Controlling Matter Phases beyond Markov

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Controlling phase transitions in quantum systems via coupling to reservoirs has been mostly studied for idealized memory-less environments under the so-called Markov approximation. Yet, most quantum materials and experiments in the solid state, atomic, molecular and optical physics are coupled to reservoirs with finite memory times. Here, using the spectral theory of non-Markovian dissipative phase transitions developed in the companion paper [Debecker, Martin, and Damanet (to be published)], we show that memory effects can be leveraged to reshape matter phase boundaries, but also reveal the existence of dissipative phase transitions genuinely triggered by non-Markovian effects.

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Introduction—Finding new ways to control phase transitions in quantum systems to access desired properties is at the forefront of research for developing new materials and technologies. In this context, driven-dissipative mechanisms obtained via the coupling of systems to engineered environments and fields offer opportunities to generate matter phases otherwise inaccessible [1–3].

However, thus far, dissipative phase transitions (DPTs), which have been observed in controlled experiments [4–12], have mostly garnered theoretically attention in systems coupled to memoryless reservoirs [13–15]. However, most realistic systems are coupled to reservoirs with a spectral structure [16], giving the latter a memory of past system-bath exchanges, which considerably complicates their dynamics. Such non-Markovian effects are crucial to be understood, not least because they can be used as a resource to generate useful phenomena, such as non-Markovian-assisted steady state entanglement [17], quantum transport [18], spin squeezing [19], chaotic behaviors [20], or new dynamical phases [21]. Moreover, from a computational perspective, it is sometimes desirable to derive reduced descriptions of a large Markovian open quantum system in order to deal with a smaller Hilbert space, which usually implies dealing with non-Markovian effects [22,23].

Here, using the theoretical framework developed in the companion paper [24], which generalizes the spectral theory of dissipative phase transitions to non-Markovian systems, we highlight how non-Markovian effects can be used to reshape phase boundaries but also capture DPTs totally overlooked by the Markov approximation; this opens many possibilities to explore DPTs in a wider range of systems and to access matter phases in new regimes of parameters and experimental setups. By contrast with previous studies of non-Markovian effects in DPTs via other techniques [25–28], which mostly focus on the paradigmatic spin-boson model [29–31], here we provide a general method, which we apply in two specific examples.

Below, we first summarize the theoretical framework presented in [24] and in particular its central element: the generalization of the Liouvillian for non-Markovian systems, its properties and its connections with DPTs and symmetries. Then, we apply the method to a generalized Lipkin-Meshkov-Glick model [32] and show that deviations from a Markovian reservoir lead to a shift of the phase transition boundary. Then, even more remarkably, we reveal the existence of DPTs triggered by non-Markovianity.

Theoretical framework—Consider a system S coupled to a bosonic environment E at zero temperature [33]. The total Hamiltonian ($\hbar = 1$) is

$$H = H_S + \underbrace{\sum_k \omega_k a_k^\dagger a_k}_{\equiv H_E} + \underbrace{\sum_k (g_k a_k L^\dagger + g_k^* a_k^\dagger L)}_{\equiv H_{\rm int}}, \quad (1)$$

where H_S (H_E) is the system (environment) Hamiltonian, with a_k ($a_k^\dagger$) the annihilation (creation) operator for the kth mode of frequency ω_k, and $H_{\rm int}$ is the interaction Hamiltonian with L being an arbitrary system operator and g_k being the system-bath coupling strengths. The effect of the environment on the system is encoded in the spectral density $J(\omega) = \pi \sum_k |g_k|^2 \delta(\omega - \omega_k)$ or, equivalently, in the bath correlation function $\alpha(\tau) = \sum_k |g_k|^2 e^{-i\omega_k \tau} = (1/\pi) \int_0^\infty J(\omega) e^{-i\omega\tau} {\rm d}\omega$. We assume that the correlation function, which depends on the model, is a sum of M decaying exponentials

$$\alpha(\tau) = \sum_{j=1}^{M} G_j e^{-i\omega_j \tau - \kappa_j |\tau|}, \quad \kappa_j, \omega_j \in \mathbb{R}, \quad G_j \in \mathbb{C}. \quad (2)$$

This decomposition, which is not unique [34], can be performed either exactly or with great precision in a wide

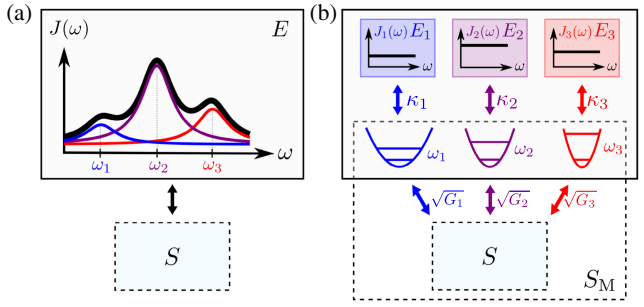

FIG. 1. (a): Sketch of a system S interacting with a structured environment E characterized by a spectral density $J(\omega)$ that can be decomposed into three Lorentzians, as if the system was coupled to three pseudomodes coupled to their own unstructured bath (b). If one enlarges the system S by including the pseudomodes (system S_M in the dashed black box), the dynamics can be treated by a Lindblad description.

range of situations [35–38]. For $G_j \in \mathbb{R}$, this amounts to decompose the non-Markovian environment E into a set of M modes of frequencies $\{\omega_j\}$ which are damped with rates $\{\kappa_j\}$ due to their coupling to independent Markovian baths, as illustrated in Fig. 1. This pseudomode picture [39–46] is relevant for atoms in cavities, superconducting qubits coupled to resonators [47,48], electron-phonon systems [49,50], or emitters in plasmonic cavities [51]. Using complex G_j is even more general and allows, for instance, an efficient fit of Ohmic spectral density [34,52] and the study of critical behaviors [53].

When the global system is initially in the state $\rho(0) = \rho_S(0) \otimes \rho_B(0)$, the exact dynamics of S can be described by the HEOM method which takes the form [38,54–57]

$$\begin{aligned}\frac{d\rho^{(\vec{n},\vec{m})}}{dt} &= -i[H_S, \rho^{(\vec{n},\vec{m})}] - (\vec{w}^*\cdot\vec{n} + \vec{w}\cdot\vec{m})\rho^{(\vec{n},\vec{m})} \\ &+ \sum_{j=1}^{M} \Big\{ G_j n_j L \rho^{(\vec{n}-\vec{e}_j,\vec{m})} + G_j^* m_j \rho^{(\vec{n},\vec{m}-\vec{e}_j)} L^\dagger \\ &+ \Big[\rho^{(\vec{n}+\vec{e}_j,\vec{m})}, L^\dagger\Big] + \Big[L, \rho^{(\vec{n},\vec{m}+\vec{e}_j)}\Big] \Big\}, \end{aligned} \tag{3}$$

with $\vec{n} = (n_j)$ and $\vec{m} = (m_j)$ multi-indices in $\mathbb{N}^M$, $\vec{w} = (\kappa_j + i\omega_j) \in \mathbb{C}^M$, $\vec{e}_j = (\delta_{jj'})$ unit vectors, and $\vec{a}\cdot\vec{b} = \sum_j a_j^* b_j$ the inner product on $\mathbb{C}^M$. In Eq. (3), $\rho^{(\vec{0},\vec{0})} \equiv \rho_S$ corresponds to the physical density operator of the system S with which all the mean values of system observables are computed, while $\rho^{(\vec{n},\vec{m})}$ for $(\vec{n},\vec{m}) \neq (\vec{0},\vec{0})$, which are also operators acting on the system space, correspond to auxiliary states from which bath correlations can be obtained [19]. Although the hierarchy is formally infinite, it can be truncated at large hierarchy depth indices $\vec{n}$ and $\vec{m}$. In practice, the stronger the non-Markovianity, the larger the number of auxiliary states we need to retain to obtain convergence of the results. Here, we choose the triangular truncation $\rho^{(\vec{n},\vec{m})} = 0 \ \forall \ \vec{n}, \ \vec{m}: \ \sum_j (n_j + m_j) > k_{\max}$, where $k_{\max}$ is the truncation order, yielding a total of $K = (2M + k_{\max})!/((2M)!k_{\max}!)$ auxiliary states [19].

By stacking in a vector $|\rho\rangle\!\rangle$ all the vectorized versions of the matrices $\rho^{(\vec{n},\vec{m})}$, Eq. (3) can be written in the form

$$\frac{d|\rho\rangle\!\rangle}{dt} = \mathcal{L}_{\text{HEOM}}(k_{\max})|\rho\rangle\!\rangle, \tag{4}$$

where $\mathcal{L}_{\text{HEOM}}(k_{\max})$ is the *HEOM Liouvillian*, the generator of the non-Markovian dynamics of the system, exact for $k_{\max} \to +\infty$ and which generalizes Lindblad's Liouvillian. Instead of using $\mathcal{L}_{\text{HEOM}}$, one can sometimes as noted above enlarge the system by including explicit pseudomode degrees of freedom damped by standard Lindblad decay channels, as illustrated in Fig. 1(b). This would define a Markovian Liouvillian $\mathcal{L}_M$ for the global system S_M. However, as shown in [24] using $\mathcal{L}_{\text{HEOM}}$ is computationally more favorable than $\mathcal{L}_M$, especially for large M.

Properties of the HEOM Liouvillian—The superoperator $\mathcal{L}_{\text{HEOM}}$ is linear and in general non-Hermitian. We assume it is diagonalizable and denote its eigenvectors and eigenvalues by $|\rho_i\rangle\!\rangle$ and λ_i. For a truncation order $k_{\max}$, its dimension is $D = K\dim(\mathcal{H}_S)^2$. It admits the following properties: (i) its spectrum is symmetric with respect to the real axis; (ii) it preserves the trace of the physical state $\rho^{(\vec{0},\vec{0})}$; (iii) the eigenvalue 0 is always in its spectrum, guaranteeing the existence of a stationary state; (iv) all the eigenvalues must have a negative real part in the limit $k_{\max} \to +\infty$; (v) $\text{Tr}[\mathbb{1}^{(\vec{0},\vec{0})}\rho_i] = 0$ with $\mathbb{1}^{(\vec{0},\vec{0})}$ the projector onto the physical state space if ρ_i is a right eigenoperator of $\mathcal{L}_{\text{HEOM}}$ associated with the eigenvalue λ_i with $\text{Re}[\lambda_i] \neq 0$. As in [13,14], we order the eigenvalues of $\mathcal{L}_{\text{HEOM}}$ so that $|\text{Re}[\lambda_0]| < |\text{Re}[\lambda_1]| < \ldots < |\text{Re}[\lambda_D]|$, where $\lambda_0 = 0$.

DPT and HEOM Liouvillian spectrum—Consider a system described by Eq. (4) which admits a valid thermodynamic limit $N \to \infty$ and a unique steady state ρ_{ss} for all finite N. We say that the system undergoes a phase transition of order p when a non-analytical change in a g-independent system observable O occurs when the parameter g tends to a critical value g_c for $N \to \infty$, i.e., [13]

$$\lim_{g\to g_c} \left| \frac{\partial^p}{\partial g^p} \lim_{N\to+\infty} \langle O \rangle_{ss} \right| = +\infty, \tag{5}$$

where $\langle O \rangle_{ss} = \text{Tr}[O\rho_{ss}^{(\vec{0},\vec{0})}]$. This definition of DPTs is the same as for Markovian systems. The only difference is that the steady state is now obtained from the HEOM Liouvillian (4). Like for the Markovian case, a nonanalytical change as described by (5) must occur due to a level crossing in the spectrum of $\mathcal{L}_{\text{HEOM}}$, which implies the closing of the HEOM Liouvillian gap $\text{Re}[\lambda_1]$. For 1st-order DPTs, the connection is even stronger as a DPT occurs iff

$\mathrm{Re}[\lambda_1] = 0$ at $g = g_c$ and $\mathrm{Im}[\lambda_1] = 0$ in a finite domain around g_c for $N \to \infty$ [24].

Symmetries and DPTs—We call *weak symmetry* of $\mathcal{L}_{\mathrm{HEOM}}$ any unitary superoperator $\mathcal{U}$ such that $[\mathcal{L}_{\mathrm{HEOM}}, \mathcal{U}] = 0$. The matrix representing $\mathcal{L}_{\mathrm{HEOM}}$ in the eigenvector basis of $\mathcal{U}$ is block diagonal, i.e., $\mathcal{L}_{\mathrm{HEOM}} = \bigoplus_{u_k} \mathcal{L}_{u_k}$, where each block $\mathcal{L}_{u_k}$ is associated with distinct eigenvalues u_k of $\mathcal{U}$ where $k \in \{0, 1, \ldots\}$. We define the *symmetry sector* L_{u_k} as the subspace spanned by the eigenvectors of $\mathcal{U}$ associated with the eigenvalue u_k. We can prove, in close analogy with the Markovian case [13] that if the steady state $|\rho_{ss}\rangle\!\rangle$ of (4) is unique, then $|\rho_{ss}\rangle\!\rangle \in L_{u_0=1}$ [24]. A spontaneous symmetry breaking (SSB) corresponds to the emergence of a zero eigenvalue in each symmetry sector k in the limit $N \to \infty$. To be specific, if $\mathcal{L}_{\mathrm{HEOM}}$ is a direct sum of $n+1$ blocks and if its eigenvalues are sorted in each block k as $|\mathrm{Re}[\lambda_0^{(k)}]| < |\mathrm{Re}[\lambda_1^{(k)}]| < \ldots$, a SSB is signaled by $\lambda_0^{(k)} \to \lambda_0^{(0)} = 0 \ \forall \ k > 0$ for $g \geq g_c$, $N \to +\infty$ [58]. This means that the independent hierarchies associated with each block k mix in the limit $N \to +\infty$ so that steady state that explicitly break the symmetry emerge.

1st-order DPT—We first illustrate our approach for a Lipkin-Meshkov-Glick (LMG) model of the form

$$H_{\mathrm{LMG}} = \frac{V}{N}(S_x^2 - S_y^2) = \frac{V}{2N}(S_+^2 + S_-^2), \tag{6}$$

where $S_\alpha = \sum_{j=1}^N \sigma_\alpha^{(j)}/2$ ($\alpha = x, y, z$) are the collective spin operators defined in terms of single-spin Pauli operators $\sigma_\alpha^{(j)}$ and $S_\pm = S_x \pm iS_y$. When the spin system undergoes collective decay as described by Lindblad's master equation

$$\dot{\rho} = -i[H_{\mathrm{LMG}}, \rho] + \frac{\gamma}{2N}\mathcal{D}_{S_-}[\rho], \tag{7}$$

where $\mathcal{D}_o[\cdot] = 2o\cdot o^\dagger - \{o^\dagger o, \cdot\}$, as would occur if coupled to an unstructured bath with $\alpha(\tau) = (\gamma/N)\delta(\tau)$, the model exhibits a 1st-order DPT at the critical point $V_c^M = \gamma/2$ [32], separating a steady state phase, where $\langle S_z\rangle \to -N/2$ ($V < V_c^M$) to a phase where $\langle S_z\rangle \to 0$ ($V > V_c^M$) for $N \to \infty$, as can be seen in Fig. 2(a). Here, we generalize the study of this DPT to the non-Markovian regime by considering a finite memory time for the bath with a correlation function of the form $\alpha(\tau) = Ge^{-\kappa|\tau| - i\omega\tau}$, as if the damping of the collective spin was originating from the coupling of the system to a *structured* bath via an interaction Hamiltonian $H_{\mathrm{int}} = \sqrt{G}(S_- a^\dagger + S_+ a)$ with $G = \gamma\kappa/(2N)$ and a the annihilation operator of a damped pseudomode of Hamiltonian $H_E = \omega a^\dagger a$. This model allows us to study non-Markovian effects on the DPT and compare them to the Markovian case by tuning the "loss" rate κ of the pseudomode. Indeed, the collective spin

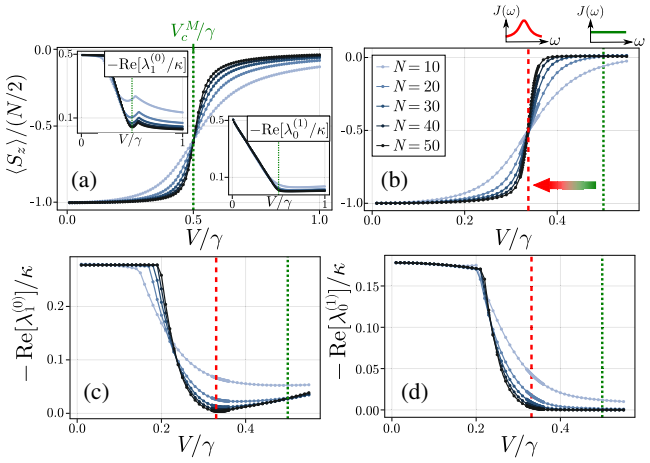

FIG. 2. Signatures of the 1st-order DPT for the generalized dissipative LMG model (6) obtained from $\mathcal{L}_{\mathrm{HEOM}}$, showing how environmental spectral structures affect the DPT. (a),(b) Steady state magnetization $\langle S_z\rangle$ as a function of V/γ for $\kappa/\omega = 50$ (a) and $\kappa/\omega = 1$ (b). The vertical green and red dashed lines indicate the critical points for $\kappa/\omega = 50$ and 1, respectively. (c),(d): Liouvillian gap $-\mathrm{Re}[\lambda_1^{(0)}]$ (c) and $-\mathrm{Re}[\lambda_0^{(1)}]$ (d) as a function of V/γ, indicating, respectively, the DPT and the SSB associated with the DPT. The insets of (a) show the same quantities for the Markovian case. Truncation orders are $k_{\max} = 2$ (a) and $k_{\max} = 6$ ($N = 10$–30), 7 ($N = 40$), 9 ($N = 50$) (b)–(d).

and the pseudomode form an extended Markovian system governed by the master equation

$$\dot{\rho}_{\mathrm{tot}} = -i[H, \rho_{\mathrm{tot}}] + \kappa\mathcal{D}_a[\rho_{\mathrm{tot}}] \tag{8}$$

with $H = H_{\mathrm{LMG}} + H_E + H_{\mathrm{int}}$. Adiabatic elimination of the pseudomode's degrees of freedom recovers Eq. (7) in the limit $\kappa \to \infty$ (see Supplemental Material [59]), as expected since $\alpha(\tau) \to (\gamma/N)\delta(\tau)$ for $\kappa \to \infty$. When κ is finite, memory effects arise and affect the DPT as described below. Note that Eq. (8) has a $\mathbb{Z}_2$ symmetry represented by $\mathcal{U}_2 = U_2 \cdot U_2^\dagger$ with $U_2 = e^{i\pi(S_z + a^\dagger a)}$. $\mathcal{U}_2$ has two distinct eigenvalues $u_k = e^{ik\pi} = \pm 1$ with $k = 0, 1$, so there are two symmetry sectors, with $L_{k=0}$ containing ρ_{ss}.

The impact of memory effects on the DPT based on $\mathcal{L}_{\mathrm{HEOM}}$ for the spin system can be seen in Fig. 2. First, we see in panel (b) that the steady state spin magnetization $\langle S_z\rangle$ exhibits a sharp transition at a critical point smaller than in the Markovian case shown in Fig. 2(a). This demonstrates that deviations from a flat spectral density can reshape phase boundaries. A mean-field analysis of (8) shows that the shift in the critical point increases as κ decreases (see Supplemental Material for all details [59]). Physically, this can be understood as follows: the smaller κ, the greater the probability that excitations escaping from the system will be reabsorbed at later times. The degree of openness of the system therefore decreases as κ decreases, which leads to a stabilization of the phase dominated by the Hamiltonian (6)

for small V. For $\kappa \to 0$ (i.e., for a closed system), the phase transition disappears because the Hamiltonian dynamics no longer competes with dissipative dynamics. In the opposite limit $\kappa \to \infty$, we recover the Markovian case. The HEOM Liouvillian spectrum correctly captures all DPT signatures. Indeed, it captures the emergence of both the level touching at the critical point in the symmetry sector $k = 0$, i.e., $-\mathrm{Re}[\lambda_1^{(0)}] \to 0$ as $N \to \infty$ and the SSB associated to the DPT, i.e., $-\mathrm{Re}[\lambda_0^{(1)}] \to 0$ for V above the critical point as $N \to \infty$, as can be seen in panels (c) and (d).

Note that this DPT cannot be studied via an approximate reduced description of the spin dynamics obtained after adiabatic elimination of the pseudomode, as the related mean-field approach predicts qualitatively different steady states (see Supplemental Material [59]). In general, reduced descriptions cannot account for all the features of a DPT and can even fail to capture DPTs, as elaborated on further below and strongly motivates again the use of our framework.

2nd-order DPT—The second model we consider, also experimentally relevant for cavity QED [63], is of the form (1) with $H_S = H_{\mathrm{LMG}} + hS_z$ and $L = S_x$. For an unstructured bath with $\alpha(\tau) = \gamma\delta(\tau)$, the system dynamics is governed by the master equation

$$\dot{\rho} = -i[H_{\mathrm{LMG}} + hS_z, \rho] + \frac{\gamma}{2N}\mathcal{D}_{S_x}[\rho], \tag{9}$$

whose unique steady state is the maximally mixed state $\rho_{\mathrm{ss}} \propto \mathbb{1}_{N+1}$, preventing the emergence of any DPT. However, if we add again a realistic finite memory time for the bath by considering $\alpha(\tau) = (\gamma\kappa/2N)e^{-i\omega\tau-\kappa|\tau|}$, we unveil the existence of two consecutive 2nd-order DPTs separating three different phases upon varying the squeezing strength V. This can be seen in Fig. 3, where we show that our approach captures all the features of the DPTs in agreement with mean-field predictions detailed in the Supplemental Material [59]. Panels (a)–(c) show the steady states expectations $\langle S_z\rangle$ and $\langle S_y^2\rangle$ as a function of V, which distinguish the phases [labeled as (I), (II), and (III)], as we have $\langle S_y^2\rangle = 0$ in phases (I) and (II) and $\langle S_z\rangle = -N/2$ in phase (II) only. In addition, as there is a $\mathbb{Z}_2$ symmetry represented by $\mathcal{U}_2$ in our model akin to the symmetry that is broken in the DPT of the Dicke model [22], we expect a SSB manifesting as $\lambda_0^{(1)} \to 0$ as $N \to +\infty$ in phases (I) and (III), accompanied by an exponential closure of the gap [23]. This behavior is illustrated in panels (b) and (d). Also, note that as the critical points are at $V = -h + \gamma\kappa\omega/[2(\kappa^2+\omega^2)]$ and $V = h$, taking the limit $\kappa \to \infty$ does not recover the prediction of the Lindblad scenario, i.e., no DPT. In other words, we have $\lim_{\kappa\to\infty}\lim_{N\to\infty} \neq \lim_{N\to\infty}\lim_{\kappa\to\infty}$. Physically, taking $\kappa \to \infty$ amounts to consider equal absorption and emission rates for the system, pushing it inevitably to the infinite temperature state. Considering a finite κ restores a memory for the bath, i.e., a system-frequency-dependent response, thereby providing the necessary competition between Hamiltonian and dissipative dynamics for the emergence of DPTs.

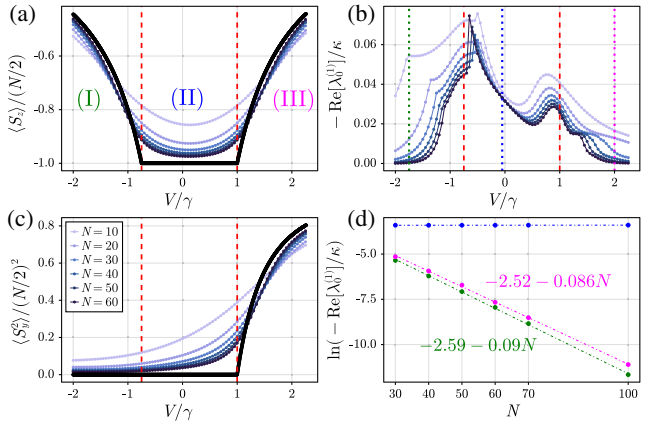

FIG. 3. Signatures of the 2nd-order DPT of the second model obtained from $\mathcal{L}_{\mathrm{HEOM}}$, showing the emergence of three phases [(I), (II), and (III)] as V/γ is varied. (a) and (c) Steady-state values of $\langle S_z\rangle$ and $\langle S_y^2\rangle$ as a function of V/γ for different N, allowing for distinguishing the phases. The solid black lines correspond to mean-field predictions and the vertical dashed red lines indicate the critical points. (b) Real part of $\lambda_0^{(1)}$ (i.e., the gap) as a function of V/γ, signaling the SSBs associated with the DPT. Three vertical dotted lines indicate the values of V/γ taken for the finite-size scaling of the gap shown in panel (d), i.e., $V/\gamma = -1.75$ (green), -0.05 (blue), 2 (pink), revealing an exponential closure of the gap in phases (I) and (III), as shown by the straight line fits. Parameters $\omega = \kappa = 2h = 2\gamma$. Truncation orders $k_{\max} = 6$ ($N = 10$–30), 7 ($N = 40$–60), 9 ($N = 70$, 100).

Conclusion—We applied the spectral theory of non-Markovian dissipative phase transitions developed in the companion paper [24] to demonstrate non-Markovian reservoir engineering of a 1st-order DPT with a discrete SSB and how DPTs can be genuinely triggered by non-Markovian effects. Our Letter highlights the importance of exploring out-of-equilibrium matter phases beyond the idealized Markovian limit, featuring non-Markovianity as a resource for triggering or controlling them. This is so far uncharted territory as most works dealing with dissipative many-body dynamics is generally constrained to Lindblad dissipation, which potentially hinders the evidences of DPTs [22,23].

There are many perspective of our Letter, such as studies of initial system-bath correlations [64] or connections in the non-Markovian regime between DPTs and symmetry breaking [65,66], geometric phase curvature [67,68], or dynamical [69,70] or measurement-induced [71,72] phase transitions, or dissipation engineering of long-range order [73], also for systems with non-Lorentzian environments that exhibit criticality [26,64,74].

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