dependence and performing the integral over dt' , accounting only for the time dependence of the interaction picture operator $\hat{O}_l(t')$.

If we focus on late times, this procedure is somewhat strange, as it is clear that for a problem which has a time-independent Hamiltonian in the Schrödinger picture it is the density matrix in the Schrödinger picture which will be time independent. As such, an alternate procedure suggests itself: to consider $\rho^{(I)}(t') = e^{i\hat{H}_0 t'} \rho^{(S)} e^{-i\hat{H}_0 t'}$. The explicit time dependence of this density matrix can be eliminated using $\text{Tr}[\hat{O} \rho^{(I)}(t')] = \text{Tr}[e^{-i\hat{H}_0 t'} \hat{O} e^{i\hat{H}_0 t'} \rho^{(S)}]$ so that we have

$$\partial_t F_{ij} = \sum_{kl} \int_0^\infty d\tau \eta_{kl}(\tau) \langle [[\hat{\psi}_i^\dagger(\tau) \hat{\psi}_j(\tau), \hat{O}_k(\tau)], \hat{O}_l] \rangle_S,$$

where $\langle \dots \rangle_S = \text{Tr}[\dots \rho^{(S)}]$ and we have written $\tau = t - t'$. Following this prescription, one can again find an equation for the vector of real quantities \mathbf{f} in the form $\partial_t \mathbf{f} = -\mathbf{M}^S \mathbf{f} + \mathbf{f}_0$, but the matrix \mathbf{M}^S has a different form. The matrix is now given by

$$\mathbf{M}^S = \begin{pmatrix} 2\varphi_a^2 K'_a & 0 & \varphi_a \varphi_b K'_a & \varphi_a \varphi_b K''_a \\ 0 & 2\varphi_b^2 K'_b & \varphi_b \varphi_a K'_b & \varphi_b \varphi_a K''_b \\ 2\varphi_a \varphi_b K'_a & 2\varphi_b \varphi_a K'_b & \Gamma_0^S & -E_0^S \\ -2\varphi_a \varphi_b K''_a & 2\varphi_b \varphi_a K''_b & E_0^S & \Gamma_0^S \end{pmatrix}, \quad (21)$$

where now $E_0^S = (\omega_b - \varphi_b^2 K''_a) - (\omega_a - \varphi_a^2 K''_b)$ and $\Gamma_0^S = \varphi_a^2 K'_b + \varphi_b^2 K'_a$. For want of a better name, we refer to this as the Schrödinger-picture Bloch-Redfield (SpBR) equation. The constant vector \mathbf{f}_0 is unchanged.

The difference between the BR and the SpBR equations has a simple structure: it corresponds to swapping the frequency at which the bath is to be sampled in the third and fourth columns. The origin of this change is the unitary transformation $e^{i\hat{H}_0 t'}$ between the interaction and the Schrödinger pictures, which has the effect of swapping the time dependence of some ‘‘off-diagonal’’ terms. These small changes to matrix \mathbf{M} have several remarkable consequences. First, we may immediately check

that the sum rule as written in Eq. (20) is now exactly satisfied. Second, one may also consider the behavior of the eigenvalues of Eq. (21). Unlike Eq. (13), there is no simple closed-form expression for the eigenvalues in the general case; Eq. (13) was special in having a structure where the secular equation could be written as a quadratic in $\mu_i - \text{Re}[\tilde{K}_a + \tilde{K}_b]$, but this does not hold for Eq. (21). However, one can perform perturbation theory around the point $\Delta = 0$. Clearly the eigenvalues of \mathbf{M} and \mathbf{M}^S match at this point, as the only distinctions occur if $K_a \neq K_b$. Thus, using standard (non-self-adjoint) perturbation theory in terms of the small parameters Δ and $K_a - K_b$ one finds that the lowest SpBR eigenvalue takes the form

$$\mu_0^S = \frac{8\varphi_a^2\varphi_b^2\Delta^2\tilde{K}'}{(\varphi_a^2 + \varphi_b^2)^3|\tilde{K}|^2} + O(\Delta^3). \quad (22)$$

Remarkably, this is identical to the exact solution, further confirming the idea that this SpBR equation is an improvement over the BR population equations discussed previously.

As we have seen above, the BR master equation matches the exact solution well as long as damping is weak enough and the Markov approximation is well justified. The decay rates near resonance have further shown that while the BR master equation is correct to leading order in the damping rate, the SpBR equation is correct to higher order. This suggests that as the damping rate becomes higher, the SpBR may give a better numerical agreement with the exact solution. This is indeed the case and is shown in Fig. 3, where we compare the steady-state values of F_{ij} . Comparing the coherence $F_{ab}(t)$, it is clear that the SpBR matches the exact solution better than the BR approach. The lower panel shows that the two theories give very similar results for the populations. For the parameters corresponding to Fig. 2 the BR and SpBR lines would be indistinguishable.

Note that at $\Delta = 0$, the SpBR and BR formalisms are identical, and so one might expect the results to match at this point. However, the matrices \mathbf{M} are singular at $\Delta = 0$ (as seen earlier from their eigenvalues). As such, the finite population and coherence at $\Delta \rightarrow 0$ correspond to a singular limit.

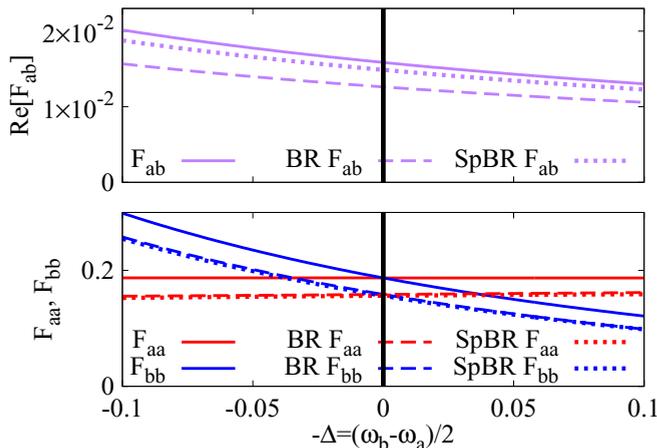


FIG. 3. Comparison of steady-state values of F_{ij} between the exact solution (solid line), BR master equation (dashed line), and SpBR master equation (dotted line), plotted for a higher bath density of states $J_0 = 0.02$, with all other parameters as for Fig. 2.

VI. EXTENSION TO MULTIPLE BATHS

Extending either the exact solution or the BR master equation to multiple baths is simple. For the BR master equation, one just finds a separate set of Lamb-shift terms h_{ij} and dissipator terms L_{ij}^σ for each bath, so that Eq. (10) involves a summation over contributions from the baths. Similarly, the expressions for the matrix \mathbf{M} follow as before, but now with a sum over baths, and even the analytic form of the eigenvalues remains true, with $\tilde{K}_i \mapsto \sum_n \tilde{K}_i^{(n)}$ in Eq. (14). The exact solution is, however, more complicated. Equation (2) still holds, however, there is a sum over baths, and each term now acquires a bath label: $J(\nu), n_B(\nu), W_i(\nu, t) \mapsto J^{(n)}(\nu), n_B^{(n)}(\nu), W_i^{(n)}(\nu, t)$. The last of these quantities now has a more complicated form,

$$W_i^{(n)}(\nu, t) = \int \frac{d\zeta}{2\pi} \frac{e^{-i\zeta t}}{\nu - \zeta - i0} \sum_j \mathcal{G}_{ij}(\zeta) \varphi_j^{(n)}, \quad (23)$$

where the matrix \mathcal{G} can be defined in terms of its inverse, $[\mathcal{G}(\zeta)^{-1}]_{ij} = i\delta_{ij}(\omega_i - \zeta - i0) + \sum_n \varphi_i^{(n)*} \varphi_j^{(n)} K^{(n)*}(\zeta)$.

In the presence of multiple baths, the singular behavior at $\omega_a = \omega_b$ no longer occurs; one may check this by calculating the zeros of $\text{Det}[\mathcal{G}(\zeta)^{-1}]$. One now finds that there is no longer a zero mode, unless the coefficients $\varphi_i^{(n)}$ happen to be parallel for different n . The physical origin of this is that with multiple linearly independent baths there is no longer a linear combination of fields $\hat{\psi}_i$ which decouples from the baths, and so all modes are damped. As such, the collective dephasing model is never correct for predicting the steady-state coherence. The nonsecularized BR approach continues to correctly describe the system as one varies the detuning.

VII. CONCLUSIONS

In conclusion, we have compared the exact and Bloch-Redfield (BR) solutions for a system of two bosonic modes coupled to a common bath. The late-time behaviors show a singular dependence on the detuning: exactly on resonance, significant coherence exists at late times, but for arbitrarily small detuning the coherence drops to a smaller value which depends on the frequency dependence of the density of states. This singular limit appears only at late times, corresponding to a slow decay rate for coherence that vanishes at the degenerate point. All aspects of this behavior are reproduced correctly by a nonsecularized BR theory, whereas secularization leads to incorrect predictions. The BR theory does not guarantee positivity, nonetheless, one can prove that the equations describe bounded dynamics of physical observables, as long as the Markov approximation remains valid. A modification to the BR theory—assuming it is the Schrödinger-picture density matrix that evolves slowly, rather than the interaction-picture one—leads to an improved time-local theory which satisfies the required sum rules and exactly matches the damping rates near resonance.

The research data supporting this publication can be accessed, see Ref. [47].

ACKNOWLEDGMENTS

We are very happy to acknowledge discussions with V. Oganasyan and T. Stace. P.G.K. and J.K. acknowledge financial support from the EPSRC program TOPNES (EP/I031014/1). P.G.K. acknowledges support from the EPSRC (EP/M010910/1). J.K. acknowledges support from

the Leverhulme Trust (IAF-2014-025) and EPSRC program Hybrid Polaritonics (EP/M025330/1). H.M.C. acknowledges support from the EPSRC (EP/G03673X/1). B.W.L. acknowledges support from the Leverhulme Trust (RPG-080) and a joint EPSRC (EP/I035536)/NSF (DMR-1107606) Materials World Network grant.

-
- [1] N. G. van Kampen, *Stochastic Processes in Physics and Chemistry*, 3rd ed. (North-Holland, Amsterdam, 2007).
- [2] H.-P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, Oxford, UK, 2002).
- [3] A. Badolato, K. Hennessy, M. Atatüre, J. Dreiser, E. Hu, P. M. Petroff, and A. Imamoglu, *Science* **308**, 1158 (2005).
- [4] A. J. Ramsay, T. M. Godden, S. J. Boyle, E. M. Gauger, A. Nazir, B. W. Lovett, A. M. Fox, and M. S. Skolnick, *Phys. Rev. Lett.* **105**, 177402 (2010).
- [5] F. Nissen, J. M. Fink, J. A. Mlynek, A. Wallraff, and J. Keeling, *Phys. Rev. Lett.* **110**, 203602 (2013).
- [6] L. Henriët, Z. Ristivojević, P. P. Orth, and K. Le Hur, *Phys. Rev. A* **90**, 023820 (2014).
- [7] P. Kaer, T. R. Nielsen, P. Lodahl, A.-P. Jauho, and J. Mørk, *Phys. Rev. Lett.* **104**, 157401 (2010).
- [8] C. Roy and S. Hughes, *Phys. Rev. Lett.* **106**, 247403 (2011).
- [9] H. Kim, T. C. Shen, K. Roy-Choudhury, G. S. Solomon, and E. Waks, *Phys. Rev. Lett.* **113**, 027403 (2014).
- [10] A. Nysteen, P. Kaer, and J. Mørk, *Phys. Rev. Lett.* **110**, 087401 (2013).
- [11] T. Yuge, K. Kamide, M. Yamaguchi, and T. Ogawa, *J. Phys. Soc. Jpn.* **83**, 123001 (2014).
- [12] S. Diehl, A. Micheli, A. Kantian, B. Kraus, H. P. Büchler, and P. Zoller, *Nat. Phys.* **4**, 878 (2008).
- [13] J. Paavola and S. Maniscalco, *Phys. Rev. A* **82**, 012114 (2010).
- [14] T. V. Tscherbul and P. Brumer, *Phys. Rev. Lett.* **113**, 113601 (2014).
- [15] J. P. Paz and A. J. Roncaglia, *Phys. Rev. Lett.* **100**, 220401 (2008).
- [16] F. Benatti, R. Floreanini, and M. Piani, *Phys. Rev. Lett.* **91**, 070402 (2003).
- [17] D. P. S. McCutcheon, A. Nazir, S. Bose, and A. J. Fisher, *Phys. Rev. A* **80**, 022337 (2009).
- [18] F. Galve, L. A. Pachón, and D. Zueco, *Phys. Rev. Lett.* **105**, 180501 (2010).
- [19] A. F. Estrada and L. A. Pachón, *New J. Phys.* **17**, 033038 (2015).
- [20] P. R. Eastham, A. O. Spracklen, and J. Keeling, *Phys. Rev. B* **87**, 195306 (2013).
- [21] C. Roy and S. Hughes, *Phys. Rev. X* **1**, 021009 (2011).
- [22] F. Benatti and R. Floreanini, *Int. J. Mod. Phys. B* **19**, 3063 (2005).
- [23] R. Dümcke and H. Spohn, *Z. Phys. B* **34**, 419 (1979).
- [24] C. Joshi, P. Öhberg, J. D. Cresser, and E. Andersson, *Phys. Rev. A* **90**, 063815 (2014).
- [25] D. A. Steck, Lecture notes, “Quantum and atom optics” (unpublished).
- [26] F. Benatti, R. Floreanini, and U. Marzolino, *Europhys. Lett.* **88**, 20011 (2009).
- [27] R. Wangsness and F. Bloch, *Phys. Rev.* **89**, 728 (1953).
- [28] A. G. Redfield, *IBM J. Res. Dev.* **1**, 19 (1957).
- [29] F. Bloch, *Phys. Rev.* **105**, 1206 (1957).
- [30] G. Lindblad, *Commun. Math. Phys.* **48**, 119 (1976).
- [31] A. K. Rivas, A. D. K. Plato, S. F. Huelga, and M. B. Plenio, *New J. Phys.* **12**, 113032 (2010).
- [32] J. Jeske, D. Ing, M. B. Plenio, S. F. Huelga, and J. H. Cole, *J. Chem. Phys.* **142**, 064104 (2015).
- [33] R. S. Whitney, *J. Phys. A Math. Theor.* **41**, 175304 (2008).
- [34] S. Maniscalco, *Phys. Rev. A* **75**, 062103 (2007).
- [35] S. Nakajima, *Prog. Theor. Phys.* **20**, 948 (1958).
- [36] R. Zwanzig, *J. Chem. Phys.* **33**, 1338 (1960).
- [37] M. Hell, M. R. Wegewijs, and D. P. DiVincenzo, *Phys. Rev. B* **89**, 195405 (2014).
- [38] J. Salmilehto, P. Solinas, and M. Möttönen, *Phys. Rev. A* **85**, 032110 (2012).
- [39] C. Karlewski and M. Marthaler, *Phys. Rev. B* **90**, 104302 (2014).
- [40] I. L. Aleiner, B. L. Altshuler, and Y. G. Rubo, *Phys. Rev. B* **85**, 121301 (2012).
- [41] S. M. Barnett, J. D. Cresser, and S. Croke, [arXiv:1508.02442](https://arxiv.org/abs/1508.02442).
- [42] H. Grabert, P. Schramm, and G.-L. Ingold, *Phys. Rep.* **168**, 115 (1988).
- [43] C.-H. Chou, T. Yu, and B. L. Hu, *Phys. Rev. E* **77**, 011112 (2008).
- [44] T. M. Stace, A. C. Doherty, and D. J. Reilly, *Phys. Rev. Lett.* **111**, 180602 (2013).
- [45] J. D. Cresser, *J. Mod. Opt.* **39**, 2187 (1992).
- [46] R. Simon, *Phys. Rev. Lett.* **84**, 2726 (2000).
- [47] <http://dx.doi.org/10.17630/e2b9091b-810a-40d4-a7e1-52a2282c2c5e>.