Limits on the decay rate of quantum coherence and correlation

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The study and control of coherence in quantum systems is one of the most exciting recent developments in physics. Quantum coherence plays a crucial role in emerging quantum technologies as well as fundamental experiments. A major obstacle to the utilization of quantum effects is decoherence, primarily in the form of dephasing that destroys quantum coherence, and leads to effective classical behavior. We show that there are universal relationships governing dephasing that constrain the relative rates at which quantum correlations can disappear. These lead effectively to speed limits which become especially important in multipartite systems.

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I. INTRODUCTION

One of the principal distinguishing features between classical and quantum systems is the existence of quantum coherences leading to correlations that cannot be accounted for classically. For example, the phenomenon of entanglement [1] and the violation of nonlocal realism [2] are such consequences. The manipulation and preservation of such coherences are vital for tasks such as the construction of quantum information-processing devices [3], quantum communication [4], cryptography [5], and metrology [6]. It has also been suggested that quantum coherence plays a role in certain biological processes [7]. Unfortunately, the inevitable interaction of quantum systems with the environment leads to decoherence, the dominant form of which is dephasing or the disappearance of the off-diagonal elements of the density operator [8]. The rates at which these coherences decay are crucial as they determine how quickly the system approaches classicality when quantum correlations are lost. Considerable effort has been expended on understanding fundamental properties of decoherence and quantum correlations and how to protect the latter and prevent rapid decay using encodings in protected subspaces or subsystems, for example [9].

Some surprising relationships between the dephasing rates in multilevel quantum systems have been previously uncovered [10,11]. The need to preserve positivity of the density operator, or more generally the complete positivity of the superoperator [12] leads to constraints on the relative rates of dephasing. The surface of this phenomenon has only been scratched and in this work we present a general framework to elucidate the general nature of these constraints focusing on decoherence induced by pure dephasing processes. Experimental observations for many systems suitable for quantum information, from trapped ions [13] and cavity QED [14] to the solid state [15,16], reveal dephasing (T_2) times much shorter than the relaxation (T_1) times; that is, the observed decoherence rates are much greater than what can be attributed to relaxation. Therefore, although relaxation processes may be significant, for example, in determining the steady states of the system and the dynamics of relaxation to equilibrium, the rate of coherence loss will be determined by the fastest decoherence processes. The experimental results above suggest that on the time scales relevant for coherence decay most physical decoherence processes can approximated by a pure dephasing model, and we expect the constraints on the rate of coherence loss to be mostly determined by pure dephasing. An important consequence of the constraints is an effective speed limit on the decay of correlations and entanglement in multipartite systems. These limits are independent of the details of the Hamiltonian evolution or dephasing mechanisms and therefore apply to a large class of quantum systems from nuclear spins, to atoms, molecules, and quantum dots.

II. DECOHERENCE MODEL AND DEPHASING OPERATORS

The Markovian evolution of a quantum system can be described by a Lindblad master equation [17,18]. A *pure dephasing process* leaves the populations of the basis states invariant but leads to the decay of the the off-diagonal elements (coherences), as well as anomalous frequency shifts. Previous work on three-level systems found that the decay rates and frequency shifts were constrained [10,11]. Additionally, [10] gave partial results for four-level systems and showed that similar constraints exist in higher dimensions but the equations were intractable in general. Here, we present a canonical form for pure dephasing Lindblad operators that allows us to explicitly state the constraints for N-level systems and showing that they form a hierarchy of inequalities, defining a convex cone of allowed dephasing rates. The general form also allows us to invert physically observed dephasing rates to define a unique set of canonical dephasing operators, which reflect correlations in noise processes such as fluctuations in the energy levels, and may serve as a useful diagnostic tool. In multi-partite systems these constraints induce speed limits on the decay of nonlocal quantum correlations and entanglement in terms of the local dephasing rates.

We begin with the result that pure dephasing of an N-level system may be modeled by a diagonal Hamiltonian $H = \mathrm{diag}(\lambda_n)$ and a canoncial set of $N-1$ or fewer diagonal

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Lindblad operators $\{V_k\}_{k=1}^{N-1}$ of the special form

$$V_k = \mathrm{diag}(\underbrace{0\ldots0}_{k}, a_{k+1}^{(k)}, \ldots, a_N^{(k)}), \tag{1}$$

where the nonzero diagonal elements $a_n^{(k)}$ can be complex except for the first nonzero element, $a_{k+1}^{(k)}$, which is set to be real and positive. The density operator elements evolve as

$$\rho_{mn}(t) = e^{-t(i\omega_{mn}+\Gamma_{mn})}\rho_{mn}(0), \tag{2}$$

with effective frequencies given by $\omega_{mn} = \lambda_m - \lambda_n + \Delta\omega_{mn}$ and dephasing-induced frequency shifts and decoherence rates,

$$\Delta\omega_{mn} = -\sum_k \mathrm{Im}\big(a_m^{(k)} a_n^{(k)*}\big), \tag{3a}$$

$$\Gamma_{mn} = \frac{1}{2}\sum_k \big(\big|a_m^{(k)}\big|^2 + \big|a_n^{(k)}\big|^2\big) - \mathrm{Re}\big(a_m^{(k)} a_n^{(k)*}\big). \tag{3b}$$

The populations are constant as $\omega_{nn} = \Gamma_{nn} = 0$ and the off-diagonal elements decay with the damping rate Γ_{mn},

$$|\rho_{mn}(t)| = e^{-t\Gamma_{mn}}|\rho_{mn}(0)|. \tag{4}$$

If all $a_n^{(k)}$ are real then the expressions simplify, $\Gamma_{mn} = \frac{1}{2}\sum_k (a_m^{(k)} - a_n^{(k)})^2$, and there are no frequency shifts, $\Delta\omega_{mn} = 0$.

As shown in Appendix A, any set of pure dephasing Lindblad operators can be transformed to this form leaving the total superoperator unchanged. The key idea is to recombine the Lindblad operators to form a new set without changing the observable dynamics. Choosing the special form of the operators [Eq. (1)] reduces an arbitrary number of parameters, specified by the nonzero elements of an arbitrary set of dephasing operators, to $N(N-1)/2$ parameters. The number of free parameters matches the number of dephasing rates Γ_{mn} and frequency shifts $\Delta\omega_{mn}$ for an N-level system.

III. INVERTING DEPHASING RATES

In this way we can determine a set of standard operators consistent with experimentally observed dephasing rates Γ_{mn} and frequencies ω_{mn} (or frequency shifts). The inversion process relies on the fact that the dephasing rates involving the first $k+1$ levels depend only on the first k dephasing operators; that is, Γ_{12} determines the first nonzero element of V_1, which together with $\{\Gamma_{13}, \Gamma_{23}, \Delta\omega_{23}\}$ then determines a further three real parameters, and so forth. Hence, it is a simple matter of iteratively solving a nested set of quadratic equations, as detailed in Appendix B.

A set of constraints on the allowed dephasing rates and frequency shifts naturally arises in the inversion process. This takes the form of $N-1$ inequalities involving the first N levels,

$$2\Gamma_{1n} - \sum_{\ell=1}^{n-2}\big|a_\ell^{(n)}\big|^2 \geqslant 0, \quad \forall\, n = 2,\ldots,N, \tag{5}$$

where the $a_\ell^{(n)}$ can be expanded in terms of the $\Gamma_{mn'}$ with $m,n' \leqslant n$. These inequalities form a convex cone of allowed dephasing rates whose boundary is formed by "hypersurfaces"

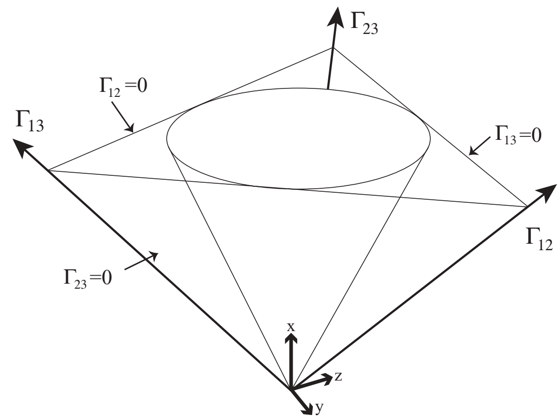

FIG. 1. Convex cone of allowed dephasing rates for $N = 3$ and real dephasing, that is, $\Delta\omega_{23} = 0$. The axes are $x = (\Gamma_{12} + \Gamma_{23} + \Gamma_{13})/\sqrt{3}$, $y = (\Gamma_{13} - \Gamma_{23})/\sqrt{2}$ and $z = \sqrt{\frac{2}{3}}(\Gamma_{12} - \frac{1}{\sqrt{2}}(\Gamma_{13} + \Gamma_{23}))$ and the constraint equation becomes $x^2/2 \geqslant y^2 + z^2$, which defines a circular cone in the positive octant of in the parameter space of $\{\Gamma_{12}, \Gamma_{23}, \Gamma_{13}\}$, tangential to the $\Gamma_{12}\Gamma_{23}$, $\Gamma_{23}\Gamma_{13}$, and $\Gamma_{13}\Gamma_{12}$ planes.

defined by $a_{k-1}^{(k)} = 0$ for some $k > 1$. For $N = 3$ there is only a single constraint equation and the convex cone of allowed dephasing rates can be visualized as shown in Fig. 1.

A symmetric form of the constraints is possible, for example, for $N = 3$,

$$2(\Gamma_{12}\Gamma_{23} + \Gamma_{23}\Gamma_{13} + \Gamma_{12}\Gamma_{13}) \geqslant \Gamma_{12}^2 + \Gamma_{23}^2 + \Gamma_{13}^2 + \Delta\omega_{23}^2,$$

which reduces to Eq. (25) in [10] if $\Delta\omega_{23} = 0$. However, as the number of dephasing rates grows as $N(N-1)/2$ and the inequalities involve products of $(N-1)\Gamma_{mn}$, there is a combinatorial explosion in the number of terms in the constraints, which is why previous attempts to obtain a general form for the constraints failed. For example, the four-level constraint contains 22 terms and the five-level constraint contains 130 terms.

IV. SPEED LIMITS FOR ENTANGLEMENT DECAY

The constraints for the decoherence rates and frequency shifts have important implications for a wide range of physical, chemical, and biological systems where phase relaxation is the dominant process. A consequence is the imposition of speed limits on the relative rates at which coherences can decay, especially in multipartite systems where entanglement decay is strictly bounded above by the single qubit dephasing rates. Though the Markovian condition constrains the temporal correlations in the noise, noise can be spatially correlated and the dephasing rates can be of a nonlocal form in general.

Let us start with two qubits where we label the basis states by $|1\rangle = |00\rangle$, $|2\rangle = |01\rangle$, $|3\rangle = |10\rangle$, and $|4\rangle = |11\rangle$. Assuming that both qubits have the same local dephasing rate, that is, $\Gamma = \Gamma_{12} = \Gamma_{13} = \Gamma_{24} = \Gamma_{34}$, then the allowed decoherence rates for the nonlocal coherences Γ_{14} and Γ_{23} are determined by Γ. The first nontrivial constraint $(a_2^{(3)})^2 \geqslant 0$ gives $0 \leqslant \Gamma_{23} \leqslant 4\Gamma$. The second constraint $(a_3^{(4)})^2 \geqslant 0$

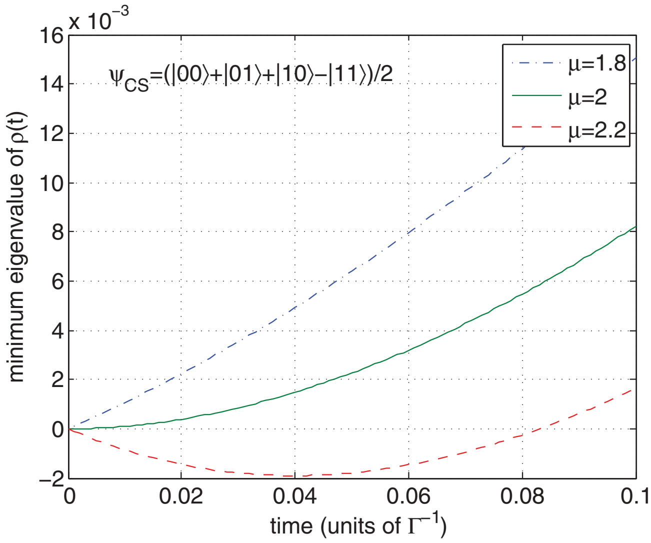

FIG. 2. (Color online) Constraint violation leads to nonphysical states. A plot of the minimum eigenvalue of $\rho(t)$ starting with $\rho(0) = |\Psi_{CS}\rangle\langle\Psi_{CS}|$ subject to pure dephasing ($H = 0$) with $\Gamma_{23} = \Gamma_{14} = \mu\Gamma$ for different values of μ shows the emergence of negative eigenvalues for $\mu > 2$.

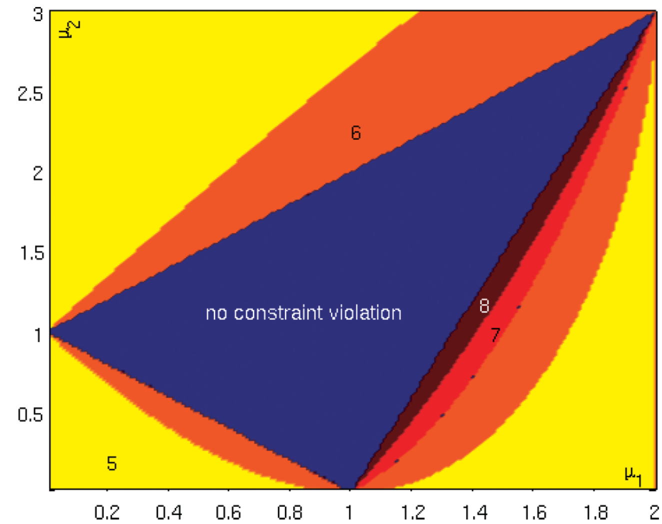

FIG. 3. (Color online) Constraint violation map for three qubits with local dephasing rates Γ, two-qubit dephasing rates $\mu_1\Gamma$, and three-qubit dephasing rates $\mu_2\Gamma$. For $0 \leqslant \mu_1 \leqslant 2$ the first four of the eight constraints are satisfied but additional constraints may be violated; for example, in the yellow region, constraint 5 is violated. Additional constraints further restrict the set of allowed rates.

leads to $\Gamma_{23} + \Gamma_{14} \leqslant 4\Gamma$.[1] Thus, to ensure complete positivity of the evolution, the nonlocal coherences ρ_{23} and ρ_{14} can decay at most four times as fast as the local coherences, and the sum of the nonlocal decay rates can be no more than 4Γ. If they are equal $\Gamma_{23} = \Gamma_{14} = \Gamma_e$ we obtain $\Gamma_e \leqslant 2\Gamma$, and Fig. 2 demonstrates that violation of the bound leads to violations of positivity, that is, nonphysical states.

The constraints on nonlocal coherence decay induce limits to the decay of entanglement between qubits. For example, for an array of identical noninteracting qubits, starting with the maximally entangled Bell state $|\Psi_0\rangle = \frac{1}{\sqrt{2}}(|00\rangle + |11\rangle)$, the state evolving under pure dephasing,

$$\rho_{\Psi_0}(t) = \frac{1}{2}\begin{pmatrix} 1 & 0 & 0 & e^{-t\Gamma_{14}} \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ e^{-t\Gamma_{14}} & 0 & 0 & 1 \end{pmatrix},$$

has concurrence $C(t) = e^{-t\Gamma_{14}}$ [19]; thus, $\Gamma_{14} + \Gamma_{23} \leqslant 4\Gamma$ implies that the concurrence cannot decay faster than four times the local decoherence rate Γ. Here, the decay of nonlocal coherences is not lower bounded; that is, nonlocal coherences can survive indefinitely even for finite local decay rates and in this case the entanglement is preserved if $\Gamma_{14} = 0$; that is, there is no sudden death of entanglement [21].

[1]An upper bound of 4Γ on the nonlocal dephasing time was also found in the Markovian limit for a specific exactly solvable model of phonon decoherence, in contrast to the non-Markovian regime, where much faster entanglement decay was shown to be possible [20].

Alternatively, an initial maximally entangled two-qubit cluster state $|\Psi_{CS}\rangle = \frac{1}{2}(|00\rangle + |01\rangle + |10\rangle - |11\rangle)$ decays as

$$\rho_{CS}(t) = \frac{1}{4}\begin{pmatrix} 1 & e^{-\Gamma t} & e^{-\Gamma t} & -e^{\Gamma_{14} t} \\ e^{-\Gamma t} & 1 & e^{-\Gamma_{23} t} & -e^{-\Gamma t} \\ e^{-\Gamma t} & e^{-\Gamma_{23} t} & 1 & -e^{-\Gamma t} \\ -e^{-\Gamma_{14} t} & -e^{-\Gamma t} & -e^{-\Gamma t} & -1 \end{pmatrix}.$$

Here, the entanglement may decay even if both nonlocal dephasing rates vanish, $\Gamma_{14} = \Gamma_{23} = 0$, in which case the concurrence satisfies $2C_1(t) = |e^{-\Gamma t} + 1| - |e^{-\Gamma t} - 1|$, which tends to zero as $t \to \infty$. If one of the two nonlocal concurrences is 4Γ and the other is 0, for example, $\Gamma_{14} = 4\Gamma$, $\Gamma_{23} = 0$, the concurrence similarly decays asymptotically but faster. When the nonlocal coherences decay at the same rate $\Gamma_{14} = \Gamma_{23} = 2\Gamma$ we have $C_2(t) = \max\{0, \frac{1}{2}(2e^{-\Gamma t} + e^{-2\Gamma t} - 1)\}$, and the concurrence vanishes when $2e^{-\Gamma t} + e^{-2\Gamma t} - 1 = 0$, that is, $t_* = -\Gamma^{-1}\ln(\sqrt{2} - 1) \approx 0.383\Gamma^{-1}$, that is, we observe sudden death of entanglement.

Extending this, given n qubits with the same local dephasing rate Γ we can simply apply the results above to any subsystem consisting of two qubits; that is, the entanglement between any two qubits in the system cannot decay faster than 2Γ. As the number of qubits grows, there are more constraints so in practice the rate of entanglement decay between any two qubits would be even more restricted. For example, for a three-qubit system ($N = 2^3 = 8$), assuming the local dephasing rate for each qubit is Γ, and the dephasing rate involving two- and three-qubit transition terms are $\mu_1\Gamma$ and $\mu_2\Gamma$ respectively, there are eight constraints restricting the allowed values for μ_1 and μ_2. From the constraints for the two-qubit system we know that $0 \leqslant \mu_1 \leqslant 2$, but Fig. 3 shows that the set of (μ_1, μ_2) that satisfies all the constraints is much smaller. Each additional constraint reduces the set of allowed dephasing rates.

V. DISCUSSION

The underlying basis for the dephasing constraints is correlation between noise acting on different energy levels of the system. The canonical dephasing operators reflect underlying physical processes with different correlation properties. For example, a canonical dephasing operator with a single nonzero element can be interpreted as the result of the fluctuation of a single energy level. Multiple nonzero diagonal entries correspond to correlated perturbations of more than one level. An example of noise correlation is magnetic field fluctuations acting on a spin-1 particle where the coupling is of the form $B_Z S_Z$. This leads to antiphase perturbations of the $S_Z = \pm 1$ levels and the canonical operator in the basis $\{|0\rangle,|1\rangle,|-1\rangle\}$ is $\propto \mathrm{diag}(0,1,-1)$. If the coupling was instead of the form $B_Z S_Z^2$, then the canonical operator would be $\propto \mathrm{diag}(0,1,1)$.

Dephasing not only leads to exponential damping of the coherences but may also produce shifts in their frequencies. Not all of these frequency perturbations can be accommodated by modifying the system Hamiltonian. In general, the residual shifts are intrinsic to the decoherence processes. While pure damping can be generated by phase diffusion due to random drift of the energy levels (Wiener-Levy process) [22], the frequency shifts can be caused by phase kicks or discrete random phase jumps with a Poissonian arrival time [23]. Phase kicks can occur by collisional processes in gases, for example, whereas phase diffusion can be generated by white noise acting on the energy levels. It is possible to derive dephasing constraints from physical models of the noise directly, though deriving the general multilevel constraints is considerably more difficult than the methods shown here. However, once the observed dephasing rates have been decomposed into their corresponding canonical set of dephasing operators, we can assign physical mechanisms by which they occur, and hence perform system diagnostics or analysis. The ability to identify sources of dephasing will be vital in producing coherent quantum devices and improving their performance.

In the context of multipartite systems, the constraints we have derived have implications for the preservation of nonlocal correlations. As the number of parties increases, the decay of the nonlocal coherences becomes constrained even more by the local dephasing rates. This reflects the general robustness of the nonlocal correlations in multipartite systems [24]. Conversely, there are suggestions that dephasing can play a positive role in biological processes [25,26], where it has been mooted to enhance the transport of energy in networks such as photosynthetic harvesting complexes. Measurement and analysis of the dephasing may illuminate these processes and lead to better energy collection devices.

Although as we have seen, relaxation processes dominated by dephasing play a crucial role in determining coherence and entanglement decay in many systems, it is an interesting question whether simple canonical forms can be derived for other types of relaxation processes, and whether these permit a systematic inversion that leads to a canonical form of the constraints. This may be useful in those cases where dephasing processes do not dominate, such as in optical phonons [27] and transmon qubits [28].

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APPENDIX A: DERIVATION OF CANONICAL DEPHASING OPERATORS

We start with the Lindblad master equation (LME) for Markovian open quantum system evolution,

$$\dot{\rho}(t) = -i(H\rho(t) - \rho(t)H) + \mathfrak{L}_D(\rho(t)), \tag{A1}$$

where $\rho(t)$ is the density operator describing the system state (defined on the system Hilbert space $\mathcal{H}_s$), H is an effective Hamiltonian H, and the superoperator $\mathfrak{L}_D(\rho)$ takes the form $\mathfrak{L}_D(\rho) = \sum_k \mathcal{D}[V_k]\rho$ with [17,18]

$$\mathcal{D}[V_k]\rho = V_k \rho V_k^\dagger - \tfrac{1}{2}(V_k^\dagger V_k \rho + \rho V_k^\dagger V_k) \tag{A2}$$

for operators V_k on $\mathcal{H}_S$.

The operators (H, V_k) define a pure dephasing process with respect to the basis $\mathfrak{B} = \{|n\rangle\}_{n=1}^N$ if and only if H and all V_k are simultaneously diagonal with respect to $\mathfrak{B}$; that is, we have

$$H = \sum_n \lambda_n \Pi_n = \mathrm{diag}(\lambda_n), \quad \lambda_n \in \mathbb{R}, \tag{A3a}$$

$$V_k = \sum_n \gamma_{nk} \Pi_n = \mathrm{diag}(\gamma_{nk}). \tag{A3b}$$

This is easy to see since by definition of a pure dephasing process the populations of the basis states remain constant, and thus each basis state $|n\rangle$ is a steady state of the system. This is possible only if the subspace spanned by each basis state $|n\rangle$ is V_k invariant for all V_k [29]. This shows that all V_k must be diagonal in the chosen basis. Since Π_n is diagonal and diagonal operators commute we have $\mathcal{D}[V_k](\Pi_n) = 0$ for all n and all k. As $|n\rangle$ is a steady state, that is, $\dot{\Pi}_n(t) = 0$, it also follows that $-iH\Pi_n + i\Pi_n H = 0$ for all n. Inserting this into the general form of the LME (A1) gives the explicit equation

$$\dot{\rho}_{mn}(t) = -(i\omega_{mn} + \Gamma_{mn})\,\rho_{mn}(t) \tag{A4}$$

for the evolution of the matrix elements $\rho_{mn} = \langle m|\rho|n\rangle$ of the density operator, or in integral form (2) with frequencies $\omega_{mn} = \lambda_m - \lambda_n + \Delta\omega_{mn}$ and dephasing-induced frequency shifts and decoherence rates given by (3).

Any set of diagonal Lindblad operators $\{V_k\}$ generates pure dephasing dynamics but the set of Lindblad operators $\{V_k\}$ generating a certain dynamical evolution is not unique. In particular, we have *unitary invariance*, that is, given any set of Lindblad operators $\{V_k\}$, the set of operators $\{W_j\}$ defined by

$$W_j = \sum_k u_{jk} V_k, \tag{A5}$$

where u_{jk} are elements of a unitary matrix, generates the same dynamics as $\sum_k \mathcal{D}[W_k]\rho = \sum_k \mathcal{D}[V_k]\rho$. Furthermore, *adding multiples of the identity*, $V_k \to V_k + \alpha\mathbb{I}$, to a Lindblad operator V_k only changes the effective Hamiltonian

$$\mathcal{D}[V_k + \alpha\mathbb{I}]\rho = \tfrac{1}{2}[\alpha V_k^\dagger - \alpha^* V_k, \rho(t)] + \mathcal{D}[V_k]\rho, \tag{A6}$$

and thus the dynamics is unchanged if we replace V_k with $V_k + \alpha\mathbb{I}$ and H with $H + \frac{i}{2}(\alpha V_k^\dagger - \alpha^* V_k)$.

The invariance of the dephasing dynamics under the two "gauge transformations" (A5) and (A6) allows us to transform any set of dephasing operators $\{V_\ell\}$ into an equivalent set of dephasing operators in canonical form defined in Eq. (1), which yield the same observable dephasing rates $\{\Gamma_{mn}\}$ and dephasing shifts $\{\Delta\omega_{mn}\}$, using the algorithm given here.

$V \leftarrow$ CanonicDephasing (W)
Calculate Canonical Dephasing Operators

In: W Matrix $(N \times K)$, kth column equals diagonal elements of Lindblad operator V_k

Out: V Lower triagonal matrix, columns equals diagonal elements of canonical V_k

```
R ← Number of rows of W
C ← Number of columns of W
V ← W−ONES(R,1) * W[1,:]
k ← 1  // Running column index
for r ← 2,...,N
    i1 ← Index 1st nonzero entry of V[r,k:C]
    i1 ← i1 + k − 1  // shift index
    while more than one element of V[r,k:C] non-zero
        i2 ← Index 2nd nonzero entry of V[r,k:C]
        i2 ← i2 + k − 1  // shift index
        r1 ← |V[r,i1]|, r2 ← |V[r,i2]|
        φ1 ← PHASE(V[r,i1]), φ2 ← PHASE(V[r,i2])
        n_c ← sqrt(r1^2 + r2^2)
        V[:,i1] ← (r1 e^{+iφ2} V[:,i1] + r2 e^{+iφ1} V[:,i2])/n_c
        V[:,i2] ← (r2 e^{−iφ1} V[:,i1] − r1 e^{−iφ2} V[:,i2])/n_c
    if V[r,k:C] has non-zero entries
        k0 ← Index of 1st non-zero entry
        k0 ← k0 + k − 1
        V ← Swap columns k and k0 of V
        k ← k + 1
V ← Remove 0 columns of V, apply phase corrections
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The process is constructive and, using the notation a_{nk} instead of $a_n^{(k)}$, the key steps can be described as follows.

(1) Using (A6) we ensure that $a_{1k} = 0$ for all V_k, modifying the Hamiltonian by

$$\Delta H = \frac{i}{2}\sum_k a_{1k} V_k^\dagger - a_{1k}^* V_k \tag{A7}$$

as necessary.

(2) We replace the Lindblad operators $V_1 = \mathrm{diag}(0, a_{21}, a_{31}, \ldots)$ and $V_2 = \mathrm{diag}(0, a_{22}, b_{32}, \ldots)$ with $a_{21} = r_{21}e^{i\phi_{21}}$ and $a_{22} = r_{22}e^{i\phi_{22}}$ by $\{W_1, W_2\}$ with

$$W_1 = u_{11}V_1 + u_{12}V_2 = \mathrm{diag}(0, c, \ldots), \tag{A8a}$$

$$W_2 = u_{21}V_1 + u_{22}V_2 = \mathrm{diag}(0, 0, *, \ldots), \tag{A8b}$$

with the unitary coefficient matrix

$$u = \frac{1}{c}\begin{pmatrix} r_{21}e^{i\phi_{22}} & r_{22}e^{i\phi_{21}} \\ r_{22}e^{-i\phi_{21}} & -r_{21}e^{-i\phi_{22}} \end{pmatrix} \tag{A9}$$

and $c = \sqrt{r_{21}^2 + r_{22}^2}$, which is dynamically equivalent to $\{V_1, V_2\}$ due to (A5).

This result allows us to reduce an arbitrary number of parameters, specified by the nonzero elements of a general set $\{V_k\}$ of dephasing operators to $N(N-1)/2$ parameters in the canonical form. Note that the number of free parameters matches the number of dephasing rates Γ_{mn} for an N-level system. The procedure will produce a set of canonical dephasing operators that reproduce the observed dephasing rates and shifts provided that these satisfy the positivity constraints. Furthermore, if the observed dephasing rates and shifts lead to constraint violations these will be detected and flagged, and this information can be used to further investigate if the violations can be explained in terms of uncertainty in the observed data, for example, due to noise, or if they are indicative of processes that would invalidate the Markovian dephasing assumption.

We note that if one has the usual Kossakowski form of Markovian evolution [18], we can "diagonalize" the sets of operators to arrive at a Lindblad form [17] where the decoherence operators are orthogonal and traceless. However, this standard form is not convenient for inversion, nor does it give much physical insight into the possible processes leading to dephasing. The canonical form Eq. (1) decomposes the dephasing into operators representing correlated level perturbations of orders 1 to $N-1$.

APPENDIX B: CONSTRAINTS FOR MULTIPARTITE SYSTEMS

The constraints derived on the decay rates of nonlocal coherences are based on the assumption that the dominant sources of decoherence are pure dephasing processes. For many experimental systems of interest it has been found that T_2 times are much shorter than the T_1-relaxation times—supporting this assumption—even for systems involving many qubits. For example, work on 14-qubit ion entanglement [13] has shown decoherence to be dominated by dephasing (T_2 100 ms for a single qubit), in this case nonlocal dominated by long-wavelength background field inhomogeneities, despite one of the qubit levels being metastable with a lifetime of only 1 s. Pure dephasing by definition implies the existence of a Lindblad relaxation operator that commutes with the Hamiltonian, or equivalently, that decoherence takes place in the eigenbasis of the Hamiltonian. One may ask what this means for a multipartite system such as a qubit register.

For qubits far removed from each other, in a quantum communications setting or distributed quantum computer, any local noise which is purely dephasing will also produce global dephasing. More generally, for a qubit register consisting of noninteracting qubits the intrinsic system Hamiltonian H is the sum of the single qubit system Hamiltonians, the computational basis states $|00\ldots\rangle$, $|100\ldots\rangle$, etc., are eigenstates of H, and any dephasing operator V that is a sum or product of local dephasing operators $\sigma_z^{(n)}$ acting on the nth qubit commutes with the Hamiltonian. Note that ideal quantum registers are generally assumed to consist of qubits that are noninteracting except when a two-qubit control Hamiltonian is applied, and these interactions are switched off for most qubits most of the time. For instance, in 14-ion example above the coupling between ions is switched off once they are entangled during the delay period in the Ramsey sequence and during this time the dephasing operator commutes with the Hamiltonian. Thus, the dephasing rates can be interpreted in terms of local and nonlocal dephasing, and the constraints on the dephasing rates are directly applicable and induce speed limits on entanglement decay in this case.

If there are always-on interactions between qubits, as is the case for nuclear spin qubits in close proximity, for example, then the Hamiltonian is no longer exactly diagonal in the computational basis and the local single qubit dephasing operators $\sigma_z^{(n)}$ generally will not commute with the intrinsic system Hamiltonian. Instead, pure dephasing in this case takes place in the eigenbasis of the Hamiltonian and involves an element of nonlocality, mediated by a common bath. For example, consider the Hamiltonian for N spins with homogeneous nearest-neighbor Heisenberg coupling,

$$H = \sum_{n=1}^{N} \sigma_z^{(n)} + J \sum_{n=1}^{N-1} \sigma_x^{(n)}\sigma_x^{(n+1)} + \sigma_y^{(n)}\sigma_y^{(n+1)} + \sigma_z^{(n)}\sigma_z^{(n+1)}. \tag{B1}$$

For $N = 2$ the Hamiltonian takes the explicit form

$$H = \begin{pmatrix} J+2 & 0 & 0 & 0 \\ 0 & -J & 2J & 0 \\ 0 & 2J & -J & 0 \\ 0 & 0 & 0 & J-2 \end{pmatrix} = UEU^\dagger, \tag{B2}$$

where

$$U = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 2^{-0.5} & -2^{-0.5} & 0 \\ 0 & 2^{-0.5} & 2^{-0.5} & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \tag{B3}$$

$$E = \begin{pmatrix} J+2 & 0 & 0 & 0 \\ 0 & J & 0 & 0 \\ 0 & 0 & -3J & 0 \\ 0 & 0 & 0 & J-2 \end{pmatrix}. \tag{B4}$$

The computational basis states $|01\rangle$ and $|10\rangle$ are eigenstates only for $J = 0$. Thus, for $J \neq 0$ a pure dephasing operator V that is diagonal, $V = \mathrm{diag}(0,a_1,a_2,a_3)$, in the eigenbasis of H, takes the form

$$\tilde{V} = UVU^\dagger = \frac{1}{2}\begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & a_1+a_2 & a_1-a_2 & 0 \\ 0 & a_1-a_2 & a_1+a_2 & 0 \\ 0 & 0 & 0 & 2a_3 \end{pmatrix} \tag{B5}$$

in the computational basis. The constraints on the dephasing rates Γ_{nk} still apply, although Γ_{nk} now refers to dephasing rate between the (nonlocal) eigenstates n and k of the Hamiltonian and thus we can no longer directly identify Γ_{12}, Γ_{13}, Γ_{24}, and Γ_{34} with local single qubit dephasing rates, for instance, although we can still relate Γ_{14} to entanglement. For example, the concurrence of the Bell state $\frac{1}{\sqrt{2}}[|00\rangle + |11\rangle]$ still decays as $e^{-\Gamma_{14}t}$, while the Bell states $\frac{1}{\sqrt{2}}[|01\rangle \pm |10\rangle]$ are eigenstates of H and thus not affected by pure dephasing.

Futhermore, if the interqubit couplings are weak ($J \ll 1$) and the always-on interaction Hamiltonian is thus only a small perturbation to the local Hamiltonian then the interpretation of the dephasing rates in terms of local and nonlocal coherences is still approximately valid. This is the case in NMR experiments where the couplings between spins are very small compared to the local Hamiltonians; hence, the local eigenstates are very close to those of the entire system. Again, even in experiments involving 12 spins T_2 processes have been shown to dominate over T_1 and the evolution is well approximated as a pure dephasing process [30].

Where subsystems become close enough to interact so strongly as to perturb the eigenstates greatly, it may not be a safe assumption that the environment still interacts locally with each subsystem. For instance, for spins located close to each other in the solid state, it is likely that they will interact with the same phonon bath. The deformation potential that one sees will be highly correlated with the potential the others see. Hence, it is quite possible for the coupling to the bath to be of a collective nature yet still be diagonal in the energy eigenbasis of the system. One would have to specifically engineer a system whereby the subsystems interacted strongly but only interacted with local baths. We therefore contend that even when looking at multipartite systems, unless they are of a very special type, if T_2 type processes dominate then we can well approximate the decoherence as pure dephasing and our results apply.

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