Editors' Suggestion

Reaction-Limited Quantum Reaction-Diffusion Dynamics

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We consider the quantum nonequilibrium dynamics of systems where fermionic particles coherently hop on a one-dimensional lattice and are subject to dissipative processes analogous to those of classical reaction-diffusion models. Particles can either annihilate in pairs, $A + A \to \emptyset$, or coagulate upon contact, $A + A \to A$, and possibly also branch, $A \to A + A$. In classical settings, the interplay between these processes and particle diffusion leads to critical dynamics as well as to absorbing-state phase transitions. Here, we analyze the impact of coherent hopping and of quantum superposition, focusing on the so-called reaction-limited regime. Here, spatial density fluctuations are quickly smoothed out due to fast hopping, which for classical systems is described by a mean-field approach. By exploiting the time-dependent generalized Gibbs ensemble method, we demonstrate that quantum coherence and destructive interference play a crucial role in these systems and are responsible for the emergence of locally protected dark states and collective behavior beyond mean field. This can manifest both at stationarity and during the relaxation dynamics. Our analytical results highlight fundamental differences between classical nonequilibrium dynamics and their quantum counterpart and show that quantum effects indeed change collective universal behavior.

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Introduction.—In reaction-diffusion (RD) models, classical reactants, or particles, are transported by diffusion and react when they meet, see, e.g., Refs. [1–3]. These are paradigmatic nonequilibrium systems displaying universal dynamical properties and stationary-state transitions from fluctuating phases to absorbing states, i.e., states that once reached cannot be left. In one dimension, in particular, spatial fluctuations of the particle number dominate the kinetics and both exact analytical results [4–9] and dynamical field-theory renormalization calculations [10–16] have shown that the dynamical critical behavior is universal and it is not captured by the mean-field approximation. This is especially true in the "diffusion-limited" regime, i.e., when the diffusive mixing of the particles is not too strong [4,6,17–19]. In the opposite "reaction-limited" regime, where the diffusive motion is fast, the density of reactants rapidly uniformize (leading to the alternative name of well-stirred-mixture approximation) and one recovers mean-field results [1,2,11,20,21].

Quantum effects can alter the universal properties of absorbing-state phase transitions. This has been shown for Markovian open quantum systems [22–30], for systems with kinetic constraints [31–45], and for the quantum contact process [38,46]. Quantum dissipative RD spin chains, where the diffusive motion is replaced by coherent hopping, have been investigated in Ref. [47]. However, results in this and other works are limited to small systems, due to the complexity of the numerical simulation of many-body quantum dynamics. As a consequence, very little is known about the impact of quantum effects on universal aspects of RD dynamics and on absorbing-state phase transitions.

In this Letter, we make progress in this direction, deriving exact analytical results for the case of reaction-limited open quantum RD processes in fermionic chains. We consider a series of prototypical reaction processes, such as annihilation $A + A \to \emptyset$, coagulation $A + A \to A$, and branching $A \to A + A$ (see Fig. 1), and show that the reaction-limited regime of quantum RD models cannot be described within a mean-field approach, in stark contrast to the classical settings. We demonstrate that the presence of quantum effects strongly affects the approach to stationarity and the stationary state itself. For annihilation and coagulation, the density of particles features an algebraic (power-law) decay. This power law changes and may deviate from the mean-field predictions when the initial state of the dynamics features quantum coherence. In the presence of the branching process, quantum RD models display an absorbing-state phase transition. Here, annihilation processes that couple to coherent superpositions of adjacent particle pairs lead to the emergence of dark states that are locally protected against dissipation. These local dark states, which are not captured by the mean-field approach, establish quantum correlations between fermionic particles.

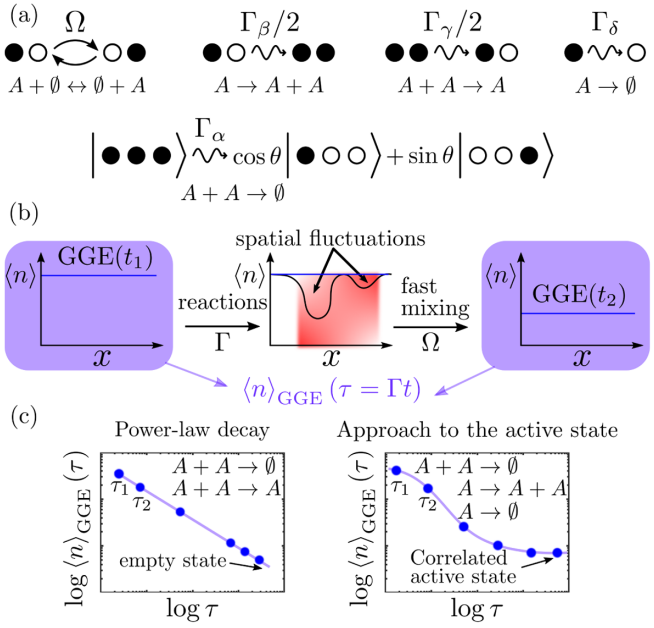

FIG. 1. Quantum RD dynamics in the reaction-limited regime. (a) Quantum chain with sites that can either be occupied by a fermion $|\cdots \bullet_j \cdots\rangle$ or empty $|\cdots \circ_j \cdots\rangle$. Particles can hop between nearest-neighboring sites with hopping rate Ω, Eq. (2). Dissipation consists of irreversible reactions at rate Γ_ν, Eqs. (4)–(6). The parameter θ controls coherent superposition from pair annihilation events. (b) In the reaction-limited regime, $\Gamma \ll \Omega$, reaction dynamics is slow and takes place on the timescale $\sim\Gamma^{-1}$. Fast hopping rapidly smooths out spatial fluctuations (highlighted in red), due to local reactions, and the state of the systems is described by a homogeneous GGE(τ) (blue horizontal lines) at any rescaled time $\tau = \Gamma t$. (c) The total particle density $\langle n \rangle_{\rm GGE}(\tau)$ decays algebraically in rescaled time τ (blue points) for annihilation or coagulation with exponent dependent on initial-state coherence. When branching is included, an absorbing-state phase transition to an active, finite density of particles, state can occur. The latter displays correlation when $\theta \neq 0, \pi/2$.

Our analysis is performed by exploiting the time-dependent generalized Gibbs ensemble method (TGGE) [48–51], which naturally leads to large-scale Boltzmann-like equations. The latter provides an exact description for the reaction-limited regime in the thermodynamic limit. Our analytical findings show that quantum effects lead to rich nonequilibrium behavior, significantly different from that of classical systems. Our results connect to the physics of cold atoms, where losses are of central experimental [52–59] and theoretical [60–67] relevance.

Quantum reaction-diffusion models.—We consider fermionic quantum chains of length L. Each site j can be either occupied $n_j|\cdots \bullet_j \cdots\rangle = |\cdots \bullet_j \cdots\rangle$ or empty $n_j|\cdots \circ_j \cdots\rangle = 0$, where $n_j = c_j^\dagger c_j$ and the operators c_j, $c_j^\dagger$ obey the fermionic anticommutation relations $\{c_j, c_{j'}^\dagger\} = \delta_{j,j'}$. The fermionic statistics prevents double occupancy of lattice sites, typically assumed in RD classical models [1–3]. The dynamics is ruled by the quantum master equation [68–70] ($\hbar = 1$ henceforth)

$$\dot{\rho}(t) = -i[H, \rho(t)] + \mathcal{D}[\rho(t)]. \tag{1}$$

Here, we assume that the diffusive motion of the particles in classical RD models is replaced by coherent hopping, which is accounted for by the quantum Hamiltonian

$$H = -\Omega \sum_{j=1}^{L} (c_j^\dagger c_{j+1} + c_{j+1}^\dagger c_j), \tag{2}$$

with Ω the hopping rate [cf. Fig. 1(a)]. Such Hamiltonian is diagonalized with Fourier-space fermionic operators $\hat{c}_k$, $\hat{c}_k^\dagger$, where k is the quasimomentum, and the number operators $\hat{n}_k = \hat{c}_k^\dagger \hat{c}_k$ [71]. It conserves the total number $N = \sum_j n_j = \sum_k \hat{n}_k$ of particles: $[H, N] = 0$. The irreversible reaction processes are encoded in the dissipator $\mathcal{D}$. It takes the (Lindblad) form [68–70]

$$\mathcal{D}[\rho] = \sum_{j,\nu} \left[L_j^\nu \rho L_j^{\nu\dagger} - \frac{1}{2} \left\{ L_j^{\nu\dagger} L_j^\nu, \rho \right\} \right], \tag{3}$$

where L_j^ν are local jump operators. We consider four different reactions, labeled by the parameter ν, which are sketched in Fig. 1(a). The first is binary annihilation, $A + A \to \emptyset$, of a pair of neighboring particles (rate Γ_α), which is described by the jump operators

$$L_j^\alpha = L_j^\alpha(\theta) = \sqrt{\Gamma_\alpha} c_j (\cos\theta c_{j+1} - \sin\theta c_{j-1}). \tag{4}$$

The sum of the two terms, whose balance is controlled by the angle $\theta \in [0, \pi)$, allows for the possibility that interference between two quantum mechanical amplitudes contributes to the pair annihilation process. Such structure naturally emerges in the Bose-Hubbard model subject to strong two-body losses. In this limit, the model can be mapped to free fermions (2) with weak, $\Gamma_\alpha \ll \Omega$, two-body losses (4), as shown in Refs. [52,60,62]. The classical-incoherent annihilation process is recovered for $\theta = 0, \pi/2$. The second reaction is coagulation, $A + A \to A$, of a particle upon meeting a neighboring one (rate $\Gamma_\gamma/2$), with jump operators

$$L_j^{\gamma\pm} = \sqrt{\Gamma_\gamma/2} c_j n_{j\pm1}. \tag{5}$$

The third reaction is one-body annihilation, $A \to \emptyset$, (rate Γ_δ) with jump operators

$$L_j^\delta = \sqrt{\Gamma_\delta} c_j. \tag{6}$$

These three reactions break number conservation and, due to continued particle loss, drive the system toward an absorbing state devoid of particles. To establish a nontrivial

steady state, we consider a fourth reaction, namely, branching, $A \to A + A$. This process allows for creation of a particle in the neighborhood of an occupied site (rate $\Gamma_\beta/2$)

$$L_j^{\beta\pm} = \sqrt{\Gamma_\beta/2}\, c_j^\dagger n_{j\pm1}. \tag{7}$$

The competition between the branching process and one-body annihilation (as in the contact process [2,3]) gives rise to a nonequilibrium absorbing-state phase transition, from the empty state to a stationary active one with finite density of particles. Coagulation (5) and branching (7) can be experimentally implemented in the facilitation regime [72] of cold-atomic gases dressed with Rydberg interactions [73–75]. For convenience, in the following, when multiple reactions are present, we rescale rates as $\Gamma_\nu = \Gamma\nu$, so that Γ sets the timescale of the dissipation, while the dimensionless parameters α, β, γ, and δ encode the relative strength of the reactions [see Figs. 1(b) and 1(c)].

There are two important timescales in the dynamics: the reaction time $\sim\Gamma^{-1}$, which gives the typical time needed for neighboring particles to react, and the hopping time (or diffusion time in classical RD models) $\sim\Omega^{-1}$, which sets the timescale for two reacting particles to meet. In classical settings [1,2], the dynamics qualitatively changes depending on the ratio Γ/Ω. The regime with $\Gamma/\Omega \gg 1$ is named "diffusion limited," as the propagation of particles is the limiting factor for reactions to occur. In this regime, spatial fluctuations are relevant and in one dimension the total particle density $\langle n\rangle(t) = \langle N\rangle(t)/L$ decays algebraically as $\langle n\rangle(t) \sim (\Omega t)^{-1/2}$ [4–9,17–19], which is slower than the corresponding mean-field prediction $\langle n\rangle_{\rm MF}(t) \sim (\Gamma t)^{-1}$ (note the different rescaling of time).

The opposite regime, $\Gamma/\Omega \ll 1$, is the "reaction-limited" one. Here, spatial fluctuations are irrelevant, as fast motion makes the particle density homogeneous in space. For classical systems [1,2,11,20,21] this regime is described by law of mass action rate equations, which assert that the rate of change of reactants is proportional to the product of their global densities. This approach disregards spatial correlations among particles and it indeed reproduces the mean-field result $\langle n\rangle_{\rm MF}(t) \sim (\Gamma t)^{-1}$. In what follows, we consider the quantum analog of this regime, see Figs. 1(b) and 1(c). As we show, this regime is much richer than its classical counterpart, as coherent effects give rise to collective behavior and quantum correlations beyond mean field.

Reaction-limited TGGE.—For our quantum RD models, the reaction-limited [55] regime $\Gamma/\Omega \ll 1$ is equivalent to a weak dissipation limit, which can be analyzed with the recently proposed time-dependent generalized Gibbs ensemble (TGGE) of Refs. [48–51]. Because of fast hopping, one can consider the state of the system $\rho(t)$ to be relaxed with respect to the stationary manifold of the Hamiltonian, $[H, \rho(t)] = 0$, at any time t. The dynamics of $\rho(t)$ within this manifold is set by the timescale Γ^{-1} and it is determined by the dissipation. This aspect is pictorially shown in Fig. 1(b). The TGGE approach then makes an ansatz among the set of relaxed states of the Hamiltonian, which is the GGE, see, e.g., Refs. [76,77]. In the specific case of the Hamiltonian (2), the GGE can be written as

$$\rho_{\rm GGE}(t) = \frac{1}{\mathcal{Z}(t)} \exp\left(-\sum_k \lambda_k(t)\hat{n}_k\right), \tag{8}$$

where $\mathcal{Z}(t) = \prod_k [1 + e^{-\lambda_k(t)}]$. The GGE state (8) describes averages $\langle\ldots\rangle_{\rm GGE}(t)$ of local observables in the thermodynamic limit. It is entirely fixed from the knowledge of the Lagrange multipliers $\lambda_k(t)$ or, equivalently, of the occupation functions $\langle\hat{n}_q\rangle_{\rm GGE}(t) = C_q(t)$, which obey the equations [62–64,67]

$$\frac{dC_q(t)}{dt} = \sum_{j,\nu} \langle L_j^{\nu\dagger}[\hat{n}_q, L_j^\nu]\rangle_{\rm GGE}(t), \qquad \forall q. \tag{9}$$

The solution $C_q(\tau)$ of this equation clearly depends on the rescaled time $\tau = \Gamma t$, consistent with the above discussion on the reaction-limited regime. The equation of motion (9) describes the large-scale dynamics of the system and it has a structure akin to the Boltzmann equation. The right-hand side can be, crucially, exactly computed in the GGE state (8) through Wick's theorem. To explore the impact of quantum-coherent effects on the RD dynamics, we consider two different initial conditions for Eq. (9). The first is the coherent Fermi-sea (FS) state with density filling $0 < n_0 \le 1$: $C_q(t=0) = 1$ if $q \in [-\pi n_0, \pi n_0]$, and zero otherwise. The second is the incoherent state $\rho_0 = \exp(-\lambda N)/\mathcal{Z}_0$, with a flat initial distribution in momentum space, $C_q(0) = n_0$.

Annihilation and coagulation.—In Fig. 2(a), we plot, from Eq. (9) (see Supplemental Material [78]), the particle density as a function of time for the pair annihilation reaction only ($\Gamma_\gamma = \Gamma_\beta = \Gamma_\delta = 0$), Eq. (4) with $\theta = 0$, so that interference effects are excluded. The density decays as $\langle n\rangle_{\rm GGE}(\tau = \Gamma_\alpha t) \sim (\Gamma_\alpha t)^{-1/2}$ for the FS initial state for any filling $n_0 \neq 1$. The $1/2$ decay exponent does not necessarily require considering pure states. It also occurs for initial mixed states with an inhomogeneous in q initial occupation function $C_q(0)$ [78]. In contrast, for the initial state ρ_0 and any n_0, the law of mass action is recovered and the density is exactly given by mean field, $\langle n\rangle_{\rm MF}(\tau) \sim (\Gamma_\alpha t)^{-1}$. This shows the relevance of coherent effects in the critical dynamics of the model, since the algebraic decay of the density in the reaction-limited regime is not described by the mean-field approximation whenever the initial state is quantum coherent. In the latter case, the decay of the particle density is slower than in the classical counterpart of the model, where only incoherent initial states are possible

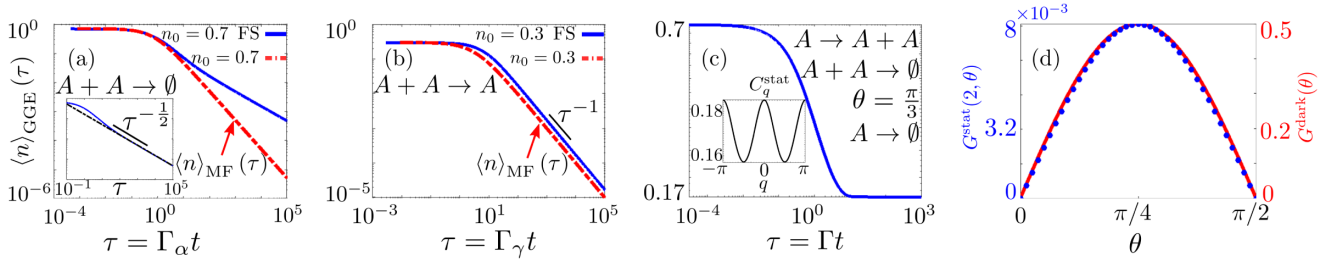

FIG. 2. Dynamics and active phase in quantum reaction-limited RD systems. (a) Log-log plot of the particle density $\langle n \rangle_{\rm GGE}(\tau)$ as a function of the rescaled time $\tau = \Gamma_\alpha t$ for the binary annihilation reaction (4) with $\theta = 0$. In the top-blue curve, the initial state is the coherent Fermi sea (FS) state with filling $n_0 = 0.7$. The density decays asymptotically as a power law $\langle n \rangle_{\rm GGE}(\tau) \sim \tau^{-1/2}$. Inset: the black dashed curve is a power-law fit $\langle n \rangle_{\rm GGE}(\tau) = a\tau^{-b}$ performed over the time window $\tau \in [10^6, 10^7]$, with the resulting fitting parameter for the exponent being $b = 0.50025 \pm 5 \times 10^{-5}$. In the red dashed curve, the initial state is the incoherent state ρ_0 with the same mean density $n_0 = 0.7$. In this case, the density is exactly described by the mean-field (MF) law of mass action and $\langle n \rangle_{\rm GGE}(\tau) = \langle n \rangle_{\rm MF}(\tau) \sim \tau^{-1}$. (b) Log-log plot of the density of particles $\langle n \rangle_{\rm GGE}(\tau)$ as a function of $\tau = \Gamma_\gamma t$ for the coagulation reaction (5). The top blue curve corresponds to the FS initial state at filling $n_0 = 0.3$, while the red dashed one corresponds to the incoherent state ρ_0 at the same filling. For the FS state, the asymptotic exponent $\langle n \rangle_{\rm GGE}(\tau) \sim \tau^{-1}$ is the same as in MF. (c) Log-log plot of the density as a function of $\tau = \Gamma t$ for the CP with pair annihilation Eqs. (4)–(6) and $\Gamma_\gamma = 0$, from the FS initial state at $n_0 = 0.7$. For $\beta > \delta$ an active stationary state is reached. The associated stationary momentum distribution function $C_q^{\rm stat}$ is shown in the inset as a function of q. (d) Stationary correlations $G^{\rm stat}(2,\theta)$ at distance 2 (left, blue axis) and dark state contribution $G^{\rm dark}(\theta) = \sin(2\theta)/2$ (right, red axis) in the CP as a function of θ. Parameters are $\beta = \alpha = 1$, $\delta = 0.5$.

and the long-time behavior of the density is independent on the initial density n_0 [79–81].

In Fig. 2(b), we plot the particle density as a function of time for the coagulation reaction only ($\Gamma_\alpha = \Gamma_\beta = \Gamma_\delta = 0$), Eq. (5) [78]. We find that $\langle n \rangle_{\rm GGE}(\tau = \Gamma_\gamma t) \sim (\Gamma_\gamma t)^{-1}$ both for the incoherent state ρ_0 and for the FS state. For all initial conditions, we see mean-field-like decay [82], which is different from the situation for pair annihilation at $\theta = 0$, Fig. 2(a). This difference between annihilation and coagulation processes is in stark contrast with classical RD models, where both processes belong to the same universality class and decay in the same way independent of initial conditions [5,6,18,80,81,83–85].

For quantum RD, only when starting from the incoherent initial state ρ_0 annihilation and coagulation behave in a similar way. In fact, the densities $\langle n \rangle^{\rm ann}_{\rm GGE}(\tau, n_0)$ and $\langle n \rangle^{\rm coag}_{\rm GGE}(\tau, n_0)$ obey

$$\langle n \rangle^{\rm coag}_{\rm GGE}(\tau, n_0) = 2\langle n \rangle^{\rm ann}_{\rm GGE}(\tau, n_0/2), \tag{10}$$

for $\Gamma_\alpha = \Gamma_\gamma$. Equation (10) is proved noting that the dynamics from the incoherent state ρ_0 according to Eq. (9) remains at all times fully incoherent and the quantum master equation (1) can then be mapped onto a classical master equation (see Supplemental Material [78]). For the coherent FS initial state, off-diagonal elements of the density matrix $\rho(t)$ are relevant, the quantum master equation does not reduce to its classical counterpart, and Eq. (10) does not apply. This shows that the quantum RD annihilation and coagulation processes do not generically belong to the same universality class and they can display different asymptotic behavior.

Contact process.—We now consider the contact process (CP) with pair annihilation, cf. Eqs. (4)–(6) with $\Gamma_\nu = \Gamma\nu$ ($\nu = \alpha, \beta, \delta$) and $\Gamma_\gamma = 0$ and Fig. 1(c). In Fig. 2(c), we plot the density as a function of the rescaled time $\tau = \Gamma t$. We find a phase transition between an absorbing and an active state: the stationary-state density $\langle n \rangle^{\rm stat}_{\rm GGE}$ becomes nonzero when $\beta > \beta_c$, with $\beta_c = \delta$ independent of α and θ. This β_c is the same as that of the mean-field classical CP [2,3]. Furthermore, we find that the associated critical exponents for the stationary density $\langle n \rangle^{\rm stat}_{\rm GGE} \propto (\beta - \beta_c)^1$ and for the decay of the density at the critical point β_c, $\langle n \rangle_{\rm GGE} \sim (\Gamma t)^{-1}$, are those of the (mean-field) directed percolation universality.

Interestingly, however, the stationary state is strongly affected by the quantum coherence introduced by the annihilation reaction in Eq. (4), beyond what can be predicted by a mean-field approach. The inset of Fig. 2(c) shows that the different quasimomenta q are not evenly populated in the stationary state. This applies when $\theta \neq 0$, $\pi/2$. The nontrivial structure of $C_q^{\rm stat}$ implies that the stationary state has spatial correlations. To quantify this, we compute the two-point fermionic correlation function $G^{\rm stat}(x - y, \theta) = \langle c_x^\dagger c_y \rangle^{\rm stat}_{\rm GGE}$, which for the mean-field (product) state would be zero unless $x = y$. We find that $G^{\rm stat}(l, \theta)$ is nonzero at even distances $l = 2, 4, 6\ldots$ with a dominant contribution at $l = 2$. The value of $G^{\rm stat}(2, \theta)$ as a function of θ is shown in Fig. 2(d) and is approximately equal to $A(\theta) = \varepsilon \sin(2\theta)/2$. Considering only these dominant next-to-nearest-neighbor correlations, we can identify the (approximate) Lagrange multipliers $\lambda_q^{\rm stat}$ for the stationary GGE $\rho^{\rm stat}_{\rm GGE}$ expanding to first order in $A(\theta)$ [since ε is small as shown in Fig. 2(d)]. One obtains $\lambda_q^{\rm stat} = \lambda_{\rm MF} + \lambda_2 \cos(2q)$ and therefore $\rho^{\rm stat}_{\rm GGE} \propto e^{-\lambda_{\rm MF} N - \lambda_2 Q_2/2}$, with

$Q_2 = \sum_j (c_j^\dagger c_{j+2} + c_{j+2}^\dagger c_j)$. The contribution $\lambda_{\mathrm{MF}} = \log(1/\langle n\rangle_{\mathrm{GGE}}^{\mathrm{stat}} - 1)$ represents the mean-field component of the state, while $\lambda_2 = -A(\theta)/[\langle n\rangle_{\mathrm{GGE}}^{\mathrm{stat}}(1 - \langle n\rangle_{\mathrm{GGE}}^{\mathrm{stat}})]$ accounts for deviations from it. We show in the Supplemental Material [78] that $\rho_{\mathrm{GGE}}^{\mathrm{stat}}$ can be written in terms of an incoherent state plus a coherent correction, where projectors onto the "local dark states"

$$|\psi\rangle_j^{\mathrm{dark},\circ/\bullet} = \pm\cos\theta|\bullet(\circ/\bullet)_j\circ\rangle + \sin\theta|\circ(\circ/\bullet)_j\bullet\rangle, \quad (11)$$

emerge out of the uncorrelated mean-field state. The states $|\psi\rangle_j^{\mathrm{dark},\circ/\bullet}$ are both dark with respect to the annihilation process (4) centered in j, i.e., $L_j^\alpha(\theta)|\psi\rangle_j^{\mathrm{dark},\circ/\bullet} = 0$. Moreover, $|\psi\rangle_j^{\mathrm{dark},\bullet}$ is dark to branching (7) in j and is connected through one-body annihilation (6) in j to the state $|\psi\rangle_j^{\mathrm{dark},\circ}$. These local dark states determine the correlations $G^{\mathrm{stat}}(2,\theta)$ in $\rho_{\mathrm{GGE}}^{\mathrm{stat}}$, as shown in Fig. 2(d).

Summary.—We provided a fully analytical treatment of quantum many-body RD systems in their reaction-limited regime, where the irreversible reaction rates are much smaller than the coherent hopping rate. While for classical RD models, this regime is well described by a mean-field approach, we have shown that quantum RD displays instead much richer behavior. In particular, for annihilation, quantum coherence in the initial state can give rise to an algebraic density decay whose power-law exponent differs from the mean-field one. Furthermore, we have shown that quantum annihilation and coagulation do not belong to the same universality class. For the contact process plus pair annihilation, we have found that the stationary state can feature correlations, which emerge as a consequence of destructive interference. This inherently quantum feature gives rise to locally protected and correlated dark states. The RD systems discussed here connect the soft-matter physics of chemical reactions to that of cold atoms, where reactions translate into dissipative particle losses or creations [53–67], which can be implemented via Rydberg dressing [73–75]. Quantum reaction-diffusion systems are an ideal benchmark to investigate the impact of quantum effects on large-scale universal properties via numerical methods [38,39,46] and dynamical Keldysh-field theory [86,87].

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