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Constraints on relaxation rates for *N*-level quantum systems

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We study the constraints imposed on the population and phase relaxation rates by the physical requirement of completely positive evolution for open *N*-level systems. The Lindblad operators that govern the evolution of the system are expressed in terms of observable relaxation rates, explicit formulas for the decoherence rates due to population relaxation are derived, and it is shown that there are additional, nontrivial constraints on the pure dephasing rates for $N>2$. Explicit, experimentally testable inequality constraints for the decoherence rates are derived for three- and four-level systems, and the implications of the results are discussed for generic ladder, Λ, and V systems and transitions between degenerate energy levels.

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I. INTRODUCTION

Understanding the dynamics of open systems is crucial in many areas of physics including quantum optics [1,2], quantum measurement theory [3], quantum state diffusion [4], quantum chaos [5], quantum-information processing [6], and quantum control [7–9]. Yet, despite much effort to shed light on these issues [3,5,10–12], many important questions remain.

For instance, it was recognized early by Kraus [13], Lindblad [14,15], and Gorini, Kossakowki, and Sudarshan [16] that the dynamical evolution of an open system must be completely positive[1] to ensure that the state of the open system remains physically valid at all times. Unfortunately, if relaxation rates are introduced *ad hoc* based on a phenomenological description of the system, the resulting equations often do not satisfy this condition. For example, the Agarwal-Redfield equations of motion for a damped harmonic oscillator have been shown to violate complete and even simple positivity for certain initial conditions [17]. Although such master equations may provide physical solutions in some cases, serious inconsistencies such as negative or imaginary probabilities, unbounded solutions, and other problems may arise.

For two-level systems the implications of the complete positivity requirement have been studied extensively in the literature, for example by Gorini *et al.*, who first showed that there are constraints on the relaxation rates in the weak coupling limit [16,18], Alicki and Lendi who provided a comprehensive in-depth analysis of the dissipative optical Bloch equations [12], and more recently Kimura who extended earlier work by Gorini *et al.* to the strong coupling regime [19]. Recently, there has also been considerable research activity on quantum Markov channels for two-level systems, motivated by their importance in quantum computing and communication. See, for instance, Ref. [20] for a comprehensive analysis. A few simple higher-dimensional systems such as a three-level V system with decay from two upper levels to a common ground state have also been studied, for instance by Alicki and Lendi [12].

In general, however, ensuring complete positivity of the evolution is often neglected for open systems with more than two (or degenerate) energy levels. For instance, the general expressions (6.A.11) in [21] for the relaxation rates ensure complete positivity only for two-level systems, as we shall show. For higher-dimensional systems additional constraints must be imposed if complete positivity is to be maintained. One reason for this neglect of positivity constraints is that, although the general form of the admissible generators for quantum dynamical semigroups is known, it can be difficult to verify whether a proposed dynamical law for an open system is consistent with positivity requirements. The main objective of this paper is to address this issue.

The paper is organized as follows. Starting with a purely phenomenological description of the interaction of an open system with its environment in terms of observable population and phase relaxation rates—analogous to the T_1 and T_2 relaxation times for a two-level system—we derive a general form for the dissipation superoperator in Sec. II. We then explicitly demonstrate with simple examples in Sec. III that the relaxation rates cannot be chosen arbitrarily if the evolution of the system is to be physical in the sense that it satisfies complete positivity. In particular, we show that the phase relaxation rates for $N>2$ are correlated even in the absence of population relaxation, i.e., there exist constraints on the phase relaxation rates that are independent of population decay. To understand the nature of these constraints we express the empirically derived relaxation superoperator in Lindblad

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[1]A map Λ acting on the algebra of bounded operators $B(\mathcal{H})$ on a finite-dimensional Hilbert space $\mathcal{H}$ is completely positive if the composite map $\Lambda\otimes I_n$ acting on $B(\mathcal{H})\otimes M(n)$, where $M(n)$ is algebra of complex $n\times n$ matrices and I_n the identity in $M(n)$, is positive for any $n\geqslant 0$.

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form (Sec. IV), and show that it can always be decomposed into two parts, one accounting for population relaxation and the other for pure phase relaxation processes (Sec. V). This decomposition provides a general formula for the decoherence rates induced by population decay, which is consistent with physical expectations and evidence, and *additional* positivity constraints on the decoherence rates resulting from pure phase relaxation for $N>2$.

In Sec. VI we study the implications of these additional constraints in depth for three- and four-level systems. In particular, we use the abstract positivity constraints to derive explicit inequality constraints for the observable decoherence rates. Such explicit constraints are important from a theoretical and practical perspective because they allow us to make concrete, empirically verifiable predictions about the decoherence rates and the dynamics of the system, as we show in Sec. VII for several common, generic three- and four-level systems such as ladder, Λ, V, and tripod systems, and transitions between doubly degenerate energy levels. Experimental data consistent with positivity constraints would be significant and would validate the chosen model for the open system dynamics. On the other hand, if the observed relaxation rates for a system do not satisfy the constraints required to ensure complete positivity, it would be a strong indication that the model used is not sufficient to properly describe the dynamics of the system. This does not necessarily mean that the model is useless; it might well be adequate for some purposes, but there will be cases where the model makes unphysical predictions and better models, consistent with physical constraints, are required.

II. QUANTUM LIOUVILLE EQUATION FOR DISSIPATIVE SYSTEMS

The state of an N-level quantum system is usually represented by a density operator ρ acting on a Hilbert space $\mathcal{H}$. If the system is closed then its evolution is given by the quantum Liouville equation

$$i\hbar\frac{d}{dt}\rho(t) = [\![H,\rho(t)]\!], \tag{1}$$

where H is the Hamiltonian. Formally, the dynamics of an open system S that is part of a closed supersystem $S+E$ (possibly the entire universe) is determined by the Hamiltonian dynamics (1) of $S+E$, and the state of the subsystem S can be obtained by taking the partial trace of the entire system's density operator ρ_{S+E} over the degrees of freedom of the environment E. Often, however, the evolution of the (closed) supersystem is unknown or too complicated, and we are interested only in the dynamics of S. It is therefore useful to define a density operator ρ based on the degrees of freedom of S, and describe its nonunitary evolution by amending the quantum Liouville equation to account for the non-Hamiltonian dynamics resulting from the interaction of S with the environment E.

In this paper we restrict our attention to the (common) case where the effect of the environment E leads to population and phase relaxation (decay and decoherence, respectively) of the system S, and ultimately causes it to relax to an equilibrium state. To clearly define what we mean by the terms population and phase relaxation, note that given an N-dimensional quantum system we can choose a complete orthonormal basis $\{|n\rangle : n=1,2,\ldots,N\}$ for its Hilbert space and expand its density operator with respect to this basis:

$$\rho = \sum_{n=1}^{N}\left[\rho_{nn}|n\rangle\langle n| + \sum_{n'>n}\rho_{nn'}|n\rangle\langle n'| + \rho_{nn'}^{*}|n'\rangle\langle n|\right]. \tag{2}$$

Although we can theoretically choose any Hilbert space basis, physically there is usually a preferred basis. Since the interaction with the environment usually causes the system to relax to an equilibrium state that is a statistical mixture of its energy eigenstates,[2] it is sensible to choose a suitable basis of (energy) eigenstates of the system for modeling the relaxation process. In this setting the diagonal elements ρ_{nn} in the expansion Eq. (2) of ρ determine the populations of the (energy) eigenstates $|n\rangle$, and the off-diagonal elements $\rho_{nn'}$ $(n\neq n')$ are called coherences, since they distinguish coherent superpositions of energy eigenstates $|\Psi\rangle = \Sigma_{n=1}^{N} c_n|n\rangle$ from statistical (incoherent) mixtures of energy eigenstates $\rho=\Sigma_{n=1}^{N} w_n|n\rangle\langle n|$.

Population relaxation occurs when the populations of the energy eigenstates change, typically due to spontaneous emission or absorption of quanta of energy at random times. To account for population relaxation as a result of the interaction with an environment we must modify the system's quantum Liouville equation (1) to

$$\dot{\rho}_{nn}(t) = -\frac{i}{\hbar}([\![H,\rho(t)]\!])_{nn} - \sum_{k\neq n}\gamma_{kn}\rho_{nn}(t) + \sum_{k\neq n}\gamma_{nk}\rho_{kk}(t), \tag{3}$$

where H represents the Hamiltonian dynamics of S, and γ_{kn} is the rate of population relaxation from state $|n\rangle$ to state $|k\rangle$, which depends on the lifetime of state $|n\rangle$, and in case of multiple decay pathways, the probability for the particular transition, etc. The γ_{kn} are thus by definition real and nonnegative. Population relaxation necessarily induces phase relaxation, and we will later derive explicit expressions for the contribution of population relaxation to the phase relaxation rates.

In general, phase relaxation occurs when the interaction of the system with the environment destroys phase correlations between quantum states, and thus converts coherent superposition states into incoherent mixed states. Since coherence is determined by the off-diagonal elements in our expansion of the density operator, this effect can be modeled as decay of the off-diagonal elements of ρ:

$$\dot{\rho}_{kn}(t) = -\frac{i}{\hbar}([\![H,\rho(t)]\!])_{kn} - \Gamma_{kn}\rho_{kn}(t), \tag{4}$$

where Γ_{kn} (for $k\neq n$) is the dephasing rate of the transition $|k\rangle \leftrightarrow |n\rangle$.

[2] An energy eigenstate of the system is a Hilbert space wave function $|n\rangle$ that satisfies $H|n\rangle = E_n|n\rangle$, where H is the Hamiltonian of the system.

Hence, population and phase relaxation change the evolution of the system and force us to rewrite its quantum Liouville equation as

$$\dot{\rho}(t) = -\frac{i}{\hbar}[\![H,\rho(t)]\!] + L_D[\rho(t)], \tag{5}$$

where $L_D[\rho(t)]$ is the dissipation (super)operator determined by the relaxation rates. It is convenient to note here that the $N\times N$ density matrix $\rho(t)$ can be rewritten as an N^2 (column) vector, which we denote as $|\rho(t)\rangle\rangle$, by stacking its columns. Since the commutator $[\![H,\rho(t)]\!]$ and the dissipation (super-)operator $L_D[\rho(t)]$ are linear operators on the set of density matrices, we can write Eq. (5) in matrix form:

$$\frac{d}{dt}|\rho(t)\rangle = \left(-\frac{i}{\hbar}\mathcal{L}_H + \mathcal{L}_D\right)|\rho(t)\rangle, \tag{6}$$

where $\mathcal{L}_H$ and $\mathcal{L}_D$ are $N^2\times N^2$ matrices representing the Hamiltonian and dissipative parts of the dynamics, respectively. Comparison with Eqs. (3) and (4) shows that the nonzero elements of $\mathcal{L}_D$ are

$$\begin{aligned}
(\mathcal{L}_D)_{(m,n),(m,n)} &= -\Gamma_{mn}, \quad m \neq n,\\
(\mathcal{L}_D)_{(m,m),(m',m')} &= +\gamma_{mm}, \quad m \neq m',\\
(\mathcal{L}_D)_{(m,m),(m,m)} &= -\sum_{\substack{k=1\\ k\neq m}}^{N} \gamma_{km},
\end{aligned} \tag{7}$$

where the index (m,n) should be interpreted as $m+(n-1)N$. $\Gamma_{mn}=\Gamma_{nm}$ implies $(\mathcal{L}_D)_{(m,n),(m,n)}=(\mathcal{L}_D)_{(n,m),(n,m)}$.

For a three-level system subject to population and phase relaxation, for instance, Eq. (7) gives a dissipation superoperator of the form

$$\mathcal{L}_D = -\begin{bmatrix}
\gamma_{21}+\gamma_{31} & 0 & 0 & 0 & -\gamma_{12} & 0 & 0 & 0 & -\gamma_{13}\\
0 & \Gamma_{12} & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & \Gamma_{13} & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & \Gamma_{12} & 0 & 0 & 0 & 0 & 0\\
-\gamma_{21} & 0 & 0 & 0 & \gamma_{12}+\gamma_{32} & 0 & 0 & 0 & -\gamma_{23}\\
0 & 0 & 0 & 0 & 0 & \Gamma_{23} & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & \Gamma_{13} & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \Gamma_{23} & 0\\
-\gamma_{31} & 0 & 0 & 0 & -\gamma_{32} & 0 & 0 & 0 & \gamma_{13}+\gamma_{23}
\end{bmatrix}, \tag{8}$$

where γ_{kn} and Γ_{kn} are the population and phase relaxation rates, respectively.

III. PHYSICAL CONSTRAINTS ON THE DYNAMICAL EVOLUTION

Although Eq. (7) gives a general form for the dissipation superoperator of a system subject to population and phase relaxation, not every superoperator $\mathcal{L}_D$ of this form is acceptable on physical grounds, since the density operator $\rho(t)$ of the system must remain Hermitian with non-negative eigenvalues for all $t>0$, and its trace must be conserved.[3] It is easy to see that the relaxation parameters in Eq. (7) cannot be chosen arbitrarily if we are to obtain a valid density operator. For instance, it is well known in quantum optics that a two-level atom with decay $|2\rangle\rightarrow|1\rangle$ at the rate $\gamma_{12}>0$ also experiences dephasing at a rate $\Gamma_{12}\geqslant\frac{1}{2}\gamma_{12}$ since the coherence ρ_{12} must decay with the population of the upper level in order for $\rho(t)$ to remain positive, consistent with the constraints on the relaxation rates for two-level systems derived in [16,19].

[3]Trace conservation means that the sum of the populations of all basis states is preserved, and it is equivalent to conservation of probability. This condition may be violated, for instance, if the total population of the system is not conserved, e.g., by atoms being ionized or mapped outside the subspace S or particles being lost from a trap. However, this condition is in principle not restrictive since we can usually amend the Hilbert space $\mathcal{H}_S$ by adding a subspace B that accounts for population losses from system S so that the total population of $S+B$ is conserved under the open system evolution resulting from the interaction of $S+B$ with the environment.

In higher dimensions we also expect population relaxation from state $|n\rangle$ to $|k\rangle$ at the rate γ_{kn} to induce dephasing of this transition at the rate $\Gamma_{kn} \geqslant \frac{1}{2}\gamma_{kn}$. However, for $N>2$ the situation is more complicated. First, a single random decay $|n\rangle \to |k\rangle$ due to spontaneous emission, for instance, may affect other transitions involving the states $|k\rangle$ or $|n\rangle$. This is perhaps not too surprising but since it is a crucial motivation for the following sections, we shall consider two concrete examples.

First, consider a three-level system subject to decay $|2\rangle \to |1\rangle$ at the rate γ_{12} but no other relaxation. Suppose, for instance, we follow formula (6.A.11) in [21] and set $\Gamma_{12} = \frac{1}{2}\gamma_{12}$ and take all other relaxation rates to be zero. Then, assuming $H=0$ for convenience, the solution of Eq. (5) for this dissipation superoperator leads to the density matrix

$$\rho(t) = \begin{pmatrix} \rho_{11} + (1 - e^{-t\gamma_{12}})\rho_{22} & e^{-t\gamma_{12}/2}\rho_{12} & \rho_{13} \\ e^{-t\gamma_{12}/2}\rho_{21} & e^{-t\gamma_{12}}\rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix}, \tag{9}$$

which in general is not positive for $t>0$. For example, the superposition state

$$\rho(0) = |\Psi\rangle\langle\Psi|, \ |\Psi\rangle = \frac{1}{\sqrt{3}}(1,1,1)^T \tag{10}$$

evolves under the action of this dynamical generator to a "state" $\rho(t)$ which has a negative eigenvalue (i.e., negative populations) for all $t>0$ as shown in Fig. 1 and is thus physically unacceptable.

Furthermore, population relaxation is not the only source of constraints on the decoherence rates for $N>2$. A perhaps more surprising observation is that, even if there is no population relaxation at all, i.e., $\gamma_{kn}=0$ for all k,n, and the system experiences only pure dephasing, we cannot choose the decoherence rates Γ_{kn} arbitrarily. For example, setting $\Gamma_{12}\neq 0$ and $\Gamma_{23}=\Gamma_{13}=0$ for our three-level system gives

$$\rho(t) = \begin{pmatrix} \rho_{11} & e^{-\Gamma_{12}t}\rho_{12} & \rho_{13} \\ e^{-\Gamma_{12}t}\rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix}. \tag{11}$$

Choosing $\rho(0)$ as in Eq. (10) we again obtain a density operator $\rho(t)$ with negative eigenvalues (see Fig. 1). This shows that there must be additional constraints on the decoherence rates to ensure that the state of the system remains physical.

IV. STANDARD FORM OF DISSIPATION SUPEROPERATORS

Significant progress toward solving the problem of finding dynamical generators for open systems that ensure complete positivity of the evolution operator, and hence positivity of the system's density matrix, was made by Gorini, Kossakowski, and Sudarshan [16] who showed that the generator of a quantum dynamical semigroup can be expressed in the standard form

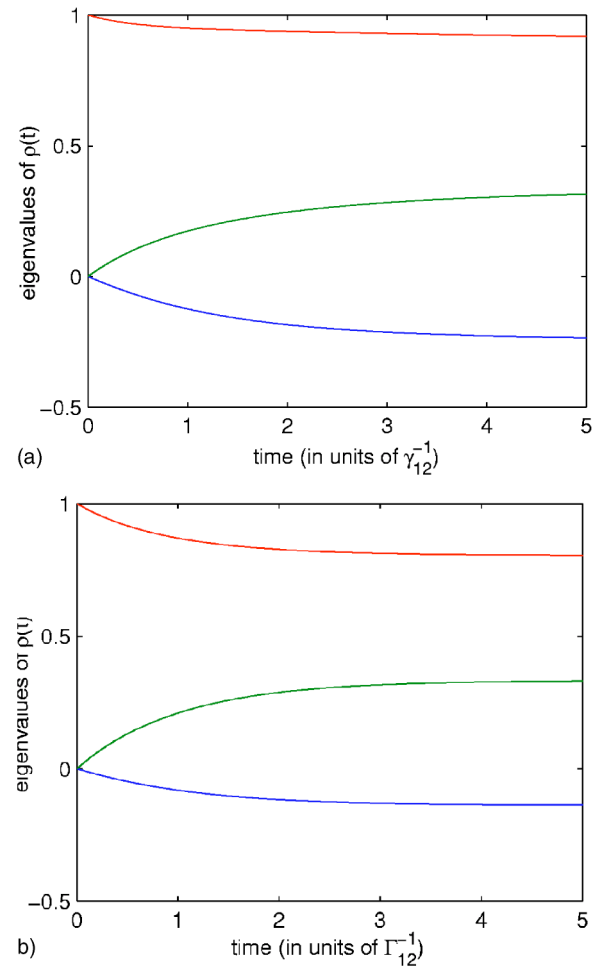

FIG. 1. Eigenvalues of Eq. (9), left, and Eq. (11), right, for $\rho(0)$ as in Eq. (10).

$$L[\rho(t)] = -i[\![H,\rho(t)]\!] + \frac{1}{2}\sum_{k,k'=1}^{N^2-1} a_{kk'}([\![V_k\rho(t),V_{k'}^\dagger]\!] + [\![V_k,\rho(t)V_{k'}^\dagger]\!]), \tag{12}$$

where H is the generator for the Hamiltonian part of the evolution and the V_k, $k=1,2,\ldots,N^2-1$, are trace-zero, orthonormal operators $(V_k,V_{k'}) := \mathrm{Tr}(V_k^\dagger V_{k'}) = \delta_{kk'}$ that together with $V_{N^2}=I/\sqrt{N}$ form a basis for the system's Liouville space. Furthermore, the resulting evolution operator is *completely positive* if and only if the coefficient matrix $a = (a_{kk'})$ is positive.

Noting that a positive matrix $(a_{kk'})$ has real, non-negative eigenvalues γ_k and can be diagonalized by a unitary transformation, we obtain the second standard representation of the dissipative dynamical generator, which was first derived (independently) by Lindblad [14]:

$$L[\rho(t)] = -i[\![H,\rho(t)]\!] + \frac{1}{2}\sum_{k=1}^{N^2-1} \gamma_k([\![A_k\rho(t),A_k^\dagger]\!] + [\![A_k,\rho(t)A_k^\dagger]\!]). \tag{13}$$

Yet, although the general expressions (12) and (13) have been known for more than two decades, it is often unknown

whether a proposed generator for the dissipative dynamics for a particular model is completely positive, and some common dissipative generators have been shown not to satisfy this condition, as in the case of the Agarwal-Redfield equations mentioned earlier. In part this may be due to the fact that it is often very difficult in practice to express phenomenologically derived dissipation generators in either of the two standard forms and hence to verify if a proposed generator satisfies complete positivity.

However, given a matrix representation for the relaxation superoperator of the form (7), which was derived from a purely phenomenological model based on observable decay and decoherence rates, we can express it in standard form (12) and transform abstract positivity requirements into concrete, easily verifiable constraints on the empirically observable relaxation rates. For this purpose we need a basis $\{V_k\}$ for the Liouville space of the system. A canonical choice is to define $N-1$ diagonal matrices

$$V_{(m,m)} = \frac{1}{\sqrt{m+m^2}}\left(\sum_{s=1}^{m} \mathbf{e}_{ss} - m\mathbf{e}_{m+1,m+1}\right) \tag{14}$$

for $m=1,2,\ldots,N-1$, as well as N^2-N off-diagonal matrices

$$V_{(m,n)} = \mathbf{e}_{mn},\ m \neq n,\ m,n = 1,2,\ \ldots,N, \tag{15}$$

where $\mathbf{e}_{mn}$ is an $N\times N$ matrix whose entries are zero except for a 1 in the mth row, nth column position. It is quite easy to verify that the N^2-1 operators $V_{(m,n)}$ thus defined are trace-zero $N\times N$ matrices that satisfy the orthonormality condition $\mathrm{Tr}(V_k V_{k'}^{\dagger}) = \delta_{kk'}$ for any dimension N.

Having defined the basis operators V_k, we can now compute the generators

$$L_{kk'}[\rho(t)] = \frac{1}{2}(\llbracket V_k\rho(t), V_{k'}^{\dagger}\rrbracket + \llbracket V_k, \rho(t)V_{k'}^{\dagger}\rrbracket) \tag{16}$$

of the dissipation superoperator (12) with respect to this basis, where $k,k'=1,2,\ldots,N^2-1$. Recalling that $L_{kk'}[\rho(t)]$ is equivalent to $\mathcal{L}_{kk'}|\ \rho(t)\rangle\rangle$, where each $\mathcal{L}_{kk'}$ is an $N^2\times N^2$ matrix and $|\ \rho(t)\rangle\rangle$ is an N^2-column vector, we note that any (trace-preserving) dissipation superoperator $\mathcal{L}_D$ can be written as a linear combination of these dissipation generators:

$$\mathcal{L}_D = \sum_{k,k'=1}^{N^2-1} a_{kk'}\mathcal{L}_{kk'}. \tag{17}$$

To compute the coefficient matrix $a=(a_{kk'})$ we can rewrite the $N^2\times N^2$ matrices $\mathcal{L}_{kk'}$ and $\mathcal{L}_D$ as column vectors $\vec{\mathcal{L}}_{kk'}$ and $\vec{\mathcal{L}}_D$ of length N^4, and a as a column vector $\vec{a}$ of length $(N^2-1)^2$, and solve the linear equation $\vec{\mathcal{L}}_D = \mathcal{A}\vec{a}$ where $\mathcal{A}$ is an $N^4\times(N^2-1)^2$ matrix whose columns are the $\vec{\mathcal{L}}_{kk'}$. This matrix equation has a solution for any trace-zero Liouville operator since the columns of $\mathcal{A}$ span the space of trace-zero Liouville operators of the system. This procedure allows us in principle to express any (trace-preserving) dissipation superoperator in the standard form (12), and to verify whether it generates a completely positive evolution operator by checking the eigenvalues of the coefficient matrix $(a_{kk'})$. However, in practice this is not very efficient, especially for large N. Instead, we would like to be able to express the coefficients $a_{kk'}$ directly in terms of observable relaxation rates. This is the aim of the following sections.

V. DECOMPOSITION OF RELAXATION SUPEROPERATOR

We now use Eq. (17) to show that the relaxation superoperator of any N-level system subject to both population and phase relaxation processes can be decomposed into a part associated with population relaxation processes and another accounting for pure decoherence. To this end, we introduce two types of decoherence rates Γ_{mn}^p and Γ_{mn}^d for decoherence due to population relaxation and pure phase relaxation (dephasing), respectively, and require that $\Gamma_{mn} = \Gamma_{mn}^p + \Gamma_{mn}^d$.

If we have population relaxation $|n\rangle \to |m\rangle$ at the rate $\gamma_{mn} \geqslant 0$ for $m,n=1,2,\ldots,N$ (with $\gamma_{mm}=0$), then setting $a_{kk} = \gamma_{mn}$ for $k=m+(n-1)N$ and $a_{kk'}=0$ otherwise in Eq. (17) leads to a dissipation superoperator

$$\mathcal{L}_D^p = \sum_{m,n=1}^{N} \gamma_{mn}\mathcal{L}_{(m,n),(m,n)}. \tag{18}$$

Inserting Eq. (16) for $\mathcal{L}_{(m,n),(m,n)}$ with $k=m+(n-1)N$ and V_k as in Eqs (14) and (15), we obtain

$$(\mathcal{L}_D^p)_{(m,m),(m,m)} = -\sum_{k=1,k\neq m}^{N} \gamma_{km},$$

$$(\mathcal{L}_D^p)_{(m,m),(m',m')} = \gamma_{mm'}, \quad m \neq m'$$

$$(\mathcal{L}_D^p)_{(m,n),(m,n)} = -\frac{1}{2}\sum_{k=1}^{N}(\gamma_{km}+\gamma_{kn}), \quad m \neq n,$$

which agrees with the general form Eq. (7) of the relaxation superoperator, yields the correct population relaxation rates, and suggests that the dephasing rates due to population relaxation are given by

$$\Gamma_{mn}^p = \frac{1}{2}\sum_{k=1}^{N}(\gamma_{km}+\gamma_{kn}), \quad m \neq n, \tag{19}$$

i.e., that the *decay-induced decoherence* of the transition between states $|n\rangle$ and $|m\rangle$ is one-half of the sum over all decay rates from either of the two states $|m\rangle$ or $|n\rangle$ to any other state. Finally, inserting

$$\Gamma_{mn} = \Gamma_{mn}^d + \frac{1}{2}\sum_{k=1}^{N}(\gamma_{km}+\gamma_{kn}), \quad m \neq n, \tag{20}$$

into Eq. (7) and solving Eq. (17) shows that the dissipation superoperator $\mathcal{L}_D$ of the system decomposes, $\mathcal{L}_D = \mathcal{L}_D^p + \mathcal{L}_D^d$, with L_D^p given by Eq. (18) and

$$\mathcal{L}_D^d = \sum_{m,m'=1}^{N-1} a_{(m,m),(m',m')} \mathcal{L}_{(m,m),(m',m')}. \tag{21}$$

Thus, given $\mathcal{L}_D$ and the population relaxation rates γ_{mn} of the system, we can compute $\mathcal{L}_D^d = \mathcal{L}_D - \mathcal{L}_D^p$ and determine the coefficients $b_{mm'} := a_{(m,m),(m',m')}$ in Eq. (21) by rewriting the superoperators $\mathcal{L}_D^d$ and $\mathcal{L}_{(m,m),(m',m')}$ as column vectors $\vec{l}$ and $\vec{l}_k$, respectively, defining a matrix $\mathcal{B}$ whose columns are given by $\vec{l}_k$, and setting $\vec{b} = \mathcal{B}^{-1}\vec{l}$ where $\mathcal{B}^{-1}$ denotes the pseudoinverse of $\mathcal{B}$. However, note that the matrix $\mathcal{B}$ has only $(N-1)^2$ instead of $(N^2-1)^2$ columns, and we can eliminate all zero rows. The resulting coefficient vector $\vec{b}$ can be rearranged into an $(N-1)\times(N-1)$ coefficient matrix $b = (b_{mm'})$ that depends only on the pure dephasing rates Γ_{mn}^d. Furthermore, the requirement of positivity of the coefficient matrix $(a_{kk'})$ in Eq. (12) now reduces to the (much simpler) requirement that the $(N-1)\times(N-1)$ matrix $(b_{mm'})$ be positive semidefinite.

It is important to note that our formula (19) for the contribution of population relaxation to the overall decoherence rates, obtained solely by imposing the physical constraint of complete positivity on the evolution of the system, agrees with the expressions given, for instance, by Shore [22] for the general Bloch equations of N-level atoms subject to various dissipative processes, but our general expression for the dissipation superoperator covers systems subject to both population decay and pure dephasing processes, and implies the existence of nontrivial constraints on the pure dephasing rates of the system for $N>2$. In the following sections, we shall analyze these constraints in detail for $N=3$ and $N=4$.

VI. CONSTRAINTS ON THE PURE DEPHASING RATES

A. Three-level systems

Expanding the relaxation superoperator (8) for a three-level system with respect to the basis

$$V_{(1,1)} = \frac{1}{\sqrt{2}}(\mathbf{e}_{11} - \mathbf{e}_{22}),$$

$$V_{(2,2)} = \frac{1}{\sqrt{6}}(\mathbf{e}_{11} + \mathbf{e}_{22} - 2\mathbf{e}_{33}), \tag{22}$$

$$V_{(m,n)} = \mathbf{e}_{mn},\ m,n = 1,2,3, m \neq n,$$

where $\mathbf{e}_{mn}$ is the 3×3 matrix whose entries are zero except for a 1 in the mth row, nth column position—which corresponds to the canonical basis (14) and (15) for $N=3$, yields an 8×8 coefficient matrix $(a_{kk'})$ whose nonzero entries are $a_{kk} = \gamma_{mn}$ for $k = m + 3(n-1)$ and $m \neq n$, as well as $a_{(m,m),(m',m')} = b_{mm'}$, where

$$\begin{pmatrix} b_{11} \\ b_{21} \\ b_{12} \\ b_{22} \end{pmatrix} = \underbrace{\begin{pmatrix} 1 & -\frac{1}{6}\sqrt{3} & \frac{1}{6}\sqrt{3} & 0 \\ \frac{1}{4} & \frac{5}{12}\sqrt{3} & \frac{1}{12}\sqrt{3} & \frac{3}{4} \\ 1 & \frac{1}{6}\sqrt{3} & -\frac{1}{6}\sqrt{3} & 0 \\ \frac{1}{4} & -\frac{5}{12}\sqrt{3} & -\frac{1}{12}\sqrt{3} & \frac{3}{4} \\ \frac{1}{4} & \frac{1}{12}\sqrt{3} & \frac{5}{12}\sqrt{3} & \frac{3}{4} \\ \frac{1}{4} & -\frac{1}{12}\sqrt{3} & -\frac{5}{12}\sqrt{3} & \frac{3}{4} \end{pmatrix}}_{\mathcal{B}}^{-1} \begin{pmatrix} \Gamma_{12}^d \\ \Gamma_{13}^d \\ \Gamma_{12}^d \\ \Gamma_{23}^d \\ \Gamma_{13}^d \\ \Gamma_{23}^d \end{pmatrix}.$$

and the pure dephasing rates Γ_{mn}^d are defined by Eq. (20). Noting that the pseudoinverse of the matrix $\mathcal{B}$ is

$$\begin{pmatrix} \frac{1}{2} & 0 & \frac{1}{2} & 0 & 0 & 0 \\ \frac{1}{2\sqrt{3}} & \frac{1}{\sqrt{3}} & \frac{1}{2\sqrt{3}} & -\frac{1}{\sqrt{3}} & 0 & 0 \\ \frac{1}{2\sqrt{3}} & 0 & -\frac{1}{2\sqrt{3}} & 0 & \frac{1}{\sqrt{3}} & -\frac{1}{\sqrt{3}} \\ -\frac{1}{6} & \frac{1}{3} & -\frac{1}{6} & \frac{1}{3} & \frac{1}{3} & \frac{1}{3} \end{pmatrix}$$

we obtain

$$b_{11} = \Gamma_{12}^d,$$

$$b_{22} = (-\Gamma_{12}^d + 2\Gamma_{13}^d + 2\Gamma_{23}^d)/3, \tag{23}$$

$$b_{12} = b_{21} = (\Gamma_{13}^d - \Gamma_{23}^d)/\sqrt{3}.$$

Therefore, the coefficient matrix $(a_{kk'})$ of the relaxation superoperator $\mathcal{L}_D$ will be positive semidefinite if and only if $\gamma_{mn} \geqslant 0$ and the real symmetric 2×2 matrix

$$\begin{pmatrix} b_{11} & b_{12} \\ b_{21} & b_{22} \end{pmatrix}$$

has non-negative eigenvalues. The second condition is equivalent to

$$b_{11} + b_{22} \geqslant 0,\ b_{11}b_{22} \geqslant b_{12}^2. \tag{24}$$

Substituting Eq. (23), these conditions become

$$0 \leqslant \Gamma_{12}^d + \Gamma_{13}^d + \Gamma_{23}^d \leqslant 2\sqrt{\Gamma_{12}^d\Gamma_{13}^d + \Gamma_{12}^d\Gamma_{23}^d + \Gamma_{13}^d\Gamma_{23}^d}. \tag{25}$$

This double inequality provides upper and lower bounds for the pure dephasing rates of the system. As expected we can also show that each relaxation rate must be non-negative since the second inequality is of the form $[a+(b+c)]^2$

$\leqslant 4[a(b+c)+bc]$, which can be rewritten as $a^2+(b-c)^2 \leqslant 2a(b+c)$, and the first inequality implies $a+b+c \geqslant 0$, which shows that $a \geqslant 0$ and $b+c \geqslant 0$, and by symmetry of a,b,c requires that Γ^d_{12}, Γ^d_{13} and Γ^d_{23} be non-negative. Hence, we can rewrite (25) as follows:

$$(\sqrt{\Gamma_b}-\sqrt{\Gamma_c})^2 \leqslant \Gamma_a \leqslant (\sqrt{\Gamma_b}+\sqrt{\Gamma_c})^2, \tag{26}$$

where $\{a,b,c\}$ is any permutation of $\{12, 13, 23\}$.

Note that the choice $\Gamma^d_{12}>0$ and $\Gamma^d_{13}=\Gamma^d_{23}=0$, which corresponds to the second example in Sec. III if there is no population relaxation, clearly violates (26), which explains why it results in nonphysical evolution. We also see that allowed choices include, for instance, $\Gamma^d_{12}=\Gamma^d_{23}>0$ and $\Gamma^d_{13}=0$.

In general, (26) shows that pure dephasing in a three-level system always affects more than one transition. Furthermore, if two of the pure dephasing rates are equal, say Γ^d, then the third rate must be between 0 and $4\Gamma^d$. For instance, consider a triply degenerate atomic energy level with basis states $|m=0\rangle$ and $|m=\pm 1\rangle$. If $\Gamma^d_{-1,0}=\Gamma^d_{0,1}$ then $0 \leqslant \Gamma^d_{-1,1} \leqslant 4\Gamma^d_{0,1}$, i.e., the decoherence rate of the transition between the outer states can be at most $4\Gamma^d_{0,1}$. But note in particular that it could be zero even if the decoherence rate between adjacent states is nonzero.

B. Four-level systems

If we expand the relaxation superoperator $\mathcal{L}_D$ for a four-level system as discussed in Sec. V with respect to the standard basis (14) and (15), we again obtain a coefficient matrix $(a_{(m,n),(m',n')})$ whose nonzero entries are $a_{(m,n),(m,n)}=\gamma_{mn}$ for $m \neq n$, as well as $a_{(m,m),(m',m')}=b_{mm'}$ with b_{11}, b_{12}, b_{21} and b_{22} as in Eq. (23), $b_{31}=b_{13}$, $b_{32}=b_{23}$, and

$$\begin{aligned} b_{13} &= \sqrt{6}(-\Gamma^d_{13}+3\Gamma^d_{14}+\Gamma^d_{23}-3\Gamma^d_{24})/12, \\ b_{23} &= \sqrt{2}(-2\Gamma^d_{12}+\Gamma^d_{13}+3\Gamma^d_{14}+\Gamma^d_{23}+3\Gamma^d_{24}-6\Gamma^d_{34})/12, \\ b_{33} &= (-\Gamma^d_{12}-\Gamma^d_{13}+3\Gamma^d_{14}-\Gamma^d_{23}+3\Gamma^d_{24}+3\Gamma^d_{34})/6. \end{aligned} \tag{27}$$

Since the reduced coefficient matrix $b=(b_{mm'})$ is a real, symmetric 3×3 matrix, necessary and sufficient conditions for it to be positive semidefinite are [23]

$$b_{11} \geqslant 0, \quad b_{11}b_{22} \geqslant b_{12}^2, \quad \det(b) \geqslant 0. \tag{28}$$

The first two of these conditions are equivalent to (24). Thus, the pure dephasing rates for a four-level system must satisfy (25) and (26), and the new constraint $\det(b) \geqslant 0$, or

$$b_{11}b_{22}b_{33}+2b_{12}b_{13}b_{23} \geqslant b_{11}b_{23}^2+b_{22}b_{13}^2+b_{33}b_{12}^2. \tag{29}$$

Unfortunately, inserting Eqs. (23) and (27) into this inequality does not yield a nice form for the additional constraint.

We can obtain a more symmetric form of the constraints by choosing a slightly different operator basis:

$$\begin{aligned} V'_{(1,1)} &= \frac{1}{2}(\mathbf{e}_{11}-\mathbf{e}_{22}+\mathbf{e}_{33}-\mathbf{e}_{44}), \\ V'_{(2,2)} &= \frac{1}{2}(\mathbf{e}_{11}-\mathbf{e}_{22}-\mathbf{e}_{33}+\mathbf{e}_{44}), \\ V'_{(3,3)} &= \frac{1}{2}(\mathbf{e}_{11}+\mathbf{e}_{22}-\mathbf{e}_{33}-\mathbf{e}_{44}), \\ V'_{(m,n)} &= \mathbf{e}_{mn}, \quad m,n=1,2,3,4, \quad m \neq n. \end{aligned} \tag{30}$$

The $V'_{(m,n)}$ are trace-zero matrices that differ from the standard operator basis only in the choice of the diagonal generators and also form an orthonormal basis for the trace-zero Liouville operators of the system. However, expanding $\mathcal{L}_D$ with respect to this basis (30) gives a more symmetric coefficient matrix b' with nonzero entries:

$$\begin{aligned} b'_{11} &= \Gamma^d_{\text{tot}}-(\Gamma^d_{13}+\Gamma^d_{24}), \\ b'_{22} &= \Gamma^d_{\text{tot}}-(\Gamma^d_{14}+\Gamma^d_{23}), \\ b'_{33} &= \Gamma^d_{\text{tot}}-(\Gamma^d_{12}+\Gamma^d_{34}), \\ b'_{12} &= b'_{21}=(\Gamma^d_{12}-\Gamma^d_{34})/2, \\ b'_{13} &= b'_{31}=(\Gamma^d_{14}-\Gamma^d_{23})/2, \\ b'_{23} &= b'_{32}=(\Gamma^d_{13}-\Gamma^d_{24})/2, \end{aligned} \tag{31}$$

where $\Gamma^d_{\text{tot}}=\frac{1}{2}\Sigma^4_{n=2}\Sigma^{n-1}_{m=1}\Gamma^d_{mn}$ is half the sum of all the pure dephasing rates. Since the eigenvalues of the coefficient matrix are independent of the operator basis, b' has the same eigenvalues as b. Furthermore, necessary conditions for b' to have non-negative eigenvalues are[4]

$$b_{11} \geqslant 0, \quad b_{22} \geqslant 0, \quad b_{33} \geqslant 0, \tag{32}$$

$$b_{11}b_{22} \geqslant b_{12}^2, \quad b_{11}b_{33} \geqslant b_{13}^2, \quad b_{22}b_{33} \geqslant b_{23}^2. \tag{33}$$

Inserting (31) into (32) and (33) yields

$$\begin{aligned} \Gamma^d_{13}+\Gamma^d_{24} &\leqslant \Gamma^d_{12}+\Gamma^d_{14}+\Gamma^d_{23}+\Gamma^d_{34}, \\ \Gamma^d_{14}+\Gamma^d_{23} &\leqslant \Gamma^d_{12}+\Gamma^d_{13}+\Gamma^d_{24}+\Gamma^d_{34}, \\ \Gamma^d_{12}+\Gamma^d_{34} &\leqslant \Gamma^d_{13}+\Gamma^d_{14}+\Gamma^d_{23}+\Gamma^d_{24}, \end{aligned} \tag{34}$$

as well as

$$\begin{aligned} (\Gamma^d_{14}+\Gamma^d_{23}-\Gamma^d_{13}-\Gamma^d_{24})^2 &\leqslant 4\Gamma^d_{12}\Gamma^d_{34}, \\ (\Gamma^d_{12}+\Gamma^d_{34}-\Gamma^d_{13}-\Gamma^d_{24})^2 &\leqslant 4\Gamma^d_{14}\Gamma^d_{23}, \end{aligned} \tag{35}$$

[4]To see that these conditions are necessary note that $b_{11} \geqslant 0$ and $b_{11}b_{22} \geqslant b_{12}^2$ implies $b_{22} \geqslant 0$. Inserting $b_{12}=b_{23}=b_{13}=0$ into (29) yields $b_{11}b_{22}b_{33} \geqslant 0$ and hence $b_{33} \geqslant 0$; inserting $b_{12}=b_{23}=0$ yields $b_{22}(b_{11}b_{33}-b_{13}^2) \geqslant 0$, and $b_{12}=b_{13}=0$ yields $b_{11}(b_{22}b_{33}-b_{23}^2) \geqslant 0$. However, these conditions are not sufficient since setting $b_{11}=b_{22}=b_{33}=b_{12}=b_{23}=1$ and $b_{13}=-1$, for instance, satisfies both (32) and (33) but gives $\det(b)=-4$ and thus violates (28).

$$(\Gamma_{12}^d + \Gamma_{34}^d - \Gamma_{14}^d - \Gamma_{23}^d)^2 \leqslant 4\Gamma_{13}^d\Gamma_{24}^d,$$

which can be written simply as

$$|b-c| \leqslant a \leqslant b+c, \quad (b-c)^2 \leqslant 4xy \tag{36}$$

if $\{a,b,c\}$ is a permutation of the set $\{\Gamma_{12}^d+\Gamma_{34}^d, \Gamma_{13}^d+\Gamma_{24}^d, \Gamma_{14}^d+\Gamma_{23}^d\}$ and we let x and y be the summands of a, e.g., if $a=\Gamma_{12}^d+\Gamma_{34}^d$ then $x=\Gamma_{12}^d$ and $y=\Gamma_{34}^d$. Setting $a=x+y$ shows in particular that $0\leqslant x+y$ and $0\leqslant 4xy$ and thus $x,y \geqslant 0$, from which we can conclude especially that $\Gamma_{mn}^d \geqslant 0$ for all m, n.

In certain cases these constraints can be simplified. For instance, if the pure dephasing rates for transitions between adjacent states are equal, i.e., $\Gamma_{12}^d=\Gamma_{23}^d=\Gamma_{34}^d=\Gamma_1^d$, as one might expect, for example, for a system consisting of the basis states of a fourfold-degenerate energy level, and we set $\Gamma_2^d=\frac{1}{2}(\Gamma_{13}^d+\Gamma_{24}^d)$ then (34) yields the following bounds on the decoherence rate Γ_{14}^d:

$$\max\{2\Gamma_2^d - 3\Gamma_1^d, \Gamma_1^d - 2\Gamma_2^d, 0\} \leqslant \Gamma_{14} \leqslant \Gamma_1^d + 2\Gamma_2^d \tag{37}$$

and combined with the second inequality of (35) we obtain $0\leqslant\Gamma_2^d\leqslant 4\Gamma_1^d$, and thus $\Gamma_{14}^d\leqslant 9\Gamma_1^d$ and $\Gamma_{13}^d,\Gamma_{24}^d\leqslant 8\Gamma_1^d$. If we further assume that the pure dephasing rates for transitions between next-to-nearest neighbor states are equal as well, i.e., $\Gamma_{13}^d=\Gamma_{14}^d=\Gamma_2^d$, then we obtain $\Gamma_{13}^d=\Gamma_{24}^d\leqslant 4\Gamma_1^d$. Hence, for a system whose pure dephasing rates depend only on the "distance" between the states, the former are bounded above by

$$\Gamma_{nk}^d \leqslant (n-k)^2\Gamma_1^d, \tag{38}$$

where $\Gamma_1^d=\Gamma_{n,n+1}^d$ is the dephasing rate for transitions between adjacent sites.

In this special case we can compare the constraints obtained from our necessary conditions with the necessary and sufficient conditions (28). Inserting $\Gamma_{nk}^d=\Gamma_{|n-k|}^d$, into Eq. (31) yields

$$b'_{11} = \frac{1}{2}(3\Gamma_1^d - 2\Gamma_2^d + \Gamma_3^d),$$

$$b'_{22} = \frac{1}{2}(\Gamma_1^d + 2\Gamma_2^d - \Gamma_3^d),$$

$$b'_{33} = \frac{1}{2}(-\Gamma_1^d + 2\Gamma_2^d - 3\Gamma_1^d),$$

$$b'_{12} = b'_{21} = 0,$$

$$b'_{13} = b'_{31} = \frac{1}{2}(\Gamma_3^d - \Gamma_1^d),$$

$$b'_{23} = b'_{32} = 0, \tag{39}$$

and the necessary and sufficient conditions (28) become

$$\Gamma_3^d \geqslant 2\Gamma_2^d - 3\Gamma_1^d,$$

$$\Gamma_3^d \leqslant 2\Gamma_2^d + \Gamma_1^d, \tag{40}$$

$$\Gamma_1^d\Gamma_3^d \geqslant (\Gamma_1^d - \Gamma_2^d)^2.$$

If $\Gamma_1^d=0$ then $\Gamma_3^d=2\Gamma_2^d=0$. Otherwise, we can multiply the second inequality by Γ_1^d and combine it with the third, which leads to $\Gamma_1^d(2\Gamma_2^d+\Gamma_1^d)\geqslant(\Gamma_1^d-\Gamma_2^d)^2$ and simplifies to $\Gamma_2^d\leqslant 4\Gamma_1^d$. Inserting this result in the first inequality gives $\Gamma_3^d\leqslant 9\Gamma_1^d$, i.e., the necessary conditions (38) are also sufficient.

VII. EXAMPLES AND DISCUSSION

We now apply the results of the previous sections to several types of generic three- and four-level atoms. The objective in each case is to derive a proper relaxation superoperator, which is consistent with both experimental data and positivity constraints, and to discuss the implications of the latter constraints. Although the emphasis is on atomic systems, the results generally apply to molecular or solid state systems with similar level structures as well.

A. Generic three-level atoms

Let us first consider the general case of a generic three-level system subject to arbitrary population and phase relaxation processes. Let γ_{mn} denote the observed rate of population relaxation from state $|n\rangle$ to state $|m\rangle$ for $m,n=1,2,3$ and $m\neq n$, and let Γ_{12}, Γ_{23}, and Γ_{13} be the observed decoherence rates for the $1\leftrightarrow 2$, $2\leftrightarrow 3$, and $1\leftrightarrow 3$ transitions, respectively. Then the pure dephasing rates of the system according to Eq. (20) are

$$\Gamma_{12}^d = \Gamma_{12} - (\gamma_{21}+\gamma_{31}+\gamma_{12}+\gamma_{32})/2,$$

$$\Gamma_{13}^d = \Gamma_{13} - (\gamma_{21}+\gamma_{31}+\gamma_{13}+\gamma_{23})/2, \tag{41}$$

$$\Gamma_{23}^d = \Gamma_{23} - (\gamma_{12}+\gamma_{32}+\gamma_{13}+\gamma_{23})/2.$$

These dephasing rates must be non-negative and satisfy the inequality constraints (26) for the evolution of the system to be completely positive. Experimental data for the observed relaxation rates that fail to satisfy these conditions should be considered a reason for concern, and might suggest that the physical system under investigation cannot be adequately modeled as a three-level system, for instance.

If the dephasing rates do satisfy the necessary constraints then a physically valid representation of the relaxation superoperator L_D for the system in terms of the observed relaxation rates is

$$L_D[\rho(t)] = \sum_{m\neq n} \gamma_{mn}L_{mn}^p[\rho(t)] + \delta_1 L_1^d[\rho(t)] + \delta_2 L_2^d[\rho(t)], \tag{42}$$

where the elementary relaxation terms are

$$2L_{mn}^p[\rho(t)] = [\![V_{(m,n)}\rho(t), V_{(m,n)}^\dagger]\!] + [\![V_{(m,n)}, \rho(t)V_{(m,n)}^\dagger]\!],$$

$$2L_m^d[\rho(t)] = [\![A_m\rho(t), A_m]\!] + [\![A_m, \rho(t)A_m]\!], \tag{43}$$

with $V_{(m,n)}$ as in Eq. (22), the diagonal "pure dephasing" generators are

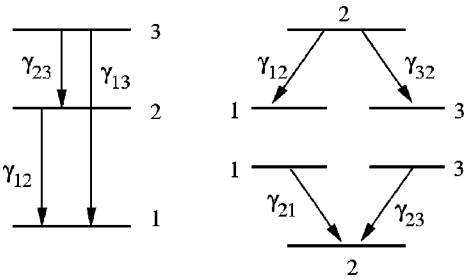

FIG. 2. Three-state atoms: ladder system (left), Λ system (top right), and V system (bottom right) with arrows indicatig population decay pathways.

$$A_1 = \frac{1}{\sqrt{2x(x-\Delta_1)}}[\sqrt{3}\Delta_2 V_{(1,1)} + (x-\Delta_1)V_{(2,2)}],$$

$$A_2 = \frac{-1}{\sqrt{2x(x+\Delta_1)}}[\sqrt{3}\Delta_2 V_{(1,1)} - (x+\Delta_1)V_{(2,2)}], \quad (44)$$

and the "effective dephasing" rates are

$$\delta_{1/2} = (\Gamma_{12}^d + \Gamma_{13}^d + \Gamma_{23}^d \pm x)/3, \quad (45)$$

where $x=\sqrt{\Delta_1^2+3\Delta_2^2}$, $\Delta_1=2\Gamma_{12}-\Gamma_{13}-\Gamma_{23}$, and $\Delta_2=\Gamma_{13}-\Gamma_{23}$.

1. Ladder configurations

For a three-level atom in a ladder configuration where the main source of population relaxation is spontaneous emission from the excited states to a stable ground state, as shown in Fig. 2, we simply set $\gamma_{21}=\gamma_{31}=\gamma_{32}=0$ in Eqs. (41) and (42), respectively, to obtain the correct pure dephasing rates

$$\Gamma_{12}^d = \Gamma_{12} - \gamma_{12}/2,$$

$$\Gamma_{13}^d = \Gamma_{13} - (\gamma_{13}+\gamma_{23})/2, \quad (46)$$

$$\Gamma_{23}^d = \Gamma_{23} - (\gamma_{12}+\gamma_{13}+\gamma_{23})/2,$$

and the corresponding relaxation superoperator $\mathcal{L}_D$. The decay-induced decoherence rates Γ_{mn}^p in this case satisfy the interesting equality $\Gamma_{12}^p+\Gamma_{13}^p=\Gamma_{23}^p$.

This is another way of seeing that, if $\gamma_{12}>0$ as in example 1 considered in Sec. III, then Γ_{23} must be at least $\gamma_{12}/2$—recall that we showed explicitly that the naive guess $\Gamma_{12}=\gamma_{12}/2$ and $\Gamma_{13}=\Gamma_{23}=0$ leads to nonphysical states with negative eigenvalues.

$\Gamma_{13}=0$, on the other hand, is possible even if $\gamma_{12}>0$ provided that state $|3\rangle$ is stable. It is interesting to note, however, that $\Gamma_{13}=0$ always implies $\Gamma_{12}=\Gamma_{23}$. If there is no pure dephasing then this is obvious since $\Gamma_{12}^p=\Gamma_{23}^p$, but it is true even if there is pure dephasing, since $\Gamma_{13}=0$ implies $\Gamma_{13}^d=0$ and thus the inequality constraint (26) for the pure dephasing rates implies $\Gamma_{12}^d=\Gamma_{23}^d$.

2. Λ systems

For a Λ system for which only the decay rates γ_{12} and γ_{32} are nonzero as shown in Fig. 2, the pure dephasing rates are

$$\Gamma_{12}^d = \Gamma_{12} - (\gamma_{12}+\gamma_{32})/2,$$

$$\Gamma_{23}^d = \Gamma_{23} - (\gamma_{12}+\gamma_{32})/2, \quad (47)$$

$$\Gamma_{13}^d = \Gamma_{13}.$$

Moreover, if the lifetime of the excited state is γ^{-1} and the system is symmetric, i.e., $\gamma_{12}=\gamma_{32}=\gamma/2$ and $\Gamma_{12}=\Gamma_{23}$, as is often the case, then we have $\Gamma_{12}=\Gamma_{23}=\Gamma^d+\gamma/2$. If $\Gamma^d=0$ then $\Gamma_{13}^d=0$ due to (25). Otherwise, setting $\Gamma_{13}^d=\alpha\Gamma^d$ gives $\Delta_1=2(1-\alpha)\Gamma^d$, $\Delta_2=-(1-\alpha)\Gamma^d$, $x=2(1-\alpha)\Gamma^d$, and thus the relaxation superoperator is simply

$$L_D[\rho(t)] = (\gamma/2)\{L_{12}^p[\rho(t)] + L_{32}^p[\rho(t)]\} + [(4-\alpha)\Gamma^d/3]L_1^d[\rho(t)] + \alpha\Gamma^d L_2^d[\rho(t)],$$

where the diagonal pure dephasing generators (44) are

$$A_1 = A_1^\dagger = (-\sqrt{3}V_{(1,1)} + V_{(2,2)})/2,$$

$$A_2 = A_2^\dagger = (V_{(1,1)} + \sqrt{3}V_{(2,2)})/2. \quad (48)$$

Positivity requires $\gamma\geqslant 0$, $4-\alpha\geqslant 0$ and $\alpha\geqslant 0$, or $0\leqslant\Gamma_{13}^d\leqslant 4\Gamma^d$, in accordance with (26) and previous observations.

3. V systems

Similarly, for a V system for which only the decay rates γ_{21} and γ_{23} are nonzero as shown in Fig. 2, the pure dephasing rates are simply

$$\Gamma_{12}^d = \Gamma_{12}^d - \gamma_{21}/2,$$

$$\Gamma_{23}^d = \Gamma_{23}^d - \gamma_{23}/2, \quad (49)$$

$$\Gamma_{13}^d = \Gamma_{13}^d - (\gamma_{21}+\gamma_{23})/2.$$

If the system is symmetric, i.e., both excited states have the same lifetime $\gamma_{21}=\gamma_{23}=\gamma$ and $\Gamma_{12}=\Gamma_{23}$ then setting $\Gamma_{13}^d=\alpha\Gamma^d$ leads to the relaxation superoperator

$$L_D[\rho(t)] = \gamma\{L_{21}^p[\rho(t)] + L_{23}^p[\rho(t)]\} + [(4-\alpha)\Gamma^d/3]L_1^d[\rho(t)] + \alpha\Gamma^d L_2^d[\rho(t)]$$

with $L_{mn}[\rho(t)]$ and $L_1[\rho(t)]$ as defined in Eq. (43) and the generators A_1,A_2 as in Eq. (48).

4. Comparison of Λ and V systems

If the excited states have the same lifetime γ^{-1} and the pure dephasing rate Γ^d for transitions between the upper and lower states is the same for the Λ and V configurations, then we have $\Gamma_{12}=\Gamma_{23}=\frac{1}{2}\gamma+\Gamma^d$ in both cases, i.e., the overall decoherence rate for transitions between ground and excited states is the same for both configurations. The main difference, as expected, is the decoherence rate of the $1\leftrightarrow 3$ transition, which is $\Gamma_{13}=\Gamma_{13}^d$ for the Λ system, and $\Gamma_{13}=\gamma+\Gamma_{13}^d$ for the V system.

Thus, if pure dephasing of the $1\leftrightarrow 3$ transition is negligible then it will remain decoherence-free for the Λ system but not for the V system. However, if pure dephasing is taken

into account then the transition between the degenerate ground states of the Λ system may not be decoherence-free, and comparison of the decoherence rates for both systems, $\Gamma_{13}^{\Lambda}=(\Gamma_{13}^{d})^{\Lambda}$ and $\Gamma_{13}^{V}=\gamma+(\Gamma_{13}^{d})^{V}$, shows that Γ_{13}^{Λ} could theoretically even be greater than Γ_{13}^{V} if the pure dephasing rate of the transition between the degenerate ground states was greater than the decay rate γ plus the pure dephasing rate Γ_{13}^{d} for the V system.

B. Generic four-level atoms

Again, we will first consider the general case of a generic four-level system subject to arbitrary population and phase relaxation processes. Let γ_{mn} denote the observed rate of population relaxation from state $|n\rangle$ to state $|m\rangle$ for $m,n=1,2,3,4$ and $m\neq n$, and Γ_{mn} be the observed decoherence rates for the $m\leftrightarrow n$, transitions, as usual. Then the pure dephasing rates of the system according to Eq. (20) are

$$\Gamma_{12}^{d}=\Gamma_{12}-(\gamma_{21}+\gamma_{31}+\gamma_{41}+\gamma_{12}+\gamma_{32}+\gamma_{42})/2,$$

$$\Gamma_{13}^{d}=\Gamma_{13}-(\gamma_{21}+\gamma_{31}+\gamma_{41}+\gamma_{13}+\gamma_{23}+\gamma_{43})/2,$$

$$\Gamma_{14}^{d}=\Gamma_{14}-(\gamma_{21}+\gamma_{31}+\gamma_{41}+\gamma_{14}+\gamma_{24}+\gamma_{34})/2,$$

$$\Gamma_{23}^{d}=\Gamma_{23}-(\gamma_{12}+\gamma_{32}+\gamma_{42}+\gamma_{13}+\gamma_{23}+\gamma_{43})/2,$$

$$\Gamma_{24}^{d}=\Gamma_{24}-(\gamma_{12}+\gamma_{32}+\gamma_{42}+\gamma_{14}+\gamma_{24}+\gamma_{34})/2,$$

$$\Gamma_{34}^{d}=\Gamma_{23}-(\gamma_{13}+\gamma_{23}+\gamma_{43}+\gamma_{14}+\gamma_{24}+\gamma_{34})/2. \quad (50)$$

These dephasing rates must satisfy the necessary and sufficient conditions (28) for complete positivity, and in particular the inequality constraints (34) and (35). If the data for a given system do not appear to satisfy these conditions then (unless the data are unreliable) it should be assumed that the system cannot be *properly* modeled as a four-level system subject to population and phase relaxation. As mentioned in the Introduction, such models are quite common and may still be adequate for some purposes but can lead to nonphysical results such as states with negative eigenvalues, etc.

If the dephasing rates do satisfy the necessary constraints then a physically valid representation of the relaxation superoperator L_D for the system in terms of the observed relaxation rates is

$$L_D[\rho(t)]=\sum_{m\neq n}\gamma_{mn}L_{mn}^{p}[\rho(t)]+\sum_{m,n=1}^{3}b_{mn}L_{mn}^{d}[\rho(t)], \quad (51)$$

where the elementary relaxation terms are

$$2L_{mn}^{p}[\rho(t)]=[\![V_{(m,n)}\rho(t),V_{(m,n)}^{\dagger}]\!]+[\![V_{(m,n)},\rho(t)V_{(m,n)}^{\dagger}]\!],$$

$$2L_{mn}^{d}[\rho(t)]=[\![V'_{(m,m)}\rho(t),V'_{(n,n)}]\!]+[\![V'_{(m,m)},\rho(t)V'_{(n,n)}]\!] \quad (52)$$

with $V'_{(m,n)}$ as defined in Eq. (30), $V_{(m,n)}=V'_{(m,n)}$ for $m\neq n$, and coefficients b_{mn} as in Eq. (31). Note that—unlike in the three-level case—we chose *not* to diagonalize the dephasing superoperator since the general expressions are quite complicated and do not confer a significant computational advantage.

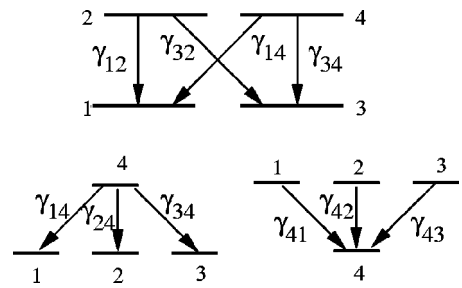

FIG. 3. Four-state atoms: transition between doubly degenerate energy levels (top), tripod system (bottom left), and inverted tripod (bottom right) with arrows indicating population decay pathways.

1. Transition between doubly degenerate levels

The results of the last paragraph apply, for instance, to a system consisting of two doubly degenerate energy levels subject to population relaxation as shown in Fig. 3 and general phase relaxation. The decay-induced decoherence rates according to Eq. (19) are

$$\Gamma_{12}^{p}=\Gamma_{23}^{p}=(\gamma_{12}+\gamma_{32})/2,$$

$$\Gamma_{14}^{p}=\Gamma_{34}^{p}=(\gamma_{14}+\gamma_{34})/2,$$

$$\Gamma_{24}^{p}=(\gamma_{12}+\gamma_{32}+\gamma_{14}+\gamma_{34})/2,$$

$$\Gamma_{13}^{p}=0. \quad (53)$$

Thus, if all decoherence is the result of population relaxation processes then the transition between the ground states remains decoherence-free. However, if there is pure dephasing then (36) implies $(b-c)^2\leqslant 4xy$ for $a=x+y$ with $x=\Gamma_{13}^{d}$, $y=\Gamma_{24}^{d}$, $b=\Gamma_{12}^{d}+\Gamma_{34}^{d}$ $c=\Gamma_{14}^{d}+\Gamma_{23}^{d}$. Thus, $x=\Gamma_{13}^{d}=0$ is possible if and only if $b=c$. Since $\Gamma_{14}^{p}+\Gamma_{23}^{p}=\Gamma_{12}^{p}+\Gamma_{34}^{p}$ according to Eq. (53), this is equivalent to $\Gamma_{12}+\Gamma_{34}=\Gamma_{14}+\Gamma_{23}$. Conversely, if $\Gamma_{12}+\Gamma_{34}\neq\Gamma_{14}+\Gamma_{23}$ then $b\neq c$, and we have $0<|b-c|<x+y$ and $0<(b-c)^2\leqslant 4xy$, which implies $x>0$, i.e., $\Gamma_{13}^{d}>0$.

Now suppose both excited states have the same T_1 relaxation time, i.e., the same spontaneous emission rate γ, and the relative probabilities for the possible decay pathways are given by the absolute values of the Clebsch-Gordan coefficients of the transition. Then $\gamma_{12}=\gamma_{34}=\gamma/3$ and $\gamma_{32}=\gamma_{14}=2\gamma/3$, and the decay-induced decoherence rates are $\Gamma_{13}^{p}=0$, $\Gamma_{12}^{p}=\Gamma_{23}^{p}=\Gamma_{14}^{p}=\Gamma_{34}^{p}=\gamma/2$, and $\Gamma_{24}^{p}=\gamma$, as one would reasonably expect.

Furthermore, if the dephasing rates satisfy $\Gamma_{12}^{d}=\Gamma_{34}^{d}=:\Gamma_{1}^{d}$ and $\Gamma_{14}^{d}=\Gamma_{23}^{d}=:\Gamma_{2}^{d}$, as one might expect due to symmetry for a typical system, then (36) implies especially that $(\Gamma_{1}^{d}-\Gamma_{2}^{d})^2\leqslant 4\Gamma_{13}^{d}\Gamma_{24}^{d}$. Thus, if we have $\Gamma_{13}^{d}=0$ then we must also have $\Gamma_{1}^{d}=\Gamma_{2}^{d}$, and conversely, if $\Gamma_{1}^{d}\neq\Gamma_{2}^{d}$ then $\Gamma_{13}^{d}>0$, i.e., the transition between the two ground states can remain decoherence-free only if $\Gamma_{1}^{d}=\Gamma_{2}^{d}$. This observation may seem trivial but it might be a convenient way of ascertaining if the transition between the ground states is decoherence-free or

not by simply measuring the decoherence of the transitions between the ground and excited states, and the decay rates of the excited states.

Moreover, if $\Gamma_{13}=0$, then we must have $\Gamma^d_{12}=\Gamma^d_{14}=\Gamma^d_{23}=\Gamma^d_{34}=:\Gamma^d$ according to our previous observations. If $\Gamma^d=0$ as well then $\Gamma^d_{24}=0$ due to (36) and we have $L^d_D[\rho(t)]=0$, i.e., no dephasing takes place. Otherwise, setting $\Gamma^d_{24}=\alpha\Gamma^d$ leads to the simplified relaxation superoperator

$$\begin{aligned} L_D[\rho(t)] = {} & (\gamma/3)\{L^p_{12}[\rho(t)] + L^p_{34}[\rho(t)]\} + (2\gamma/3)\{L^p_{32}[\rho(t)] \\ & + L^p_{14}[\rho(t)]\} + (4-\alpha)\Gamma^d([\![A_1\rho(t),A_1]\!] \\ & + [\![A_1,\rho(t)A_1]\!])/4 + \alpha\Gamma^d([\![A_2\rho(t),A_2]\!] \\ & + [\![A_2,\rho(t)A_2]\!])/2 \end{aligned} \tag{54}$$

with $A_1=V'_{(1,1)}$, $A_2=(-V'_{(2,2)}+V'_{(3,3)})/\sqrt{2}$ and $L^p_{mn}[\rho(t)]$ as defined in Eq. (43), and $V'_{(m,n)}$ as defined in Eq. (30). Again positivity requires $0\leqslant\alpha\leqslant 4$, i.e., $0\leqslant\Gamma^d_{24}\leqslant 4\Gamma^d$, consistent with our previous observations, and thus provides an upper bound of $4\Gamma^d+\gamma$ on the total decoherence of the transition between the upper levels.

2. Tripod and inverted tripod systems

Another common type of four-level system is a tripod system, i.e., a transition between a triply degenerate ground state and a nondegenerate excited state $|4\rangle$. With population relaxation due to spontaneous emission as indicated in Fig. 3, the decay-induced decoherence rates according to Eq. (19) are

$$\Gamma^p_{12}=\Gamma^p_{13}=\Gamma^p_{23}=0,$$

$$\Gamma^p_{14}=\Gamma^p_{24}=\Gamma^p_{34}=(\gamma_{14}+\gamma_{24}+\gamma_{34})/2. \tag{55}$$

Assuming that the lifetime of the excited states is γ^{-1} and all decay pathways are equally probable, we obtain $\gamma_{14}=\gamma_{24}=\gamma_{34}=\gamma/3$ and $\Gamma^p_{14}=\Gamma^p_{24}=\Gamma^p_{34}=\gamma/2$, as well as

$$L^p_D[\rho(t)] = (\gamma/3)\{\mathcal{L}^p_{14}[\rho(t)] + \mathcal{L}^p_{24}[\rho(t)] + \mathcal{L}^p_{34}[\rho(t)]\} \tag{56}$$

with $L^p_{mn}[\rho(t)]$ as defined in Eq. (43).

For comparison, the decay-induced decoherence rates (19) for an inverted tripod, i.e., a transition between a nondegenerate ground state $|4\rangle$ and a threefold-degenerate excited state with population relaxation as indicated in Fig. 3 are

$$\Gamma^p_{12}=\frac{1}{2}(\gamma_{41}+\gamma_{42}),$$

$$\Gamma^p_{13}=\frac{1}{2}(\gamma_{41}+\gamma_{43}),$$

$$\Gamma^p_{23}=\frac{1}{2}(\gamma_{42}+\gamma_{43}),$$

$$\Gamma^p_{14}=\frac{1}{2}\gamma_{41},$$

$$\Gamma^p_{24}=\frac{1}{2}\gamma_{42},$$

$$\Gamma^p_{34}=\frac{1}{2}\gamma_{43}, \tag{57}$$

and assuming that the lifetime of the excited states is γ^{-1} we thus obtain $\gamma_{41}=\gamma_{42}=\gamma_{43}=\gamma$ and $\Gamma^p_{14}=\Gamma^p_{24}=\Gamma^p_{34}=\gamma/2$, $\Gamma^p_{12}=\Gamma^p_{23}=\Gamma^p_{13}=\gamma$, as well as

$$L^p_D[\rho(t)] = \gamma\{\mathcal{L}^p_{41}[\rho(t)] + \mathcal{L}^p_{42}[\rho(t)] + \mathcal{L}^p_{43}[\rho(t)]\} \tag{58}$$

with $L^p_{mn}[\rho(t)]$ as defined in Eq. (43).

Interestingly, the decay-induced decoherence rates for transitions between the upper and lower sublevels are the same for both systems. In the absence of pure dephasing, the only difference between the two cases is that the degenerate subspace remains decoherence-free for the tripod system, while the decoherence rates for the inverted tripod are equal to the spontaneous emission rate γ for the upper levels. This basic situation does not change very much if we add pure dephasing since the tripod and inverted tripod systems are equivalent as far as pure dephasing is concerned. However, there will be additional constraints, and we shall in particular study the case $\Gamma^d_{14}=\Gamma^d_{24}=\Gamma^d_{34}=\Gamma^d$ and $\Gamma^d_{12}=\Gamma^d_{23}=\Gamma^d_2$, which one might expect to occur in many systems for reasons of symmetry.

If $\Gamma^d=0$ then the necessary conditions (35) can be satisfied only for $\Gamma^d_2=\Gamma^d_{13}=0$, i.e., if there is no pure dephasing for transitions between the upper and lower sublevels then all pure dephasing rates are zero and all decoherence in the system must be due to population relaxation. Hence, $L^d_D[\rho(t)]=0$. Otherwise, set $\Gamma^d_2=\alpha\Gamma^d$ and $\Gamma^d_{13}=\beta\Gamma^d$. Inserting these values into the coefficient matrix b' [Eq. (31)] allows us to directly derive necessary and sufficient conditions for positivity of this matrix by computing its eigenvalues:

$$\det(b'/\Gamma^d-\lambda I) = (\beta-\lambda)(\lambda^2-p\lambda+q),$$

where $p=3+2\alpha-\beta$ and $q=(4\alpha-\alpha^2-\beta)/2$, shows immediately that the eigenvalues of b' are $\lambda_1=\Gamma^d_{13}$, and $\lambda_{2/3}=(p\pm\sqrt{p^2-4q})/2$. Hence, necessary and sufficient conditions for positive semidefiniteness of b' are $\Gamma^d_{13}\geqslant 0$, $p\geqslant 0$, and $q\geqslant 0$, and the dephasing superoperator can be written as

$$\begin{aligned} L^d_D[\rho(t)] = {} & \Gamma^d_{13}\{[\![A_1\rho(t),A_1]\!] + [\![A_1,\rho(t)A_1]\!]\}/2 + \lambda_2\{[\![A_2\rho(t),A_2]\!] \\ & + [\![A_2,\rho(t)A_2]\!]\}/2 + \lambda_3\{[\![A_3\rho(t),A_3]\!] \\ & + [\![A_3,\rho(t)A_3]\!]\}/2, \end{aligned} \tag{59}$$

where we have

$$A_1=(V'_{(2,2)}-V'_{(3,3)})/\sqrt{2},$$

$$A_2=[4\tilde{\alpha}V'_{(1,1)}+(\tilde{\beta}+x)V'_{(2,2)}-(\tilde{\beta}+x)V'_{(3,3)}]/(2x),$$

$$A_3=[4\tilde{\alpha}V'_{(1,1)}+(\tilde{\beta}-x)V'_{(2,2)}-(\tilde{\beta}-x)V'_{(3,3)}]/(2x)$$

with $V'_{(m,n)}$ as defined in Eq. (30), and $\tilde{\alpha}=\alpha-1$, $\tilde{\beta}=\beta-1$, and $x=\sqrt{8(\alpha-1)^2+(\beta-1)^2}$. Furthermore, $p\geqslant 0$ implies $\beta\leqslant 3$

$+2\alpha$ and $q \geqslant 0$ is equivalent to $\alpha(4-\alpha) \geqslant \beta \geqslant 0$ and hence implies $0 \leqslant \alpha, \beta \leqslant 4$.

VIII. CONCLUSIONS

Starting with very basic assumptions we defined a simple yet general relaxation superoperator, which should be adequate to describe a wide variety of open systems not too strongly coupled to their environment, solely in terms of experimentally observable quantities such as the population relaxation and decoherence rates of the system, without imposing any restrictions on the types of population and phase relaxation that can occur.

The advantage of a relaxation superoperator thus defined is that it can describe the observed dissipative dynamics of the system in principle as accurately as we can measure the relaxation rates. Unfortunately, however, there are several problems with this approach. One is that it can lead to relaxation superoperators that do not preserve complete or even simple positivity, as we have explicitly shown for several examples. Since any violation of positivity effectively means negative or even nonreal probabilities, this is a serious problem.

To avoid such problems one must impose constraints on the relaxation rates. We have analyzed the nature of these basic constraints by expressing our relaxation superoperator in the standard form for dissipative generators of quantum dynamical semigroups derived by Gorini, Kossakowski, and Sudarshan. We have also shown that it is possible to decompose our generic relaxation superoperator into two distinct parts associated with population relaxation and pure dephasing processes, respectively, and that the coefficients of the Kossakoswki generators for the population relaxation part can be identified (usually uniquely) with the observed population relaxation rates, the only restriction being the obvious one that the decay rates be non-negative. Most importantly, the expressions we obtain for the decoherence rates induced by population relaxation agree with similar expressions found in the literature.

However, population relaxation is usually not the only source of decoherence. To account for other sources of decoherence, we have introduced pure dephasing rates for each transition by subtracting the decoherence induced by population relaxation processes from the observed overall decoherence rates. These pure dephasing rates define the pure-phase-relaxation superoperator, and we express the coefficients of the Kossakoswki generators for this part of the relaxation superoperator explicitly in terms of these pure dephasing rates for three- and four-level systems. These expressions, unlike the general expressions for the coefficients of the population relaxation superoperator, are more complicated, and the requirement of complete positivity results in nontrivial constraints on the dephasing rates, which we have analyzed specifically for three- and four-level systems, although the same type of analysis can be performed for systems of higher dimension.

Finally, we have applied these general results to study their concrete implications for several simple but commonly used three- and four-level model systems such as Λ and V systems, tripod and inverted tripod systems, and transitions between doubly degenerate energy levels. In each case we have attempted to make *concrete* predictions about inequality constraints and correlations of the decoherence rates demanded by the requirement of complete positivity, which are *experimentally verifiable*. Such experimental tests of the constraints could be useful in various ways. Confirmation of the correlations would vindicate the semigroup description of the dynamics. On the other hand, violation of the constraints required by complete positivity would suggest that our model of the system is not really adequate to capture its real dynamics although it may still be useful for certain purposes.

IX. EPILOGUE: POSITIVE MATRICES

There has been a great deal of mathematical work on the properties of positive matrices, and publications such as [24–26] appear relevant to our problem at first glance. However, they really address other problems.

Man [24], for example, studies the problem of comparing positive definite symmetric matrices, in particular answering the question under what conditions $P > Q$, i.e., $P-Q$ positive, implies $P^2 > Q^2$ for positive definite symmetric matrices P and Q. Unfortunately, these results are not applicable to our problem. However, they may be relevant for issues such as the comparison of density matrices.

Savage [25] considers the space of $n \times n$ positive definite matrices X with $\det(X)=1$ under isometries $X \to AXA^T$ where $A \in SL(n,\mathbb{R})$ and shows that it has a collection of simplices preserved by the isometries and that the volume of the top-dimensional ones has a uniform upper bound. One could perhaps say that the density matrices ρ of interest to us are positive, and that the dynamical Lie group of the system provides isometries of a sort, but our density matrices are positive matrices of trace 1, which actually rules out $\det\rho = 1$, since positivity requires the eigenvalues to be non-negative and $\mathrm{Tr}(\rho)=1$ requires that the sum of these eigenvalues be 1, which implies $\det(\rho) < 1$ unless $n=1$ and $\rho=1$.

Similarly, Leighton and Newman [26] show that the number of $n \times n$ integral, triple-diagonal matrices that are unimodular, positive definite, and whose sub- and superdiagonal elements are all 1 is the Catalan number $(2n/n)/(n+1)$, which is an interesting mathematical result but not relevant to our problem since our density matrices, although positive definite, are not usually tridiagonal, and even if they were, the elements on the sub- and superdiagonals (the coherences) would have to be less than 1 for normalized density matrices.

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