Completely Positive, Accurate Master Equation for Weakly-Damped Quantum Systems Across All Regimes

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Realistic models of quantum systems must include dissipative interactions with an environment. For weakly-damped systems the Lindblad-form Markovian master equation is invaluable for this task due to its tractability and efficiency. This equation only applies, however, when the frequencies of any subset of the system's transitions are either equal (degenerate), or their differences are much greater than the transitions' linewidths (far-detuned). Outside of these two regimes the only available efficient description has been the Bloch-Redfield master equation. This equation has long been the subject of debate because it does not guarantee to preserve the positivity of the density matrix. The ability to efficiently simulate weakly-damped systems across all regimes is becoming increasingly important, especially in the area of quantum technologies. Here we derive a completely positive, Lindblad-form master equation valid for all regimes, and demonstrate its accuracy by comparing it to exact simulations of a variety of systems coupled to an Ohmic bath. The new master equation describes all weakly-damped systems for which, like the Ohmic bath, the spectral density does not vary sharply on the scale of the transitions' Lamb shifts and linewidths. We also show via exact simulations of two examples, that the trivial adiabatic extension of this master equation can accurately describe systems with time-dependent transition frequencies that cross each other during the evolution, so long as the time dependence is not too fast. System identification tools, developed in systems engineering, play an important role in our analysis. We expect that these tools will prove useful in other areas of physics involving complex systems.

I. INTRODUCTION

Weakly damped open systems are important across a wide rang of areas in both physics and chemistry, from quantum thermodynamics [1–3] to the control of chemical reactions [4], to quantum technologies [5–12]. The Lindblad-form Markovian master equations (which we will abbreviate to "Lindblad master equations") are tremendously useful across all these areas because they provide accurate models that avoid the computationally expensive, and often prohibitive, task of simulating the thermal environment [13, 14]. However, recent developments have made it clear that the regime that existing Lindblad master equations cannot describe — the "near degenerate" regime in which non-degenerate transition frequencies are close together — while long ignored, is crucial for investigating important questions in a range of topics, including reservoir engineering and cascaded systems [15–20], adiabatic computation [21, 22], super and sub-radiance [23–26], and "weak lasing" [27, 28], with the possibility that this regime will reveal new tools for controlling quantum systems.

Weakly-damped quantum systems can be divided into three regimes depending on the frequency difference between pairs of transitions. These regimes are *degenerate* (the frequency difference is zero), *non-degenerate* (the frequency difference is much greater than the transitions' linewidths), and *near-degenerate* (everything else). The degenerate and non-degenerate regimes are described, respectively, by two quite different Lindblad master equations [29, 30]. The difference between them is exemplified by the fact that degenerate transitions exhibit super and sub-radiance, whereas non-degenerate transitions do not. These two Lindblad master equations are obtained from the Bloch-Redfield master equation by making the secular (rotating-wave) approximation. However, no Lindblad master equation has been obtained for the near-degenerate regime [31, 32]. Thus to simulate systems in which two or more distinct transitions are separated by less than a few linewidths, one must resort to the Bloch-Redfield master equation [33, 34]. This equation has long been the subject of debate because it is not guaranteed to preserve the positivity of the density matrix [35, 36]. In some subfields (e.g. photo-chemistry [37, 38]), the B-R master equation is used as the standard vehicle for treating weakly-damped systems. Practitioners in other fields, for example quantum optics and many areas of quantum technologies, do not use it because its failure to ensure such a fundamental property as positivity is seen as an indication that it cannot be trusted.

There have been a number of papers, some quite recent, arguing that the B-R equation is a valid and effective model so long as the system is close to Markovian [28, 39, 40]. In particular, Eastham *et al.* [28] considered a model of two coupled linear oscillators that can be solved exactly, and examined how well the B-R equation describes the near-degenerate regime (since this is the regime in which it is needed). They found that the B-R equation was both very accurate and preserved positivity to a very good approximation. They attributed this

to the fact that the dynamics of the coupled oscillators stays close to Markovian, which was in turn due to the relatively slow variation of the spectral density. In [40] Jeske *et al.* also noted that when transitions are close enough that they share the same value of the spectral density, the B-R equation reduces to the degenerate master equation, which is a key element in our analysis here. These recent works raise an interesting question: other authors have assumed that the near-degenerate regime is non-Markovian, due to the apparent lack of a Linblad-form master equation in that regime [31, 32]. If the dynamics of weakly-damped systems is indeed Markovian in the near-degenerate regime, then it is not unreasonable to suggest that there may be a completely positive Markovian master equation that accurately describes it.

Here we show that there is a single, Lindblad-form master equation that describes weakly-damped systems across all three regimes. Deriving this master equation does not require the secular approximation. It accurately models baths with the Ohmic spectrum, and any spectrum that is not highly complex (rapidly varying) on the scale of the Lamb shifts and linewidths (this is true, for example, of all baths consisting of fields in free space). The master equation

1. Provides a completely positive evolution that describes all weakly damped quantum systems for which the bath spectral density does not vary sharply on the scale of the Lamb shifts and linewidths.

2. Is accurate for the above systems in all regimes: degenerate, near-degenerate, and non-degenerate. It describes non-degenerate transitions more accurately than the existing Lindblad master equation for non-degenerate transitions.

3. Shows why the Bloch-Redfield master equation maintains positivity when describing typical weakly-damped systems.

4. When trivially extended to its adiabatic version, accurately models time-dependent systems in which transition frequencies cross, so long as the rate of change of these frequencies is not too fast.

5. Provides insight into the dynamics of the near-degenerate regime.

We will show though numerical simulations that the new master equation is no less accurate than the B-R equation for modeling weakly damped systems that are Markovian (that is, all except those with extremely rapidly varying bath spectral densities), but comes with the guarantee of complete positivity, along with the other benefits that the Lindblad form provides (e.g. the ability to use efficient quantum trajectory methods [41–44]). Further, if the B-R equation looses its validity outside of Markovian systems (something that is indicated in [28]) then our master equation will in fact replace the B-R equation for all weakly damped systems.

We obtain the new master equation in three steps. First, we use exact simulations of a V system coupled to an Ohmic bath, together with the method of system identification, developed in systems engineering, to show that weakly-damped quantum systems with an Ohmic spectrum are not only Markovian but also time-independent across all three regimes. This method also allows us to directly back-out the Lindblad-form equation of motion for this V system. Second, aided by the insights provided in step one, we show how to derive the new Lindblad-form master equation from the Bloch-Redfield equation valid for all regimes and all temperatures. Third, we confirm its accuracy across all regimes for slowly varying spectral densities, using the Ohmic bath as our example, by comparing its predictions to those of exact simulations of two further systems, a trident system and two co-located qubits.

We expect that many important problems involving the near-degenerate regime will also involve transition frequencies that change with time, in such a way that two or more may cross, or nearly cross, during the evolution. Examples of this are the Landau-Zener transition [45, 46] and the control of super- and sub-radiance by shifting energy levels. By simulating two examples, one involving a 4-level system with two transitions, and the other involving two co-located qubits, we show that the adiabatic extension of our master equation is able to accurately describe such time-dependent problems, so long as the rate of change of the transition frequencies is not too fast.

In the next section we perform simulations of a V system with an Ohmic bath, and obtain an accurate master equation for this system for all regimes using system identification. In Section III we use the information obtained in Section II to derive a master equation for all regimes and temperatures given a constraint on the derivative of the spectral density. In Section IV we further confirm the accuracy of the master equation with numerical simulations. In Section V we use numerical simulations to show that the master equation is able to describe time-dependent systems whose levels cross. Section V concludes with a discussion of some open questions.

II. SYSTEM IDENTIFICATION IN THE NEAR-DEGENERATE REGIME

The methods of system identification provide us with a way to determine, from the time series of a linear time-invariant system, the minimal number of variables required to generate this time-series (that is, the dimension of the system), as well as its equations of motion. System identification (SID) methods are typically concerned with input/output systems. SID involves obtaining the outputs of a system for a large enough set of distinct inputs that the equations of motion can be determined. While our system does not have inputs, SID methods are easily adapted to replace the set of inputs with a set of

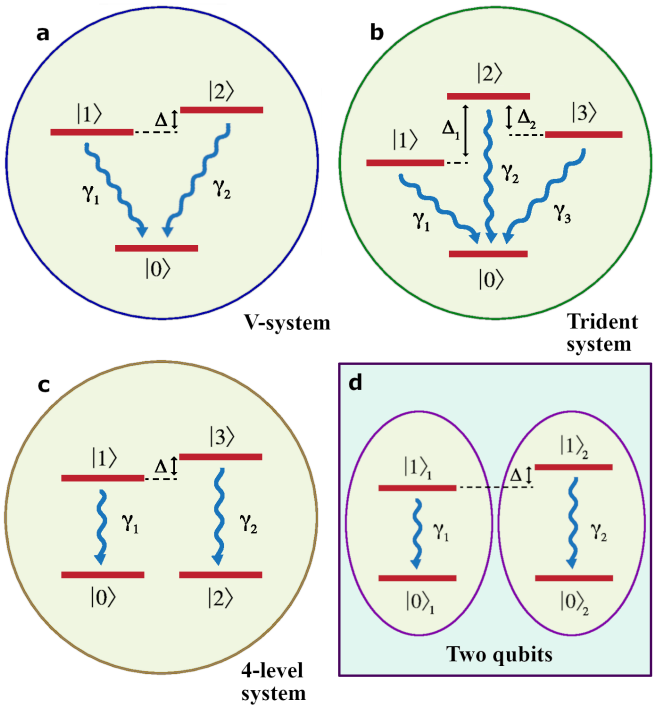

FIG. 1. (Color online) Here we depict four systems with transitions that decay due to a coupling with a thermal bath at zero temperature. The red bars are the energy eigenstates of the system and the blue wiggly lines indicate the transitions. These system are (a) V system, (b) trident system, (c) four-level system, and (d) to co-located qubits. Given that the relative energy of each level in the diagram is indicated by its vertical position, Δ denotes the detuning between the transitions in (a), (c), and (d). In system (b) there are three transitions and thus two independent detunings denoted by Δ_1 and Δ_2. The decay rate of the j^{th} transition is denoted by γ_j. The transition operators and frequencies for each of these systems are given in the supplement [51]

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initial states. (We give the details of the SID method that we use in Appendix A.) Since evolution of the V system is non-trivial only when the upper levels are populated, and the evolution does not generate coherence with the lower level, the two upper populations together with their complex coherence form a closed four-dimensional system. Performing exact simulations of the V system, SID provides us with the dynamics of a fictitious (and possibly larger) system that generates the four-dimensional dynamics. Specifically, if we denote the state of the fictitious system at time t by $\mathbf{v}(t)$, then SID provides us with a matrix $M(\tau)$ where $\mathbf{v}(\tau) = M(\tau)\mathbf{v}(0)$ for a specified time τ. The number of appreciable eigenvalues of M is the effective size of the fictitious system.

Since it is only the ratios between the rate parameters that determine the dynamical behavior (up to a scaling of time) we scale time by a single arbitrary frequency. As a result we specify all frequencies in units of this arbitrary frequency. We perform SID on the V system depicted in Fig.1a with bath cut-off frequency $\Omega = 80\pi$ (details of the bath model are given below in Section III), fix the mean transition frequency $\bar{\omega} \equiv (\omega_1 + \omega_2)/2 = 3\pi$, and choose the coupling constants g_1 and g_2 (defined in Eq.(9)) so as to give the decay rates $\gamma_1 = 0.1$ and $\gamma_2 = 0.05$ [47]. Since we wish to examine the evolution when the detuning, $\Delta\omega \equiv \omega_2 - \omega_1$, is not large compared to the damping rates, we simulate the evolution for the following four values of $\Delta\omega$: 0, $0.28\pi\gamma_1$, $2\pi\gamma_1$, and $4.8\pi\gamma_1$.

To perform the exact simulations we use the method detailed in [13, 14] which employs the matrix-product-state method of Vidal [48, 49]. This in turn requires a split operator method, for which we use a second-order method valid for time-dependent systems, and choose a time-step small enough to obtain an accuracy of about six digits of precision.

Obtaining the matrix M for each value of the detuning, $\Delta\omega$, we find that the largest four eigenvalues of M account for almost all of the dynamical behavior for all four values: the magnitudes of all the rest of the eigenvalues contribute a fraction of less than 3×10^{-4} to the 1-norm of M. This result implies that the dynamics of the system in the near-degenerate regime is both time-independent and Markovian to very good approximation.

A 4-dimensional dynamical model for the four independent variables of the V system can now be obtained merely by taking the log of the matrix $M(t)$ for some appropriate value of t [50]. Writing the four variables as the vector $\mathbf{x}$, the approximate model is $\dot{\mathbf{x}} = D\mathbf{x}$, with $D = \ln[M(t)]/t$. To determine the master equation specified by this model, we need to translate from the elements of D to the familiar terms used to express master equations. The simplest way to do this is to take a general degenerate master equation for a V system and derive its D matrix. The degenerate master equation for a V system (Fig. 1a), in which both transitions have frequency ω, is given by

$$\dot{\rho} = -\frac{i}{\hbar}\left[H_0 + H_{\rm L}, \rho\right] - \mathcal{D}[\Sigma]\rho, \tag{1}$$

where

$$H_0 = \hbar\omega_0(|1\rangle\langle 1| + |2\rangle\langle 2|), \tag{2}$$

$$\Sigma = \sqrt{\gamma_1}\sigma_1 + e^{i\phi}\sqrt{\gamma_2}\sigma_2, \tag{3}$$

with ω_0 the frequency of the both transitions, and $H_{\rm L}$ is the Lamb shift Hamiltonian, given by

$$H_{\rm L} = -\hbar\left[\sum_j \Delta_j\sigma_j^\dagger\sigma_j - \sqrt{\Delta_1\Delta_2}(e^{i\phi}\sigma_1^\dagger\sigma_2 + \text{H.c.})\right]. \tag{4}$$

Here $\sigma_1 = |0\rangle\langle 1|$, $\sigma_2 = |0\rangle\langle 2|$, and ϕ is determined by the phases of the interactions between the transitions and the bath (see Eq.(9)). We also note that $H_{\rm L}$ can be factored as $H_L = -\hbar f(\omega)D^\dagger D$ in which $D = \sqrt{\Delta_1}\sigma_1 + e^{i\phi}\sqrt{\Delta_2}\sigma_2$.

From the derivation of the degenerate master equation [29] (see also Section III) we know that the decay rates γ_j depend on the spectral density of the bath evaluated at their corresponding transition frequencies ω_j.

The Lamb shifts depend both on the damping rates (to which they are proportional) as well as a factor that is an integral of the entire spectral density. Thus if the frequencies of the transitions are changed while leaving the spectral density the same, the decay rates and Lamb shifts also change.

We find that the backed-out model for $\Delta\omega \neq 0$, in which the Hamiltonian is now

$$H_0 = \hbar\omega_0|1\rangle\langle 1| + \hbar(\omega_0 + \Delta\omega)|2\rangle\langle 2|, \tag{5}$$

has exactly the same form as the degenerate master equation. That is, it can be written as Eq.(1) with $H_{\rm L} = \sum_j \zeta_j \sigma_j^\dagger \sigma_j + (\xi\sigma_1^\dagger\sigma_2 + \text{H.c.})$ and $L = \sum_j \eta_j \sigma_j$ for some set of $\{\zeta_j, \xi, \eta_j\}$. This may be considered a little surprising, given that the non-degenerate master equation has no terms in the Lamb shift Hamiltonian proportional to $\sigma_1\sigma_2$. A simple guess for the parameters ζ_j, ξ, and η_j as functions of the Lamb shifts and the decay rates is to take exactly the expression for the degenerate master equation, but to replace $\Delta_2(\omega_0)$ and $\gamma_2(\omega_0)$ by new values implied by the new value of ω_2, namely $\Delta_2(\omega_0 + \Delta\omega)$ and $\gamma_2(\omega_0 + \Delta\omega)$.

We find that this trial master equation does indeed match the model backed out using SID for all three values of $\Delta\omega$. We compare further the evolution predicted by this master equation to the exact evolution in Fig. 2, for a range of values of $\Delta\omega$. For these simulations we use $\omega_0 = 10\pi$ and $\gamma_1(\omega_0) = 2\gamma_2(\omega_0) = 0.1$. In Fig.2c we show a measure of the difference between the evolution given by the master equation and exact simulations for a range of values of the detuning. This measure is an average of the absolute values of the differences between the populations and coherences of the density matrix averaged over time. The measure is below 10^{-3} for all values of the detuning shown.

In Fig.2b we plot the evolution of the populations of the upper levels for both the master equation and the exact simulation for a value of $\Delta\omega$ that might be considered well into the non-degenerate regime ($\Delta\omega = 100\gamma$). We see that the evolution contains "wiggles" that are correctly predicted by our trial master equation, but are not predicted by the non-degenerate master equation. As $\Delta\omega \to \infty$ these wiggles vanish. We also find that in the near-degenerate regime, for which $\Delta\omega \lesssim \Delta_j$, $j = 1, 2$, for the Ohmic bath the values of the detunings and damping rates at $\omega_0 + \Delta\omega$ are hardly changed from their values at ω_0: $\Delta_2(\omega_0 + \Delta\omega) \approx \Delta_2(\omega_0)$ and $\gamma_2(\omega_0 + \Delta\omega) \approx \gamma_2(\omega_0)$. This observation is important in deriving the general master equation, below.

We also compare the evolution of the Bloch-Redfield equation (Eq.(23) below) both to our inferred master equation and the exact simulations. These comparisons, plots of which are given in the supplemental material [51], show that the B-R equation and our master equation have essentially the same accuracy (a result that is implied by the derivation in Section III).

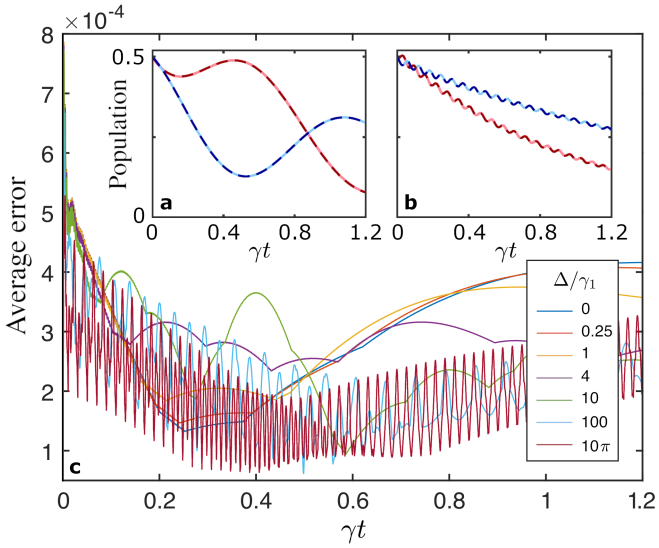

FIG. 2. (Color online) Here we compare the exact evolution of the weakly-damped V system with that predicted by the master equation in Eq.(37). In (a) and (b) we show the populations of levels $|1\rangle$ (red) and $|2\rangle$ (blue) as a function of time with the initial state $|\psi_0\rangle = (|1\rangle + |2\rangle)/\sqrt{2}$. The damping rates are $\gamma_1 = 2\gamma_2 = 0.1$. The solid curves are the exact evolution and the dashed curves are that of the master equation. The detuning is (a) $\Delta\omega = 4\gamma_1$, (b) $\Delta\omega = 100\gamma_1$. In (c), for the values of the detuning shown in the legend, and with the parameters above, we plot a measure of the deviation of the master equation from the exact dynamics as a function of time. This measure is an average taken over the deviations of the four relevant elements of the density matrix: the populations of levels $|1\rangle$ and $|2\rangle$, and the real and imaginary part of the coherence between them. Notably, this average error remains less than 8×10^{-4} for all the values of detuning we explored.

III. DERIVATION OF THE MASTER EQUATION

Having shown numerically that there is a Markovian master equation describing two arbitrarily detuned transitions, at least for the Ohmic bath, as well as obtaining the form that this equation takes, a close examination of the usual derivation of the existing Markovian master equations reveals how this more general master equation can be derived for an arbitrary number of levels. For simplicity we present this derivation first for a bath at zero temperature. We then outline the derivation for non-zero temperature, since it is essentially the same.

The Hamiltonian for the system and the bath, in which the latter consists of a continuum of independent harmonic oscillators, is given by

$$H = H_{\rm sys} + \hbar(A + A^\dagger)\int_0^\Omega \sqrt{J(\omega)}\left[b(\omega) + b^\dagger(\omega)\right] d\omega + \int_0^\Omega \hbar\omega b(\omega)^\dagger b(\omega)\, d\omega \tag{6}$$

Here $H_{\rm sys}$ is the Hamiltonian of the system which has a discrete set of energy levels. The operators $b(\omega)$ are the annihilation operators for a continuum of harmonic oscillators indexed by their frequencies ω (equivalently the modes of a quantum field). The function $J(\omega)$ is the density of oscillators per unit frequency, usually referred to as the *spectral density* of the bath. The maximum frequency of the bath oscillators is Ω and is called the "cut-off" frequency. Instead of having sharp cut-off at frequency Ω one can instead arrange $J(\omega)$ to exhibit a smooth drop-off above some frequency. We use a sharp cut-off purely for simplicity. As will be clear in what follows, in the weak-damping regime the addition of a smooth cut-off merely modifies the values of the damping rates and Lamb shifts.

We have written the Hermitian operator of the system that couples to the bath as $A + A^\dagger$. Here A is defined as containing all the matrix elements of this Hermitian operator that transform higher energy levels to lower ones. If we denote the energy levels of the system by $|n\rangle$, thus writing

$$H_{\rm sys} = \sum_n E_n |n\rangle\langle n|, \tag{7}$$

and define transition (or "decay") operators

$$\sigma_j = |n_j\rangle\langle m_j|, \;\; E_{m_j} > E_{n_j}, \;\; j = 1, \ldots, J, \tag{8}$$

then A can be written as

$$A = \sum_{j=1}^{J} g_j e^{i\phi_j} \sigma_j, \tag{9}$$

where $g_j e^{i\phi_j}$ gives the magnitude and phase of the coupling of transition j to the bath. The frequency of transition j is

$$\omega_j = \frac{E_{m_j} - E_{n_j}}{\hbar} \tag{10}$$

and the evolution of the transition operators σ_j and bath operators $b(\omega)$ in the interaction picture is

$$\sigma_j^{\rm I} = \sigma_j e^{-i\omega_j t}, \tag{11}$$

$$b^{\rm I}(\omega) = b(\omega) e^{-i\omega t}. \tag{12}$$

To derive Markovian master equations one applies a rotating-wave approximation (RWA) to the Hamiltonian above. This should not be confused with a second rotating-wave approximation which is the final step that turns the Bloch-Redfeild master equation into the degenerate and non-degenerate Lindblad master equations. To apply the first RWA we move into the interaction picture. The terms in the interaction Hamiltonian that contain the products $Ab(\omega)$ and $A^\dagger b^\dagger(\omega)$ (the so-called "off-resonant" terms) become

$$H_{\rm OR}^{\rm I} = \hbar \int_0^\Omega \sqrt{J(\omega)} \left[\sum_j g_j \sigma_j b(\omega) e^{-i(\omega_j + \omega)} + {\rm H.c} \right] d\omega. \tag{13}$$

Since the minimum frequency at which each of the terms in the sum over j oscillates is ω_j, when the damping rates and Lamb shifts (to be derived below) are much less than all the ω_j, these terms will average to zero on the timescale of the dynamics induced by the bath, and can be discarded. With this approximation the Hamiltonian of the system and bath becomes

$$H_{\rm RWA} = H_{\rm sys} + \hbar \int_0^\Omega \sqrt{J(\omega)} \left[A^\dagger b(\omega) + A b^\dagger(\omega) \right] d\omega + \int_0^\Omega \hbar\omega b(\omega)^\dagger b(\omega)\, d\omega \tag{14}$$

The regime of weak damping is defined as the regime in which we are close enough to the limit in which $\min_j(\omega_j)/\max_j(\gamma_j) \to \infty$ so that this approximation is a good one.

To proceed one now applies what are known as the Born-Markov approximations to the evolution generated by $H_{\rm RWA}$. For the details of these approximations we refer the reader to [29, 36, 52]). The result is the following expression for the evolution of the density matrix of the system in the interaction picture:

$$\frac{d\rho^{\rm I}}{dt} = -\frac{1}{\hbar^2} \int_0^\infty {\rm Tr}_{\rm B} \left[H_{\rm R}^{\rm I}(t), \left[H_{\rm R}^{\rm I}(s), \rho^{\rm I}(t) \otimes \rho_{\rm B}(0) \right] \right] ds, \tag{15}$$

where

$$H_{\rm R}^{\rm I} = \hbar \int_0^\Omega \sqrt{J(\omega)} \left[\sum_{j=1,2} g_j \sigma_j^{\rm I\dagger} b^{\rm I}(\omega) + {\rm H.c} \right] d\omega \tag{16}$$

is the interaction between the system and the bath that appears in $H_{\rm RWA}$, in the interaction picture. The operator $\rho^{\rm I}(t)$ is the density matrix of the system in the interaction picture, $\rho_{\rm B}(0)$ is the initial density matrix of the bath, and ${\rm Tr}_{\rm B}[\cdot]$ denotes the trace over the bath.

To proceed now we will examine a single term from the expression above, since all the terms are similar and each is processed in the same way. Substituting $H_{\rm R}^{\rm I}(t)$ and $H_{\rm R}^{\rm I}(s)$ into the expression above, one of the terms we obtain is

$$K = \int_0^\infty \left[\int_0^\Omega G(\omega)\, d\omega \right] \sigma_1^{\rm I\dagger}(t) \sigma_2^{\rm I}(s) \rho^{\rm I}(t)\, ds \tag{17}$$

where

$$G(\omega) = e^{-i\omega(t-s)} \int_0^\Omega \left\langle b(\omega) b^\dagger(\omega') \right\rangle \sqrt{J(\omega) J(\omega')}\, d\omega' = J(\omega) e^{-i\omega(t-s)}. \tag{18}$$

Here we have used the relation $[b(\omega), b^\dagger(\omega')] = \delta(\omega - \omega')$ and chosen the field to be at zero temperature so that $\left\langle b^\dagger(\omega') b(\omega) \right\rangle = 0$.

Now substituting $G(\omega)$ into K and rearranging we obtain

$$\begin{aligned} K &= \int_0^\infty \left[\int_0^\Omega G(\omega)\, d\omega\right] \sigma_1^{\mathrm{I}\dagger}(t)\sigma_2^{\mathrm{I}}(s)\rho^{\mathrm{I}}(t)ds \\ &= \left[\int_0^\infty \int_0^\Omega G(\omega-\omega_2)\, d\omega\, ds\right] e^{i(\omega_1-\omega_2)t}\sigma_1^\dagger\sigma_2\rho^{\mathrm{I}}(t) \\ &= \left[\int_0^\Omega \int_0^\infty G(\omega-\omega_2)\, ds\, d\omega\right] \sigma_1^{\mathrm{I}\dagger}(t)\sigma_2^{\mathrm{I}}(t)\rho^{\mathrm{I}}(t). \end{aligned} \tag{19}$$

It is useful to define

$$\Gamma_j \equiv \Gamma(\omega_j) \equiv \int_0^\Omega \int_0^\infty G(\omega-\omega_j)\, ds\, d\omega, \tag{20}$$

$$R_j = \mathrm{Re}[\Gamma_j], \tag{21}$$

$$I_j = \mathrm{Im}[\Gamma_j]. \tag{22}$$

Moving back into the Schrödinger picture, and writing down all the terms we obtain

$$\begin{aligned} \dot\rho = &-\frac{i}{\hbar}[H_{\mathrm{sys}},\rho] + i\sum_j I_j[\Sigma_j^\dagger\Sigma_j,\rho] + \sum_j 2R_j\mathcal{D}[\Sigma_j]\rho \\ &- i\sum_{k\neq j} I_k\left[\Sigma_j^\dagger\Sigma_k\rho - \rho\Sigma_k^\dagger\Sigma_j + \Sigma_j\rho\Sigma_k^\dagger - \Sigma_k\rho\Sigma_j^\dagger\right] \\ &- \sum_{k\neq j} R_k\left[\Sigma_j^\dagger\Sigma_k\rho + \rho\Sigma_k^\dagger\Sigma_j - \Sigma_j\rho\Sigma_k^\dagger - \Sigma_k\rho\Sigma_j^\dagger\right] \end{aligned} \tag{23}$$

where for compactness we have defined

$$\Sigma_j \equiv g_j\sigma_j, \tag{24}$$

and $\mathcal{D}[\cdot]\rho$ is a superoperator defined by

$$\mathcal{D}[c]\rho \equiv \tfrac{1}{2}\left(2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c\right). \tag{25}$$

for an arbitrary operator c,

We note that the decay rates γ_j will be

$$\begin{aligned} \gamma_j &= 2|g_j|^2 R_j \\ &= 2|g_j|^2\int_0^\Omega J(\omega)\left[\int_0^\infty \cos([\omega-\omega_j]s)ds\right]d\omega \\ &= 2|g_j|^2\int_0^\Omega J(\omega)\pi\delta(\omega-\omega_j)d\omega \\ &= 2\pi|g_j|^2 J(\omega_j) \\ &\equiv \gamma(\omega_j). \end{aligned} \tag{26}$$

and the Lamb shifts will be

$$\begin{aligned} \Delta_j &= |g_j|^2\, I_j \\ &= |g_j|^2\int_0^\Omega J(\omega)\left[\int_0^\infty \sin([\omega-\omega_j]s)ds\right]d\omega \\ &= |g_j|^2\,\mathbb{P}\left[\int_0^\Omega J(\omega)\left(\frac{1}{\omega-\omega_j}\right)d\omega\right] \\ &= |g_j|^2\,\mathbb{P}\left[\int_{-\omega_j}^{\Omega-\omega_j}\frac{J(\omega+\omega_j)}{\omega}d\omega\right] \\ &\equiv \Delta(\omega_j). \end{aligned} \tag{27}$$

Here $\mathbb{P}[\cdot]$ denotes the *principle value* of an integral. For readers not familiar with this quantity we give the definition and an example in Appendix B. So long as the spectral density does not decrease with ω, and $\Omega > \omega_j$, the Lamb shift Δ_j can be expected to be greater than the damping rate γ_j (this is true for the Ohmic bath, see below).

The master equation we have derived in Eq.(23), as it stands, includes arbitrary detuning between the levels, but it is not in the Linblad form, and does not guarantee that the density matrix will remain positive. It is, in fact, the Bloch-Redfield master equation. To obtain a master equation in the Lindblad form for all detunings between the transitions, we need to examine how Eq.(23) transforms into the non-degenerate master equation when $\Delta\omega \gg \gamma_j, j = 1, 2$.

Note that when transitions j and k are degenerate, $\Gamma_j = \Gamma_k$ so the last four lines of Eq.(III) collapse into two lines. The terms proportional to I_j become the Lamb shift Hamiltonian, and the terms proportional to R_j combine with the Lindblad term on the first line to give the Lindblad damping term for transitions j and k in the degenerate master equation, namely, $\mathcal{D}[\sqrt{\gamma_j}\Sigma_j + \sqrt{\gamma_k}\Sigma_k]$.

When the two transitions are non-degenerate, then every term in the last four lines will oscillate at the difference frequency $\Delta\omega_{jk} = |\omega_j - \omega_k|$. If this frequency is sufficiently high, then these terms will average to zero and we will be left only with the first two lines, which constitute the non-degenerate master equation.

How large does $\Delta\omega_{jk}$ need to be to eliminate the last four lines of Eq.(23)? It needs to be much larger than the magnitudes of the rest of the dynamical terms in the master equation (excluding the Hamiltonian of the system, since this does not change the populations of the system's eigenstates). The magnitudes of the second and third terms on the top line are Δ_j and γ_j, respectively, and those on the last four lines are

$$M_{jk} \equiv |g_j g_k| R_j \sim \sqrt{\gamma_j\gamma_k}, \tag{28}$$

$$O_{jk} \equiv |g_j g_k| I_j \sim \sqrt{\Delta_j\Delta_k}. \tag{29}$$

So the terms on the second two lines are eliminated when $\Delta\omega_{jk} \gg \max_{kj} M_{jk}$ and $\min_j \Delta\omega_{jk} \gg \max_{kj} O_{jk}$. Without loss of generality we will assume that the Lamb shifts are greater than the damping rates (in the opposite case

one merely switches the roles of O_j and M_j). To find a set of terms that are in the Linblad form, and that are an excellent approximation to the last four lines of Eq.(23), we only need concern ourselves with the regime

$$\Delta\omega_{jk} \lesssim \min_{jk} O_{jk} \tag{30}$$

(since we have assumed $O_{jk} \geq M_{jk}$). Outside of this regime, the last for lines will be eliminated by the oscillations at the detuning $\Delta\omega_{jk}$.

We now recall that γ_j and Δ_j, and therefore M_{jk} and O_{jk}, must be much smaller than both transition frequencies in order for the master equation to be valid. This is a requirement of the initial rotating wave approximation discussed above. Combining this with Eq.(30) above, we only need to consider the regime in which

$$\Delta\omega_{jk} \lesssim L_{jk} \sim \sqrt{\Delta_j \Delta_k} \ll \sqrt{\omega_j \omega_k} \tag{31}$$

Now if Γ_j (and thus R_j and I_j) does not vary rapidly on the scale of $L_{jk} \sim \sqrt{\Delta_j \Delta_k}$ (which is the scale of the Lamb shifts), then in the regime we need to consider we have $\Gamma_j \approx \Gamma_k$. More specifically, we consider systems for which the spectral density satisfies

$$J(\omega_j + \Delta_k) \approx J(\omega_j), \quad \forall j, k. \tag{32}$$

for which the more precise statement is

$$|J(\omega_j + \Delta_k) - J(\omega_j)| \ll J(\omega_j), \quad \forall j, k. \tag{33}$$

since this implies that $\Gamma(\omega)$ also satisfies the same "slow variation" conditions. An interesting question for future work is whether the above slow variation condition is necessary for the system to obey a Markovian master equation.

Given the above condition, we have $I_k \approx I_j$ and $R_k \approx R_j$ when $\Delta\omega_{jk} \lesssim L_{jk}$. Thus, in the regime we need to consider, we can make the replacements

$$R_j \approx \sqrt{R_j R_k}, \tag{34}$$

$$I_j \approx \sqrt{I_j I_k}, \tag{35}$$

in Eq.(23). The result is

$$\begin{aligned}\dot{\rho} = &-\frac{i}{\hbar}[H_{\rm sys}, \rho] + i\sum_j I_j[\Sigma_j^\dagger \Sigma_j, \rho] + \sum_j 2R_j \mathcal{D}[\Sigma_j]\rho \\ &- i\sum_{k\neq j} \sqrt{I_j I_k}\left[\Sigma_j^\dagger \Sigma_k + \Sigma_k^\dagger \Sigma_j, \rho\right] \\ &- 2\sum_{k\neq j} \sqrt{R_j R_k}\left[\Sigma_j^\dagger \Sigma_k \rho + \rho \Sigma_k^\dagger \Sigma_j - 2\Sigma_j \rho \Sigma_k^\dagger\right]\end{aligned} \tag{36}$$

The terms in this equation can be re-factored so as to write it in a much neater form, namely

$$\dot{\rho} = -\frac{i}{\hbar}\left[H_0 - \hbar D^\dagger D, \rho\right] - \mathcal{D}[\Sigma]\rho, \tag{37}$$

This is the zero-temperature Lindblad-form master equation for all regimes. The two operators we have defined are:

$$\Sigma = \sum_{j=1}^N \sqrt{\gamma_j} e^{i\phi_j} \sigma_j, \tag{38}$$

$$D = \sum_{j=1}^N \sqrt{\Delta_j} e^{i\phi_j} \sigma_j. \tag{39}$$

The term $H_{\rm L} \equiv -\hbar D^\dagger D$ is the Lamb-shift Hamiltonian. If we expand it out we see that, so long as $\sigma_j^\dagger \sigma_k \neq 0$, the upper levels of the transitions j and k are coupled together via the bath:

$$\begin{aligned}H_{\rm L} = &-\hbar \sum_{j=1}^N \Delta_j \sigma_j^\dagger \sigma_j \\ &-\hbar \sum_{j=1}^N \sum_{k=j+1}^N \sqrt{\Delta_j \Delta_k}\left(e^{i\Delta\phi_{jk}} \sigma_j^\dagger \sigma_k + \text{H.c.}\right).\end{aligned} \tag{40}$$

The phase factors appearing in $H_{\rm L}$ are

$$\Delta\phi_{jk} = \phi_k - \phi_j, \tag{41}$$

in which g_j and ϕ_j are respectively the strengths and phases of the underlying system-bath interactions, defined in Eq.(9).

The decay rates γ_j are given in Eq.(26) and are determined by the value of the spectral density $J(\omega)$ only at transition frequency ω_j. The Lamb shifts, on the other hand, depend on the whole spectral density, and in particular on the cut-off frequency. As an example, for the Ohmic spectrum with a sharp cut-off at Ω, in which $J(\omega) \propto \omega$ (we choose to define $J(\omega) = \omega/\Omega^2$) the Lamb shifts are

$$\begin{aligned}\Delta_j &= |g_j|^2 \,\mathbb{P}\left[\int_{-\omega_j}^{\Omega-\omega_j} \frac{J(\omega+\omega_j)}{\omega} d\omega\right] \\ &= \frac{|g_j|^2}{\Omega^2}\,\mathbb{P}\left[\int_{-\omega_j}^{\Omega-\omega_j} \frac{(\omega+\omega_j)}{\omega} d\omega\right] \\ &= \frac{\gamma_j}{2\pi}\left[\frac{\Omega}{\omega_j} + \ln\left(\frac{\Omega}{\omega_j} - 1\right)\right].\end{aligned} \tag{42}$$

We see that so long as Ω is larger than $2\omega_j$ the Lamb shift is larger than the damping rate by at least a factor of Ω/ω_j. Recall that the Lamb shifts are required to be much less that the transition frequencies. Using the expression for Δ_j above, we have

$$\begin{aligned}\frac{\Delta_j}{\omega_j} &= \frac{\gamma_j}{2\pi}\left[\frac{\Omega}{\omega_j} + \ln\left(\frac{\Omega}{\omega_j} - 1\right)\right] \\ &\approx \frac{1}{2\pi}\left(\frac{\gamma_j}{\omega_j}\right)\left(\frac{\Omega}{\omega_j}\right)\end{aligned} \tag{43}$$

Thus to satisfy the condition $\Delta_j \ll \omega_j$ requires that the cut-off frequency is not too large. In particular $\Omega \ll 2\pi\omega_j^2/\gamma_j$.

We can now evaluate the fidelity of our approximation explicitly for the Ohmic spectrum. Recall that we require $\Delta_k/\Delta_j \approx 1$, when $\Delta\omega = |\omega_j - \omega_k| \lesssim \Delta_j$. Denoting Δ_j by $\Delta(\omega_j)$, and writing $\omega_k = \omega_j + \Delta_j$, we have

$$\frac{\Delta(\omega_j+\Delta_j)}{\Delta(\omega_j)} - 1 = \frac{\Delta_j}{\Omega}\ln\left(\frac{\Omega}{\omega_j} - 1\right) + \mathcal{O}(\Delta\omega^2) \approx \left[\frac{\Delta_j}{\omega_j}\right]\left[\frac{\omega_j}{\Omega}\ln\left(\frac{\Omega}{\omega_j}\right)\right] \ll 1 \quad (44)$$

Since the master equation is already derived under the conditions that $\Omega \gg \omega_j$, and $\omega_j \gg \Delta_j$, the expressions in *both* of the square brackets are individually much less than unity. Thus the slowly varying Markovian approximation we have introduced is automatically a very good approximation for the Ohmic bath.

Master equation for arbitrary temperature

To derive the master equation for non-zero temperature we merely replace the zero-temperature expectation values of the bath operators with their expectation values at non-zero temperature, which are

$$\left\langle b^\dagger(\omega')b(\omega)\right\rangle = n_T(\omega)\delta(\omega-\omega') \quad (45)$$

$$\left\langle b(\omega')b^\dagger(\omega)\right\rangle = [1+n_T(\omega)]\delta(\omega-\omega') \quad (46)$$

in which

$$n_T(\omega) = \frac{1}{\exp\left[\hbar\omega/\left(k_{\rm B}T\right)\right]-1}. \quad (47)$$

Here T is the temperature of the bath and $k_{\rm B}$ is Boltzmann's constant. With these new expectation values we now obtain more terms in the master equation. For the new terms the spectral density $J(\omega)$ is multiplied by $n_T(\omega)$, so the new terms give a new integral for which we have to calculate the principle value. This integral is

$$\begin{aligned}\mathrm{Im}[\tilde{\Gamma}_j^{\rm T}] &= \mathbb{P}\left[\int_{-\omega_j}^{\Omega-\omega_j}\frac{J(\omega+\omega_j)n_T(\omega+\omega_j)}{\omega}d\omega\right] \\ &= \frac{1}{\Omega^2}\mathbb{P}\left[\int_{-\omega_j}^{\Omega-\omega_j}\frac{1+(\omega_j/\omega)}{\exp[\hbar(\omega+\omega_j)/(k_{\rm B}T)]-1}d\omega\right]\end{aligned} \quad (48)$$

Unfortunately this integral does not have an analytic solution, so we leave it as an integral and define a new set of Lamb shifts

$$\Delta_j^{\rm T} \equiv |g_j|^2\mathrm{Im}[\tilde{\Gamma}_j^{\rm T}]. \quad (49)$$

The approximations we used for the zero temperature part of the master equation can be applied in exactly the same way to the new terms that appear at non-zero temperature. The resulting master equation for arbitrary temperatures is

$$\dot{\rho} = -\frac{i}{\hbar}[H_0 + H_{\rm L}, \rho] - \mathcal{D}[\Theta(T)]\rho - \mathcal{D}[\Upsilon(T)]\rho, \quad (50)$$

where

$$\Theta(T) = \sum_{j=1}^{N}\sqrt{\gamma_j\left[1+n_T(\omega_j)\right]}\,e^{i\phi_j}\sigma_j, \quad (51)$$

$$\Upsilon(T) = \sum_{j=1}^{N}\sqrt{\gamma_j n_T(\omega_j)}\,e^{-i\phi_j}\sigma_j^\dagger \quad (52)$$

and

$$H_{\rm L} = -\hbar\left[B^\dagger B - CC^\dagger\right] \quad (53)$$

with

$$B = \sum_{j=1}^{N}\sqrt{\Delta_j + \Delta_j^{\rm T}}\,e^{i\phi_j}\sigma_j, \quad (54)$$

$$C = \sum_{j=1}^{N}\sqrt{\Delta_j^{\rm T}}\,e^{i\phi_j}\sigma_j. \quad (55)$$

Note that when $T > 0$ the bath induces a Hamiltonian coupling not only between the upper levels of the different transitions, but also the lower levels.

The non-degenerate master equation, the secular approximation, and numerical efficiency

In deriving the master equation valid for all regimes, we did not make the secular approximation, which involves dropping terms that oscillate at the frequency difference between different transitions. However, when the difference between the frequencies of two transitions is much larger than the Lamb shifts and linewidths, keeping the resulting rapidly oscillating terms greatly increases the numerical overhead while contributing little to the evolution. In this case one should drop these terms for numerical efficiency. Doing so transforms the master equation into the non-degenerate master equation, but only for those pairs of transitions for which the detuning is very large. If we write the master equation in the form given in Eq.(36), then dropping the rapidly oscillating terms means merely dropping terms in the second and third lines for the pairs of values of j and k whose transitions are detuned by much more that their Lamb shifts and linewidths.

For readers very familiar with the degenerate and non-degenerate master equations, the result of applying the secular approximation to a subset of pairs of transitions will likely be clear. For readers without this familiarity, we give an explicit example. Let us say that we can divide our transitions into two sets, where the frequencies of those in the first set differ from the frequencies of those in the second set by at least $10^3\Delta_{\rm max}$ in which $\Delta_{\rm max}$ is the maximum Lamb shift among all the transitions. If we denote the transition operators in the first set by $\sigma_j^{(1)}$, with $j = 1, \ldots, N_1$, and those in the second by $\sigma_j^{(2)}$,

with $j = 1, \ldots, N_2$, then the result of making the secular approximation on the master equation in Eq.(37) is

$$\dot{\rho} = -\frac{i}{\hbar}\left[H_0 - \hbar\sum_{m=1}^{2} D_m^\dagger D_m, \rho\right] - \sum_{m=1}^{2}\mathcal{D}[\Sigma_m]\rho, \quad (56)$$

with

$$\Sigma_m = \sum_{j=1}^{N_m}\sqrt{\gamma_j^{(m)}}e^{i\phi_j^{(m)}}\sigma_j^{(m)}, \quad (57)$$

$$D_m = \sum_{j=1}^{N_m}\sqrt{\Delta_j^{(m)}}e^{i\phi_j^{(m)}}\sigma_j^{(m)}. \quad (58)$$

IV. ACCURACY OF THE MASTER EQUATION: NUMERICAL EXAMPLES

It is clear from the derivation in the previous section that the master equation we have obtained, given in Eqs.(37) and (50), will be valid so long as the variation of the Lamb shifts $\Delta(\omega_j)$ and damping rates $\gamma(\omega_j)$ on the scale of these same Lamb shifts and damping rates is sufficiently small. This variation of the Lamb shifts and damping rates will be small if the variation of the spectral density $J(\omega)$ on the scale of the Lamb shifts and damping rates is sufficiently small. Below we will verify quantitatively the accuracy of the new master equation for the Ohmic spectrum at zero temperature, using exact simulations of two example systems. The Ohmic spectrum is appropriate for systems such as atoms, color centers, or superconducting qubits coupled to one-dimensional wave-guides or transmission lines.

Since the efficacy of the approximation used to derive the master equation does not depend on the temperature of the bath or the specific functional form of the spectral density (it depends only on the local variation of the resulting Lamb shifts and damping rates around their respective transition frequencies), simulations for the Ohmic bath at zero temperature provide a high level of confidence in the accuracy of the master equation more generally. Further, since we have exact analytic expressions for the Lamb shifts and damping rates in this case, if desired the variations of these quantities at the transition frequencies can be related directly to the accuracy determined in our simulations.

We have already compared the evolution of the master equation to exact simulations for the V system in Fig. 2. We now consider two further systems. The first is the "trident" system depicted in Fig.1b. This system has three transitions, and thus also three pairs of transitions which we can place simultaneously in the near-degenerate regime. We choose the parameters $\omega_1 = 10\pi$, $\omega_j = \omega_1 + \gamma_2/(j-1)$, for $j = 2, 3$, and $\gamma_j = (5-j)/40$, for $j = 1, 2, 3$, with the cut-off frequency $\Omega = 80\pi$. Choosing the initial state $|\psi_0\rangle = (7i|1\rangle + 3|2\rangle)/\sqrt{58}$, we plot the evolution of the populations predicted by the master equation along with the exact evolution in Fig. 3a. The maximum error in the evolution of the master equation over the time period plotted in Fig. 3a is less than 2×10^{-3}.

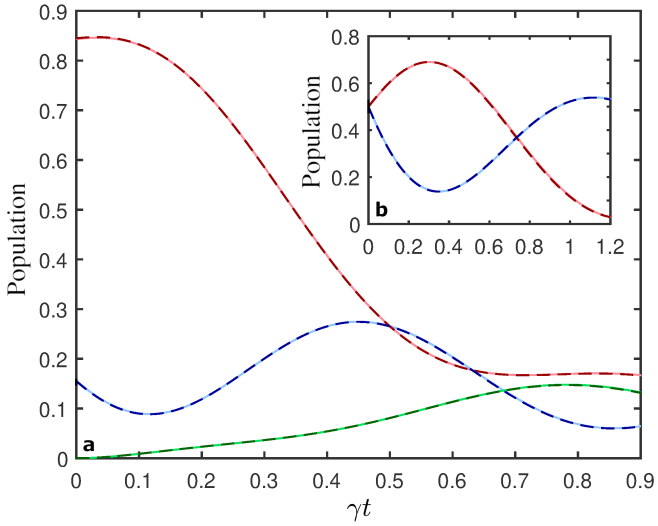

FIG. 3. (Color online) A comparison of the master equation given in Eq.(37) with the exact evolution for two open systems. The evolution of the master equation is shown as dashed lines and the exact evolution as solid lines. (a) The populations of the three upper levels of the trident system depicted in Fig.1b with initial state $|\psi_0\rangle = (7i|1\rangle + 3|2\rangle)/\sqrt{58}$. (b) The populations of the two upper levels of the two-qubit system depicted in Fig.1d with the initial state $|\psi_0\rangle = (i|1\rangle_1|0\rangle_2 + |0\rangle_1|1\rangle_2)/\sqrt{2}$ and the parameters $\gamma_1 = \gamma_2 = 0.1$, $\omega_1 = 10\pi$, and $\omega_2 = \omega_1 + 2\gamma$.

We now perform simulations for two co-located qubits, whose level structure is depicted in Fig. 1d. We choose the parameters $\omega_1 = 10\pi$, and $\omega_2 = \omega_1 + 2\gamma$, $\gamma_1 = \gamma_2 = 0.1$, with the same cut-off frequency as before. We find that this system requires significantly larger values of the weak damping parameters ("quality factors"), $Q_j \equiv \omega_j/\gamma_j$, in order for the master equation to accurately model the dynamics. Since available numerical resources place restrictions on the sizes of the Q_j's that we can practically simulate, for this system we apply the *first* rotating-wave approximation to our model Hamiltonian prior to performing the exact simulations. That is, we simulate the Hamiltonian $H_{\rm RWA}$ (Eq.(14)) instead of the full model in Eq.(6). These simulations thus show us how well the master equation will perform so long as the Q_j's are large enough to satisfy the first rotating-wave approximation. We stress that the values of the Q_j's we actually simulate here are not large enough to satisfy this approximation for this system. This fact is interesting in itself, because it shows that different systems, even with only a few levels, can require quite different quality factors to reach the weak damping regime. We believe this is due to the availability of more channels via which the off-resonant terms in the system bath interaction can excite the two-qubit system over the V and trident systems.

Choosing the initial state $|\psi_0\rangle = (i|1\rangle_1|0\rangle_2 +$

$|0\rangle_1|1\rangle_2)/\sqrt{2}$, we show the evolution of the populations for both the master equation and the exact simulations of $H_{\rm RWA}$ in Fig. 3b. The error in the evolution of the master equation over the duration shown in Fig. 3a is less than 5×10^{-3}.

V. TIME-DEPENDENT PROBLEMS: ACCURACY OF THE ADIABATIC EXTENSION

Many important problems involve open systems whose Hamiltonians change with time. Master equations are able to describe these systems if the time-dependence is not too fast. To do so one takes the master equation for a time-independent system and changes the parameters and operators that appear in it, namely $\gamma_j(\omega_j)$, $\Delta_j(\omega_j)$, and σ_j (which depend on the system eigenstates and thus on the system Hamiltonian), so that at each time they take the values corresponding to the Hamiltonian of the system at that time. The resulting time-dependent "adiabatic" master equation will be effective for sufficiently slow changes in the Hamiltonian.

Here we examine some examples to confirm that the adiabatic version of the master equation is accurate even when two levels of an open system cross each other, or move from degenerate to near-degenerate, during the evolution. We consider first the V system in which both damping rates are equal and the detuning changes with time. We start the system in the state $|\psi_-\rangle \equiv (|1\rangle - |2\rangle)/\sqrt{2}$, which for $\Delta\omega = 0$ will not decay since it is the (sub-radiant) dark state [15, 25]. We then change the detuning with time as determined by following two functions:

$$F_1(t) = \begin{cases} 0, & 0 < t < \frac{T}{4}, \\ \frac{\pi}{64}\left(t - \frac{T}{4}\right), & \frac{T}{4} < t < T, \end{cases} \tag{59}$$

$$F_2(t) = \begin{cases} 0, & 0 < t < \frac{T}{4}, \\ \frac{\pi}{2}, & \frac{T}{4} < t < \frac{3T}{4}, \\ 0, & \frac{3T}{4} < t < T. \end{cases} \tag{60}$$

The function F_1 is chosen so that the detuning increases gradually, while F_2 involves rapid changes. In Fig.4 we compare the adiabatic version of the master equation with the exact evolution for the two cases. For $\Delta\omega(t) = F_1(t)$ the maximum error of the adiabatic master equation is 3.4×10^{-3}, and for $\Delta\omega(t) = F_2(t)$ the maximum error is 1.5×10^{-2}.

As our final example we consider a generalized version of the Landau-Zener transition [45, 46], in which the energies of two coupled levels cross each other. In particular we use the 4-level system depicted in Fig.1c, in which we add a coupling between the upper two levels. In the original Landua-Zener transition, for which there is an analytic solution, the energy of one of the levels is fixed and the other increases linearly with time. We generalize this by choosing the following sinusoidal time-dependence for ω_1:

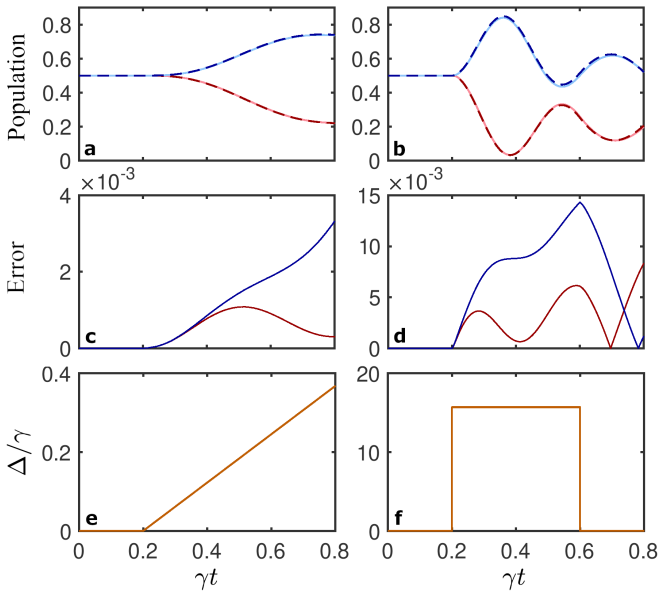

FIG. 4. (Color online) A comparison of the evolution predicted by the adiabatic extension of the new master equation, Eq.(37), with an exact simulation of a V system coupled to an Ohmic bath (Fig.1a), in which the detuning between the transitions, $\Delta\omega$, has the time-dependence given in Eqs.(59) and (60). The initial state is $|\psi_-\rangle = (|1\rangle - |2\rangle)/\sqrt{2}$ which is the dark state for degenerate transitions with equal damping rates. The transition frequency $\omega_1 = 3\pi$, the damping rates are $\gamma_1 = \gamma_2 = \gamma = 0.1$, and the simulation time is $T = 8$. (a,c,e) $\Delta\omega(t) = F_1(t)$ (Eq.(59)). a) The populations of levels $|1\rangle$ (blue) and $|2\rangle$ (red), with the evolution of the adiabatic master equation denoted by dashed lines and that of the exact simulation by solid lines. (c) The absolute value of the difference between the populations predicted by the adiabatic master equation and the exact evolution. (e) The detuning as a function of time. (b,d,f) The same set of results but with the detuning given by $F_2(t)$ (Eq.(60)).

$$\omega_1(t) = \omega_2 - \Delta_0 \cos(\nu t), \quad 0 \le t \le \tfrac{\pi}{\nu}. \tag{61}$$

The detuning, $\Delta\omega \equiv \omega_1 - \omega_2$, starts at $-\Delta_0$ and increases as a sinusoid through zero to end at Δ_0. We plot the evolution of the populations of the two levels in Fig.5. The maximum error of the adiabatic extension of the master equation is less than 5.4×10^{-3}.

DISCUSSION

We have shown that so long as the spectral density, and thus the Lamb shifts and the linewidths of an open system do not vary sharply on the scale of these same Lamb shifts and linewidths, there is a single Lindblad-form master equation that accurately describes the system across all regimes. This shows why the Bloch-Redfield master equation produces valid results for such systems,

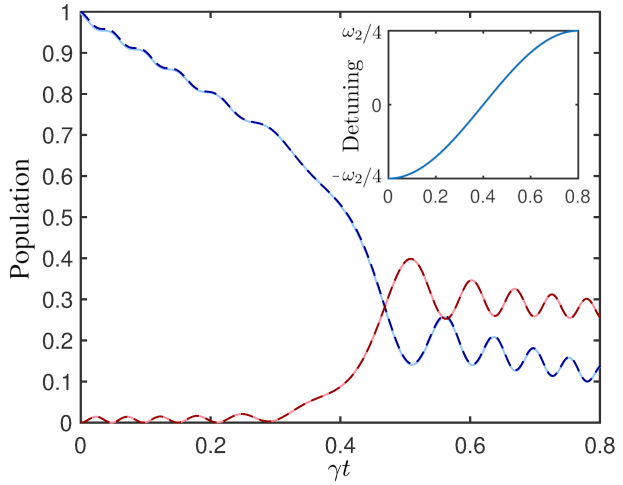

FIG. 5. (Color online) Here we plot the evolution resulting from a generalized Landau-Zener transition in which the energies of two coupled levels cross. The levels are the upper two levels of the four-level system depicted in Fig.1c. The energy of level $|2\rangle$ is fixed so that $\omega_2 = 2\pi$, and that of level $|1\rangle$, in which all the population starts, increases with time. The detuning between the levels is shown as a function of time in the inset. We plot the populations of the two levels, both the exact evolution (solid) and that predicted by the master equation (dashed). The damping rates of the two levels are $\gamma_2 = 2\gamma_1 = 0.05$, the coupling between them is $c = 0.2$, the initial detuning is $\Delta_0 = \pi/2$, and the period of the sinusoid is $2\pi/\nu = 64$.

and clarifies previous assertions that open systems are Markovian in this regime.

The new master equation provides a powerful tool for gaining analytical insights into the behaviour of open systems in the near-degenerate regime, as well as for simulating these systems. It also provides a powerful tool for simulating systems in which transitions are time-dependent and cross during the evolution, so long as this time-dependence is not too fast. This suggests that further exploration of the accuracy of the adiabatic extension of the master equation as a function of the speed of the time-dependence may be a worthwhile endeavor. Such an exploration would help to delineate the class of controlled systems for which it is effective.

It is an interesting question as to whether the Bloch-Redfield equation is able to describe any weakly-damped open systems that the master equation derived here cannot. That is, whether the B-R equation provides a useful model of systems for which the spectral density varies significantly on the scale of the Lamb-shifts and linewidths. If the answer turns out to be no, as is suggested by previous investigations [28, 40], then the master equation we have presented here can be viewed as a completely positive replacement for the B-R equation for weakly damped systems.

The paradigm of system identification played an important role in obtaining the master equation. As far as we are aware, system identification has not been used before as a tool to understand the dynamics of open quantum systems, or emergent phenomena more generally. We expect that it will prove to be powerful for exploring a wide range of problems in open systems and many-body physics.

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Appendix A: System identification for linear systems

Here we present the method we used to determine the minimal model of a linear system given a knowledge of the evolution of a subset of the system's state-space. In our case the linear system consists of a low-dimensional quantum system interacting with a high-dimensional bath, and the subset of the state space that we can observe is the density matrix of the low-dimensional system. The following method is one of a family of elegant methods referred to as *subspace identification methods*, adapted so as to use a set of initial conditions rather than a set of inputs. Further information on subspace identification methods can be found in [54–57].

Let us say we have a high dimensional system with dimension J (in our case the open system and the bath), and we have the ability to observe $N < J$ variables of the system, as well as to evolve the system with any choice of initial conditions for the N variables we can observe. We would like to find an accurate model (another linear system) that generates the evolution of the N variables but is only M dimensional with $N \leq M < J$. Let us denote a state of the full J dimensional system by the vector $\mathbf{v}$, and the subset of N variables in which we are interested by the N-dimensional vector $\mathbf{x}$. The map that gives the state of the total J-dimensional system at time τ given an initial state $\mathbf{v}(0)$ we will call $Z(\tau)$ so that $\mathbf{v}(\tau) = Z(\tau)\mathbf{v}(0)$. Defining $Z \equiv Z(\tau)$ we note that $Z(n\tau) = Z^n$. We also define the non-square projector P that projects onto the N variables so that $\mathbf{x} = P\mathbf{v}$.

Given the ability to evolve the total system with any choice of initial conditions for the N-dimensional subsystem, along with a single choice for the initial values of the rest of the variables (of which there are $N - J$), we can obtain the matrices $Y_n = PZ(n\tau)P^{\mathrm{T}}$ that maps the N variables at time 0 to their values at time τ, for any time τ.

We now construct the following two symmetric "block Hankel" matrices:

$$H_0 \equiv \begin{pmatrix} Y_0 & Y_1 & \cdots & Y_n \\ Y_1 & Y_2 & & Y_{n+1} \\ \vdots & & \ddots & \vdots \\ Y_n & Y_{n+1} & \cdots & Y_{2n} \end{pmatrix}, \tag{A1}$$

$$H_1 \equiv \begin{pmatrix} Y_1 & Y_2 & \cdots & Y_{n+1} \\ Y_2 & Y_3 & & Y_{n+2} \\ \vdots & & \ddots & \vdots \\ Y_{n+1} & Y_{n+2} & \cdots & Y_{2n+1} \end{pmatrix}. \tag{A2}$$

We now note that H_0 can be written as an outer product $H_0 = CD^{\mathrm{T}}$ of (non-square) matrices given by

$$C = \begin{pmatrix} P \\ PZ \\ \vdots \\ PZ^m \end{pmatrix} = P \begin{pmatrix} I \\ Z \\ \vdots \\ Z^m \end{pmatrix}, \tag{A3}$$

$$D^{\mathrm{T}} = \begin{pmatrix} Z & Z^2 & \cdots & Z^m \end{pmatrix} P^{\mathrm{T}}. \tag{A4}$$

We can now determine C and D by doing a singular value decomposition of H_0 to give $H_0 = USV^{\mathrm{T}}$. Note that since the smaller dimension of the matrices C and D is smaller than that of H_0 we expect many of the columns of U and the rows of V^{T} will be zero, as will many of the eigenvalues of H_0 which are given in the diagonal matrix S. Note that we can now view P and Z as defining a linear model that generates evolution of the N-dimensional subsystem. The number of eigenvalues that are appreciably non-zero tells us the dimension of the model.

To distinguish the model from the original total system we started with, we can write the matrices C and D as

$$C^{\mathrm{T}} = \begin{pmatrix} M^{\mathrm{T}} & [M^{\mathrm{T}}]^2 & \cdots & [M^{\mathrm{T}}]^m \end{pmatrix} Q^{\mathrm{T}}, \tag{A5}$$

$$D^{\mathrm{T}} = \begin{pmatrix} M & M^2 & \cdots & M^m \end{pmatrix} Q^{\mathrm{T}}. \tag{A6}$$

where M is the evolutionary map for the model and Q is the projector onto the subsystem. Let us now decompose $H_0 = USV^{\mathrm{T}}$ into the outer product of two vectors $\tilde{C} \equiv U\sqrt{S}P^{\mathrm{T}}$ and $\tilde{D} = (P\sqrt{S}V^{\mathrm{T}})^{\mathrm{T}}$. Noting that

$$H_1 = CZD^{\mathrm{T}} \tag{A7}$$

we can obtain the evolutionary map for the model, M, from H_1 using

$$M = (\tilde{C}\tilde{C}^{\mathrm{T}})^{-1}\tilde{C}^{\mathrm{T}}H_1\tilde{D}(\tilde{D}^{\mathrm{T}}\tilde{D})^{-1}. \tag{A8}$$

Appendix B: Principle value of an integral

In deriving the master equation in Section III we used the fact that

$$\int F(x)\left[\int_0^\infty \sin([\omega-\omega_0]s)ds\right]dx = \mathbb{P}\left[\int \frac{F(x)}{x}dx\right] \tag{B1}$$

for any smooth function $F(x)$, in which $\mathbb{P}[\cdots]$ denotes the *principle value* of a divergent integral. The principle value of an integral that diverges at a point a (where $a \in (b,c)$), is defined by

$$\mathbb{P}\left[\int_b^c f(x)dx\right] \equiv \lim_{\varepsilon\to 0}\left[\int_b^{a-\varepsilon} f(x)dx + \int_{a+\varepsilon}^c f(x)dx\right]. \tag{B2}$$

Since the divergent function $f(x) = 1/x$ is anti-symmetric and diverges at $a = 0$, it is simple to evaluate the principle value of $\int_{-b}^c (1/x)dx$. Assuming that b and c are positive and $c > b$ we have

$$\begin{aligned}\mathbb{P}\left[\int_{-b}^c \frac{dx}{x}\right] &\equiv \lim_{\varepsilon\to 0}\left[\int_{-b}^{-\varepsilon}\frac{dx}{x} + \int_\varepsilon^c \frac{dx}{x}\right] \\ &= \lim_{\varepsilon\to 0}\left[\int_{-b}^{-\varepsilon}\frac{dx}{x} + \int_\varepsilon^b \frac{dx}{x}\right] + \int_b^c \frac{dx}{x} \\ &= \int_b^c \frac{dx}{x} = \ln(c/b).\end{aligned} \tag{B3}$$

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