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Symmetrized Liouvillian Gap in Markovian Open Quantum Systems

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Markovian open quantum systems display complicated relaxation dynamics. The spectral gap of the Liouvillian characterizes the asymptotic decay rate towards the steady state, but it does not necessarily give a correct estimate of the relaxation time because the crossover time to the asymptotic regime may be too long. We here give a rigorous upper bound on the transient decay of auto-correlation functions in the steady state by introducing the symmetrized Liouvillian gap. The standard Liouvillian gap and the symmetrized one are identical in an equilibrium situation but differ from each other in the absence of the detailed balance condition. It is numerically shown that the symmetrized Liouvillian gap always gives a correct upper bound on the decay of the auto-correlation function, but the standard Liouvillian gap does not.

Introduction.— It is a fundamental problem in nonequilibrium physics to elucidate how fast a quantum system approaches stationarity under dissipative couplings to an external environment [1–8]. This problem is also of great practical interest in quantum technologies. Because quantum control and computations unavoidably suffer from dissipation and decoherence, it is becoming important to understand general properties of dissipative quantum dynamics [9–13]. Moreover, the strategy of utilizing engineered dissipation in controlling and manipulating quantum states, which was theoretically proposed as reservoir engineering [14, 15], is being implemented in experiments [16–18]. It was demonstrated that quantum phase transitions can be induced by controlling the strength of dissipation [18]. Those ongoing experimental developments will require a more precise theoretical understanding of open-system dynamics.

In the Markovian regime, where the environmental correlation time is much shorter than a typical time of dissipative processes, the dynamics of an open quantum system is generated by the Liouvillian superoperator of the celebrated Lindblad form [19, 20]. One might then expect that the knowledge of the eigenvalue spectrum of the Liouvillian is enough to estimate how fast the relaxation proceeds. Especially, the Liouvillian gap, which is defined as the smallest nonzero real part of the Liouvillian eigenvalue, has been investigated for various models [1, 3, 21–27]. Because the Liouvillian gap gives the decay rate of the slowest relaxation mode, it is naturally expected that its inverse bounds from above the relaxation time. However, it turns out that the problem is more elaborate. It has been shown that the relaxation time is not bounded by the Liouvillian gap [6, 7, 28, 29], although the latter characterizes the asymptotic decay rate appearing in the long-time limit [30]. It implies that the decay rate *in a transient regime* can be smaller than the Liouvillian gap. Such an anomalously small decay rate would be prominent in many-body systems [6], but not restricted to them.

In this Letter, we provide a rigorous analysis on the decay of auto-correlation functions in the steady state. Our result tells us that not the standard Liouvillian gap but the symmetrized one appears as a key quantity to bound the relaxation time in the transient regime. It turns out that the standard Liouvillian gap correctly bounds the relaxation time in an equilibrium situation, but not in a nonequilibrium situation without the detailed balance condition. In the latter, the symmetrized Liouvillian gap rigorously bounds the relaxation time.

Our results complement a series of results based on the quantum speed limit (QSL), which was originally formulated in isolated quantum systems [31] and later extended to open quantum systems [32–35]. The QSL gives a *lower* bound on the relaxation time, whereas an *upper* bound is investigated here.

In the following, we first explain the general setup, and then present main results. We demonstrate the validity of our theoretical results by numerical calculations in an interacting quantum dot coupled to reservoirs and driven-dissipative Rydberg atoms. An extension to the relaxation from a fixed initial state is briefly mentioned.

Setup.— Let us consider a Markovian open quantum system, whose state at time t is represented by the density matrix $\rho(t)$. Its time evolution is generated by the Liouvillian superoperator $\mathcal{L}$ of the Lindblad form [36]: $d\rho(t)/dt = \mathcal{L}\rho(t)$, where

$$\mathcal{L}\rho = -i[\hat{H},\rho] + \sum_k \left(\hat{L}_k \rho \hat{L}_k^\dagger - \frac{1}{2}\left\{ \hat{L}_k^\dagger \hat{L}_k, \rho \right\} \right). \quad (1)$$

The first term of the right-hand side expresses the intrinsic unitary evolution of the system with Hamiltonian $\hat{H}$, whereas the second term represents the dissipation characterized by a set of Lindblad jump operators $\{\hat{L}_k\}$. The Lindblad form ensures physically natural properties such as the complete positivity [19, 20].

Let us denote by $\{\lambda_\alpha\}$ the eigenvalues of $\mathcal{L}$. It is shown that any eigenvalue has a non-positive real part, and hence we sort the eigenvalues in the descending order:

$$0 = \lambda_0 > \mathrm{Re}\,\lambda_1 \geq \mathrm{Re}\,\lambda_2 \geq \dots. \quad (2)$$

In this work, we assume that the zero eigenvalue is not degenerate: the steady state is unique. Because of the

property $\mathcal{L}(\rho^\dagger) = \mathcal{L}(\rho)^\dagger$, it is shown that λ_α^* is also an eigenvalue. The Liouvillian gap g is defined as

$$g = -\operatorname{Re}\lambda_1, \tag{3}$$

which determines the asymptotic decay rate [30].

Let us introduce two inner products $\langle\hat{A},\hat{B}\rangle$ and $\langle\hat{A},\hat{B}\rangle_{\rm ss}$ for two operators $\hat{A}$ and $\hat{B}$. The first inner product is defined as $\langle\hat{A},\hat{B}\rangle = \mathrm{Tr}(\hat{A}^\dagger\hat{B})$. Accordingly, we define an adjoint superoperator $\tilde{\mathcal{L}}$ of $\mathcal{L}$ as follows:

$$\langle\hat{A},\mathcal{L}\hat{B}\rangle = \langle\tilde{\mathcal{L}}\hat{A},\hat{B}\rangle\,. \tag{4}$$

The expectation value of an Hermitian operator $\hat{A}$ at time t is expressed as

$$\langle\hat{A}(t)\rangle = \mathrm{Tr}[\hat{A}\rho(t)] = \langle\hat{A},e^{\mathcal{L}t}\rho\rangle = \langle e^{\tilde{\mathcal{L}}t}\hat{A},\rho\rangle\,. \tag{5}$$

Here, $\hat{A}(t) := e^{\tilde{\mathcal{L}}t}\hat{A}$ is interpreted as the time-evolved operator in the Heisenberg picture. It is explicitly given by

$$\tilde{\mathcal{L}}\hat{A} = i[\hat{H},\hat{A}] + \sum_k\left(\hat{L}_k^\dagger\hat{A}\hat{L}_k - \frac{1}{2}\left\{\hat{L}_k^\dagger\hat{L}_k,\hat{A}\right\}\right). \tag{6}$$

Since $\tilde{\mathcal{L}}$ is an adjoint of $\mathcal{L}$, $\tilde{\mathcal{L}}$ has the same eigenvalue spectrum as $\mathcal{L}$. We denote by χ_α left eigenvectors of $\mathcal{L}$: $\chi_\alpha^\dagger\mathcal{L} = \lambda_\alpha\chi_\alpha^\dagger$. We then find that χ_α are right eigenvectors of $\tilde{\mathcal{L}}$: $\tilde{\mathcal{L}}\chi_\alpha = \lambda_\alpha^*\chi_\alpha$.

The second inner product is given by

$$\langle\hat{A},\hat{B}\rangle_{\rm ss} := \mathrm{Tr}[\hat{A}^\dagger\hat{B}\rho_{\rm ss}], \tag{7}$$

which we call the steady-state inner product [37]. The corresponding adjoint superoperator $\tilde{\mathcal{L}}^*$ of $\tilde{\mathcal{L}}$ associated with $\langle\cdot,\cdot\rangle_{\rm ss}$ is defined as

$$\langle\hat{A},\tilde{\mathcal{L}}\hat{B}\rangle_{\rm ss} = \langle\tilde{\mathcal{L}}^*\hat{A},\hat{B}\rangle_{\rm ss}\,. \tag{8}$$

It should be noted that $\tilde{\mathcal{L}}^*$ depends on the steady state $\rho_{\rm ss}$. It is shown that $\tilde{\mathcal{L}}^*$ is expressed as $\tilde{\mathcal{L}}^*A = \mathcal{L}(\hat{A}\rho_{\rm ss})\rho_{\rm ss}^{-1}$ [37], where we assume that $\rho_{\rm ss}$ is invertible. Again, $\tilde{\mathcal{L}}^*$ has the same eigenvalue spectrum as $\mathcal{L}$. For later convenience, we define the steady-state norm $\|\cdot\|_{\rm ss}$ as

$$\|\hat{A}\|_{\rm ss} := \sqrt{\langle\hat{A},\hat{A}\rangle_{\rm ss}} \geq 0. \tag{9}$$

Main results.— Let us consider an auto-correlation function $C_A(t) = \mathrm{Tr}[\hat{A}(t)\hat{A}\rho_{\rm ss}] = \langle\hat{A}(t),\hat{A}\rangle_{\rm ss}$ in the steady state, where $\hat{A}$ is an Hermitian operator satisfying $\langle\hat{A}\rangle_{\rm ss} := \mathrm{Tr}[\hat{A}\rho_{\rm ss}] = 0$ (this assumption is made without loss of generality because we can always shift $\hat{A}\to\hat{A}-\langle\hat{A}\rangle_{\rm ss}\hat{I}$ with the identity operator $\hat{I}$).

We investigate how quickly $C_A(t)$ decays. It is known that the Liouvillian gap determines the asymptotic decay of $C_A(t)$:

$$|C_A(t)| \lesssim e^{-gt} \quad (t\to\infty). \tag{10}$$

However, in a transient regime, the Liouvillian gap does not necessarily give the smallest decay rate [6, 7, 28, 29, 38], i.e. the inequality $|C_A(t)| \leq e^{-gt}C_A(0)$ does not hold in general. It is thus desired to give a rigorous bound on $|C_A(t)|$ at *finite* times.

In this Letter, we give such a bound:

$$|C_A(t)| \leq e^{-g_st}C_A(0) \text{ for any } t\geq 0, \tag{11}$$

where g_s is the spectral gap (i.e. the difference between the largest and the second-largest eigenvalues) of *symmetrized Liouvillian*

$$\tilde{\mathcal{L}}_s := \frac{\tilde{\mathcal{L}}+\tilde{\mathcal{L}}^*}{2}, \tag{12}$$

which is Hermitian with respect to the second inner product $\langle\cdot,\cdot\rangle_{\rm ss}$. We call g_s the *symmetrized Liouvillian gap*. Because g_s does not depend on $\hat{A}$, Eq. (11) tells us that the inverse of the symmetrized Liouvillian gap gives a general upper bound on the decay time of any auto-correlation function.

Later, we numerically show that our bound (11) is tight in two different models. It means that the symmetrized Liouvillian gap is not a mathematical artifact but a relevant quantity in the relaxation of open quantum systems.

Although we have focused on static Hamiltonians, our formalism can be naturally extended to open Floquet systems, i.e. periodically driven systems in contact with reservoirs, which have been studied from long ago, but attracted renewed interests in the context of Floquet engineering in open systems [39]. See Supplementary Material (SM) for the detail [40].

We point out a recent work [41] in which the symmetrized Liouvillian gap is used to derive concentration bounds for *finite*-time averages of measurement outcomes in quantum Markov processes. Such a general result is applied to derive upper bounds on the size of fluctuations of trajectory observables like time-integrated currents [42], which complement lower bounds provided by thermodynamic uncertainty relations [43]. In this way, the symmetrized Liouvillian gap is a key quantity to study *finite*-time properties of Markov processes.

Properties of $\tilde{\mathcal{L}}_s$ and g_s.— Before proving Eq. (11), we summarize basic properties of $\tilde{\mathcal{L}}_s$ and g_s below:

(i) $\tilde{\mathcal{L}}_s$ has a zero eigenvalue, and $\hat{I}$ is the corresponding eigenvector.

(ii) $\tilde{\mathcal{L}}_s$ is negative semidefinite, i.e., all the eigenvalues are non-positive.

(iii) $0\leq g_s\leq g$.

(iv) $g_s = g$ when $[\tilde{\mathcal{L}},\tilde{\mathcal{L}}^*] = 0$.

The property (i) is easily confirmed by using $\tilde{\mathcal{L}}\hat{I} = \tilde{\mathcal{L}}^*\hat{I} = 0$ [recall $\tilde{\mathcal{L}}^*A = \mathcal{L}(\hat{A}\rho_{\rm ss})\rho_{\rm ss}^{-1}$ and $\mathcal{L}\rho_{\rm ss} = 0$].

The property (ii) is proved by using the following inequality for the Liouvillain of the Lindblad form [19]:

$$\tilde{\mathcal{L}}(\hat{X}^\dagger)\hat{X} + \hat{X}^\dagger\tilde{\mathcal{L}}(\hat{X}) \le \tilde{\mathcal{L}}(\hat{X}^\dagger\hat{X}) \tag{13}$$

for any bounded operator $\hat{X}$. From this inequality, we have

$$\begin{aligned}\langle \hat{X}, \tilde{\mathcal{L}}_s\hat{X}\rangle_{\rm ss} &= \frac{1}{2}\,{\rm Tr}\left\{\left[\tilde{\mathcal{L}}(\hat{X}^\dagger)\hat{X} + \hat{X}^\dagger\tilde{\mathcal{L}}(\hat{X})\right]\rho_{\rm ss}\right\} \\ &\le \frac{1}{2}\,{\rm Tr}\left[\tilde{\mathcal{L}}(\hat{X}^\dagger\hat{X})\rho_{\rm ss}\right] = \frac{1}{2}\,{\rm Tr}[\hat{X}^\dagger\hat{X}\mathcal{L}\rho_{\rm ss}] = 0, \end{aligned}\tag{14}$$

which proves (ii).

Next, we prove (iii). From the definition, $g_s \ge 0$ is obvious. Because of (i) and (ii), $-g_s$ is nothing but the second-largest eigenvalue of $\tilde{\mathcal{L}}_s$, which has the following variational expression:

$$\begin{aligned} -g_s &= \sup_{\hat{X}\neq 0:\langle\hat{X}\rangle_{\rm ss}=0} \frac{\langle \hat{X}, \tilde{\mathcal{L}}_s\hat{X}\rangle_{\rm ss}}{\langle \hat{X}, \hat{X}\rangle_{\rm ss}} \\ &= \sup_{\hat{X}\neq 0:\langle\hat{X}\rangle_{\rm ss}=0} \frac{{\rm Re}\,\langle \hat{X}, \tilde{\mathcal{L}}\hat{X}\rangle_{\rm ss}}{\langle \hat{X}, \hat{X}\rangle_{\rm ss}}. \end{aligned}\tag{15}$$

The condition $\langle\hat{X}\rangle_{\rm ss} = \langle\hat{I}, \hat{X}\rangle_{\rm ss} = 0$ in Eq. (15) guarantees that $\hat{X}$ is orthogonal to the identity $\hat{I}$, which is the eigenvector of $\tilde{\mathcal{L}}_s$ with zero eigenvalue. Since χ_1 satisfies $\langle\chi_1\rangle_{\rm ss} = 0$ and $\tilde{\mathcal{L}}\chi_1 = \lambda_1^*\chi_1$, we obtain

$$g_s \le -\frac{{\rm Re}\,\langle\chi_1, \tilde{\mathcal{L}}\chi_1\rangle_{\rm ss}}{\langle\chi_1,\chi_1\rangle_{\rm ss}} = -\,{\rm Re}\,\lambda_1^* = g, \tag{16}$$

which proves (iii).

The following observation is key to prove the last property (iv): When $[\tilde{\mathcal{L}}, \tilde{\mathcal{L}}^*] = 0$, χ_α is a simultaneous eigenvector of $\tilde{\mathcal{L}}$ and $\tilde{\mathcal{L}}^*$ with the eigenvalue λ_α^* and λ_α, respectively, which follows from the fact that any normal matrix is unitarily diagonalizable [44]. It implies that χ_α is an eigenvector of $\tilde{\mathcal{L}}_s$ with the eigenvalue ${\rm Re}\,\lambda_\alpha$. Thus, we conclude $g_s = \min_{\alpha\neq 0}[-\,{\rm Re}\,\lambda_\alpha] = g$.

The property (iv) is of physical importance. The condition $[\tilde{\mathcal{L}}, \tilde{\mathcal{L}}^*] = 0$ holds whenever the Liouvillian obeys the quantum detailed balance condition [37]. When the system is coupled to an equilibrium reservoir and its dynamics is described by the Lindblad equation with the quantum detailed balance, the standard Liouvillian gap g gives a bound on the decay of any auto-correlation function as $|C_A(t)| \le e^{-gt}C_A(0)$. While, when the system is put in a nonequilibrium situation (e.g. the system is in contact with multiple reservoirs at different temperatures), we need g_s to obtain a correct upper bound. In this sense, g_s is relevant in nonequilibrium open quantum systems.

Proof of Eq. (11).— We first express the auto-correlation function as $C_A(t) = \langle\hat{A}(t), \hat{A}\rangle_{\rm ss}$. By using the Cauchy-Schwarz inequality, we obtain

$$|C_A(t)| \le \|\hat{A}(t)\|_{\rm ss}\cdot\|\hat{A}\|_{\rm ss}. \tag{17}$$

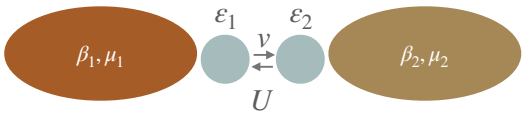

FIG. 1. A schematic of the model. Two interacting quantum dots are interacting with their own reservoirs.

Let us evaluate $f(t) := \|\hat{A}(t)\|_{\rm ss}^2 = \langle\hat{A}(t), \hat{A}(t)\rangle_{\rm ss}$. By differentiating it with respect to t, we have

$$\begin{aligned}\frac{df}{dt} &= \langle\tilde{\mathcal{L}}\hat{A}(t), \hat{A}(t)\rangle_{\rm ss} + \langle\hat{A}(t), \tilde{\mathcal{L}}\hat{A}(t)\rangle_{\rm ss} \\ &= \langle\hat{A}(t), (\tilde{\mathcal{L}} + \tilde{\mathcal{L}}^*)\hat{A}(t)\rangle_{\rm ss} \\ &= 2\frac{\langle\hat{A}(t), \tilde{\mathcal{L}}_s\hat{A}(t)\rangle_{\rm ss}}{\langle\hat{A}(t), \hat{A}(t)\rangle_{\rm ss}} f(t) \le -2g_s f(t), \end{aligned}\tag{18}$$

where Eq. (15) was used in the last inequality. By integrating it over t, we obtain

$$f(t) \le e^{-2g_s t}f(0) = e^{-2g_s t}\|\hat{A}\|_{\rm ss}^2. \tag{19}$$

By substituting it into Eq. (17) and using $\|\hat{A}\|_{\rm ss}^2 = C_A(0)$, we obtain Eq. (11).

Double quantum dot in contact with two reservoirs.— We demonstrate the relevance of our main results in a specific model. The Hamiltonian of the total system is given by $\hat{H}_T = \hat{H}_S + \hat{H}_B + \hat{H}_I$. The Hamiltonian $\hat{H}_S$ of a double quantum dot is given by

$$\hat{H}_S = \sum_{i=1}^{2}\varepsilon_i\hat{d}_i^\dagger\hat{d}_i + v(\hat{d}_1^\dagger\hat{d}_2 + \hat{d}_2^\dagger\hat{d}_1) + U\hat{d}_1^\dagger\hat{d}_1\hat{d}_2^\dagger\hat{d}_2, \tag{20}$$

where $\hat{d}_i$ is the annihilation operator of ith dot. We denote by E_n and $|n\rangle$ the energy eigenvalue and the corresponding energy eigenstate: $\hat{H}_S = \sum_n E_n|n\rangle\langle n|$. The Hamiltonian of the two reservoirs is given by

$$\hat{H}_B = \sum_k(\hat{c}_{k,1}^\dagger\hat{c}_{k,1} + \hat{c}_{k,2}^\dagger\hat{c}_{k,2}), \tag{21}$$

where $\hat{c}_{k,i}$ is the annihilation operator of fermions in the reservoir coupled to ith dot. The interaction Hamiltonian reads

$$\hat{H}_I = \sum_{i=1}^{2}\left(\hat{d}_i^\dagger\sum_k\lambda_k\hat{c}_{k,i} + {\rm h.c.}\right). \tag{22}$$

We assume that two reservoirs are in thermal equilibrium at the inverse temperature β_i and the chemical potential μ_i $(i = 1, 2)$. See Fig. 1 for a schematic of the model.

When the interaction between the system and the reservoirs is sufficiently weak, the Lindblad equation for the reduced density matrix $\rho(t)$ for the system of interest is derived by applying the Born-Markov and secular approximations [36]. The Liouvillian is block diagonalized

into sectors each of which is spanned by $|n\rangle\langle m|$ with a fixed frequency $\omega = E_n - E_m$. The sector of $\omega = 0$ corresponds to the subspace spanned by the diagonal matrix elements $\{|n\rangle\langle n|\}$ if we assume no energy degeneracy in $\hat{H}_S$. Let us consider $C_A(t)$ with $\hat{A}$ being a diagonal matrix in the energy basis. The dynamics of $\hat{A}(t)$ is then restricted to the diagonal subspace. For this reason, we focus on the diagonal sector and define the spectral gap within this sector.

The Born-Markov-secular Lindblad equation in the diagonal sector is given by the following Pauli master equation [36]:

$$\frac{dP_n}{dt} = \sum_m \left[W_{nm}P_m(t) - W_{mn}P_n(t)\right], \tag{23}$$

where $P_n(t) = \langle n|\rho(t)|n\rangle$ and the transition rate matrix W_{nm} is given by

$$W_{nm} = 2\pi J(E_n - E_m)\sum_{i=1}^{2} f_i(E_n - E_m)|\langle n|\hat{d}_i^\dagger|m\rangle|^2$$
$$+2\pi J(E_m - E_n)\sum_{i=1}^{2}[1 - f_i(E_m - E_n)]|\langle m|\hat{d}_i|n\rangle|^2. \tag{24}$$

Here, $J(\omega) = \sum_k \delta(\omega - \omega_k)|\lambda_k|^2$ is the bath spectral function and $f_i(E) = [e^{\beta_i(E-\mu_i)} + 1]^{-1}$ is the Fermi distribution at ith reservoir.

We numerically compute the auto-correlation function of the energy,

$$C_E(t) = \langle \delta\hat{H}_S(t), \delta\hat{H}_S(0)\rangle_{\rm ss}, \tag{25}$$

where $\delta\hat{H}_S = \hat{H}_S - \langle\hat{H}_S\rangle_{\rm ss}\hat{I}$. In Fig. 2, we plot $C_E(t)/C_E(0)$, our upper bound $e^{-g_s t}$, and e^{-gt} for (a) an equilibrium case ($\beta_1 = \beta_2$ and $\mu_1 = \mu_2$) and (b) a nonequilibrium case. In numerical calculations, we set $v = 1$, $\varepsilon_1 = -1.37$, $\varepsilon_2 = -2.24$, $U = 1.76$, and assume $2\pi J(\omega) = \gamma \equiv 1$. In Fig. 2 (a), we set $\beta_1 = \beta_2 = 5.5$ and $\mu_1 = \mu_2 = 0.3$, whereas in Fig. 2 (b), $\beta_1 = 6.94$, $\beta_2 = 4.06$, $\mu_1 = -1.63$, and $\mu_2 = 2.23$.

In an equilibrium case, $g = g_s$ and therefore the Liouvillian gap gives an upper bound on the relaxation time τ as $\tau \lesssim g^{-1}$. While, in a nonequilibrium case, the decay rate in a transient regime is not bounded by the Liouvillian gap. Instead, the symmetrized Liouvillian gap gives a correct bound even in this case. Moreover, it gives a tight upper bound at short times [see Fig. 2 (b)]. Thus, in general, the symmetrized Liouvillian gap is needed to evaluate the maximum relaxation time of the dissipative system.

Driven-dissipative Rydberg atoms.— In Eq. (23), dynamics of the population (i.e. diagonal elements in the energy basis) is decoupled from that of the coherence (i.e. off-diagonal elements), and thus the population dynamics is essentially classical. Now we consider dissipative dynamics that is genuinely quantum (i.e. the population and the coherence are not independent).

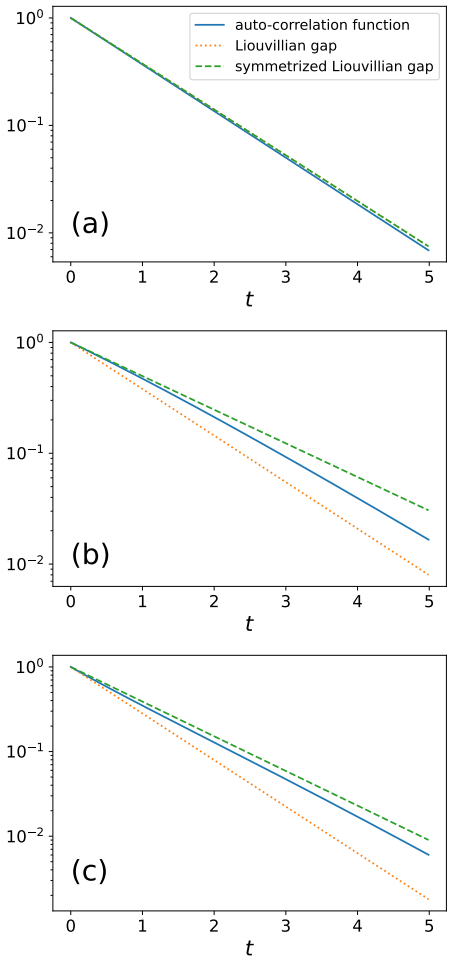

FIG. 2. Auto-correlation function $C_E(t)$ for (a) equilibrium and (b) nonequiliblium quantum dots, and for (c) driven-dissipative Rydberg atoms. The solid lines are the numerical values of $C_E(t)/C_E(0)$. The orange dotted lines and the green dashed lines are e^{-gt} and $e^{-g_s t}$, respectively.

Let us consider two Rydberg atoms under laser driving and dissipation. Each Rydberg atom is considered as a two-level system, where the spin up (down) corresponds to the Rydberg (ground) state. Laser-driven dissipative Rydberg atoms are described by the Lindblad equation with the Ising Hamiltonian [45]

$$\hat{H} = J\hat{\sigma}_1^z\hat{\sigma}_2^z + \sum_{i=1}^{2}(h^z\hat{\sigma}_i^z + h^x\hat{\sigma}_i^x), \tag{26}$$

where $\sigma_i^{x,y,z}$ denote the Pauli matrices ($i = 1, 2$), and jump operators $\hat{L}_{i,+} = \sqrt{\gamma_+}\hat{\sigma}_i^+$ and $\hat{L}_{i,-} = \sqrt{\gamma_-}\hat{\sigma}_i^-$, where $\gamma_\pm > 0$ are positive constants (see SM of Ref. [46] for the derivation of jump operators).

We numerically compute $C_E(t)$ and the result is plotted in Fig. 2 (c). Parameters are set as $J = 0.054$, $h^z = 0.107$, $h^x = 0.267$, $\gamma_- = 1$, and $\gamma_+ = 0.9$. We again see that not g but g_s bounds the decay rate in a transient regime.

Relaxation from an arbitrary initial state.— So far, we have considered the relaxation of fluctuations at stationarity. Now we briefly discuss an extension of the analysis to the relaxation dynamics from an arbitrary initial state $\rho_{\rm ini}$ towards the steady state $\rho_{\rm ss}$. We can prove the following inequality [40]:

$$\|\rho(t) - \rho_{\rm ss}\|_1 \leq \left(e^{S_2(\rho_{\rm ini}\|\rho_{\rm ss})} - 1\right)^{1/2} e^{-g_s t}, \tag{27}$$

where $\|\cdot\|_1$ denotes the trace norm, and $S_2(\rho\|\sigma) = \ln \mathrm{Tr}[\rho^2\sigma^{-1}]$ is the quantum Rényi 2-divergence. From this inequality, we find that the relaxation time $\tau_{\rm rel}$ is bounded as

$$\tau_{\rm rel} \lesssim \frac{S_2(\rho_{\rm ini}\|\rho_{\rm ss})}{g_s}. \tag{28}$$

Comparing with the inequality (11) for the auto-correlation function, we have an additional factor that grows with $S_2(\rho_{\rm ini}\|\rho_{\rm ss})$. In SM [40], it is shown that this additional factor explains anomalously long relaxation times in the single-particle asymmetric hopping model, which was reported by Haga *et al.* [7]. In contrast, it turns out that $g_s = 0$ in the boundary-dissipated system studied in Ref. [6], and hence Eqs. (11) and (27) are not informative. In general, studies beyond the spectral-gap analysis are needed to fully understand the relaxation time.

Summary and Outlook.— We have derived an upper bound on the decay of auto-correlation functions in the steady state, and numerically show that the bound is tight in a transient regime. The decay of correlations is bounded by the symmetrized Liouvillian gap, which may differ from the standard Liouvillian gap when the quantum detailed balance condition is violated. We note that it is straightforward to extend our results to classical Markov jump processes.

We believe that our results unveil a general property of nonequilibrium quantum dissipative dynamics. While, we are also convinced that the symmetrized Liouvillian gap does not capture whole physics of Markovian quantum dynamics at finite times. It is a future problem to unveil its generic properties beyond the spectral-gap analysis of the (symmetrized) Liouvillian.

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