Steady-state crystallization of Rydberg excitations in an optically driven lattice gas

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We study the conditions for attaining crystalline order in the stationary state of a continuously driven, open many-body system. Specifically, we consider resonant optical excitations of atoms in a one-dimensional lattice to the Rydberg states interacting via the van der Waals potential. Strong blockade of excitations at neighboring lattice sites steers the system toward a crystalline state while competing with the fluctuations associated with relaxation. We analyze the stationary state of the many-body system and the dynamics of its buildup employing numerically exact time-dependent density-matrix renormalization-group simulations for two- and three-level excitation schemes. We also present an approximate rate equation model which provides qualitative conditions for attaining crystalline order.

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The strong, long-range dipole-dipole or van der Waals (vdW) interactions between Rydberg atoms [1] have positioned them as promising systems for quantum information processing [2–4], which has led to considerable experimental efforts in exploring such systems [5–12]. Coupling Rydberg atoms to light can give rise to novel photonic states with highly nonclassical correlations [13,14] and unusual nonlinear spectroscopic features [15,16]. Rydberg atoms are also interesting for studies of many-body physics: It was predicted that long-range interactions lead to spontaneous symmetry breaking and crystalline order in a continuous [17,18] or lattice gas [19,20], formation of supersolids [21], and vortex lattices [22] or fractional quantum Hall states [23] in two-dimensional (2D) systems. Hints of the crystalline order of Rydberg excitations were observed in a continuous system [24], while very recently the short-range order of Rydberg excitations was demonstrated in an optically driven lattice gas [25].

Most of the theoretical work on many-body physics with Rydberg atoms deals with the ground states of model Hamiltonians and ignores dissipation and decoherence associated with the optical excitation, which become relevant on time scales necessary for the buildup of correlations. A more natural approach, closer to experimental reality, is therefore to consider correlations in the *stationary state* of open, continuously driven system [26–29]. Importantly, the stationary state is an attractor of the dynamics of the system and is immune to small perturbations. Recent work has shown that judiciously engineered reservoirs can generate correlated dark states or induce spatial coherence by dissipation [30–32]. In general, however, fluctuations associated with relaxation in open systems tend to destroy order favored by the interactions. Under what conditions long-range order survives the competition between interactions and dissipation is an open question, beyond the established theory of quantum phase transitions in unitary systems. Here we address this question in the context of a specific but experimentally relevant system: a one-dimensional (1D) lattice of atoms optically excited to the strongly interacting Rydberg states. We employ numerically exact time-dependent density-matrix renormalization-group (t-DMRG) simulations and an approximate rate equations (REs) model to derive general conditions for attaining the crystalline order of Rydberg excitations of the system in the stationary state and analyze the dynamics of its buildup. The coupling to the reservoir limits entanglement within the system, circumventing the usual bottleneck of t-DMRG, which permits full many-body calculations beyond exact diagonalization for a small system [29] or mean-field treatments [27,28].

We consider a chain of N atoms trapped in a 1D lattice potential of period a [Fig. 1(a)] and examine one- and two-photon resonant optical excitations of atoms from the ground state $|g\rangle$ to the Rydberg state $|r\rangle$ [Fig. 1(b)]. In the two-level scheme, the transition $|g\rangle \to |r\rangle$ is driven by a laser field of (effective) Rabi frequency Ω_{gr}. In the three-level scheme, the Rydberg state $|r\rangle$ is populated from the ground state $|g\rangle$ via resonant intermediate state $|e\rangle$ in the coherent population trapping (CPT) or dark-state resonance configuration [33] with Rabi frequencies $\Omega_{ge} \gtrsim \Omega_{er}$. The corresponding atom-field interaction Hamiltonians are given, respectively, by $\mathcal{V}_2^j = -\hbar(\Omega_{gr}\hat{\sigma}_{rg}^j + \text{H.c.})$ and $\mathcal{V}_3^j = -\hbar(\Omega_{ge}\hat{\sigma}_{eg}^j + \Omega_{er}\hat{\sigma}_{re}^j + \text{H.c.})$, where $\hat{\sigma}_{\mu\nu}^j \equiv |\mu\rangle_{jj}\langle\nu|$ are the transition operators for atom j. Spontaneous decay is described by Liouvillian terms in the equation of motion for the density operator ρ: $\mathcal{L}^j\rho = \frac{1}{2}(2\hat{L}^j\rho\hat{L}^{j\dagger} - \{\hat{L}^{j\dagger}\hat{L}^j, \rho\})$, where $\hat{L}^j$ are the Lindblad generators. For the two-level scheme, $\hat{L}_2^j = \sqrt{\Gamma_{rg}}\hat{\sigma}_{gr}^j$, with Γ_{rg} being the (population) decay rate of Rydberg state $|r\rangle$. Although Γ_{rg} is typically small ($\sim 10^4$ Hz), it may be comparable to the Rabi frequency Ω_{gr} associated with either direct one-photon (UV) transition $|g\rangle \to |r\rangle$ with a small dipole matrix element or two-photon transition via far-off-resonant intermediate states. For the three-level CPT scheme, we take into account only the high spontaneous decay rate Γ_{eg} ($\sim 10^7$ Hz) of the intermediate excited state $|e\rangle$ via $\hat{L}_3^j = \sqrt{\Gamma_{eg}}\hat{\sigma}_{ge}^j$; the decay rate Γ_{re} of Rydberg state $|r\rangle$ can be neglected in comparison with the loss rate $\Omega_{er}^2/\Gamma_{eg}$ due to the optical pumping through $|e\rangle$, which we verified numerically. Other losses from the Rydberg state should be small on the time scale of the buildup of correlations in the full system (see below).

For an isolated two- or three-level atom under continuous driving, the steady-state population of the Rydberg state is

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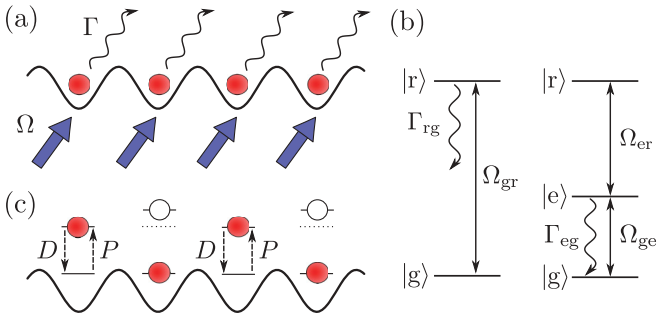

FIG. 1. (Color online) (a) Schematic of optically driven atoms in a lattice. (b) Starting from the ground state $|g\rangle$, atoms are resonantly excited to the Rydberg state $|r\rangle$ either directly (left) or via resonant intermediate state $|e\rangle$ (right). (c) Illustration of the rate equation model with Rydberg blockade of neighboring atoms.

given, respectively, by

$$\langle\hat{\sigma}_{rr}\rangle \approx \frac{|\Omega_{gr}|^2}{2|\Omega_{gr}|^2+\gamma_{rg}^2+\Delta^2}, \tag{1a}$$

$$\langle\hat{\sigma}_{rr}\rangle \approx \frac{|\Omega_{ge}|^2(|\Omega_{ge}|^2+|\Omega_{er}|^2)}{(|\Omega_{ge}|^2+|\Omega_{er}|^2)^2+\left(\gamma_{eg}^2+2|\Omega_{ge}|^2\right)\Delta^2}, \tag{1b}$$

where $\gamma_{\mu\nu}=\frac{1}{2}\Gamma_{\mu\nu}$ and Δ is the one- or two-photon detuning of the laser frequency from the $|g\rangle\rightarrow|r\rangle$ transition resonance. It follows from Eqs. (1) that the Rydberg population $\langle\hat{\sigma}_{rr}\rangle$ of an atom is a Lorentzian function of Δ, with the width $w=\sqrt{2|\Omega_{gr}|^2+\gamma_{rg}^2}$ for the direct excitation and $w=(|\Omega_{ge}|^2+|\Omega_{er}|^2)/\sqrt{\gamma_{eg}^2+2|\Omega_{ge}|^2}$ for the CPT excitation schemes.

Finally, pairs of atoms i and j interact with each other via the vdW potential [34] $\mathcal{V}_{\rm vdW}^{ij}=\hbar\hat{\sigma}_{rr}^{i}\frac{C_6}{d_{ij}^6}\hat{\sigma}_{rr}^{j}$, where $d_{ij}=a|i-j|$ is the interatomic distance. Once an atom i is excited to the Rydberg state $|r\rangle$, it shifts the atom j out of resonance by $\Delta=U/|i-j|^6$, where $U\equiv C_6/a^6$. The excitation of atom j to state $|r\rangle$ is then blocked if $\Delta\gtrsim w$, which determines the blockade distance $d_{\rm b}\simeq\sqrt[6]{C_6/w}$ [3]. Throughout this paper, we assume that $a<d_{\rm b}<2a$, so that there is a Rydberg blockade only between neighboring atoms. Note that for $N_{\rm b}>1$ atom per blockade volume, Ω_{ge} is replaced by the collective Rabi frequency $\sqrt{N_{\rm b}}\Omega_{ge}$ [12], with the blockade distance determined self-consistently [4,18,26].

The density matrix ρ of the system of N atoms obeys the master equation $\dot{\rho}=-\frac{i}{\hbar}[\mathcal{H}_l,\rho]+\mathcal{L}_l\rho$, with the Hamiltonian $\mathcal{H}_l=\sum_j\mathcal{V}_l^j+\sum_{i<j}\mathcal{V}_{\rm vdW}^{ij}$ and the Liouvillian $\mathcal{L}_l\rho=\sum_j\mathcal{L}_l^j\rho$ for $l=2$- or 3-level atoms. Since the interaction of an atom with its immediate neighbor is much stronger than that with atoms farther apart, we truncate the vdW potential $\mathcal{V}_{\rm vdW}^{ij}$ to nearest neighbor (NN): $\mathcal{V}_{\rm NN}^{ij}=\hbar\hat{\sigma}_{rr}^{i}U\hat{\sigma}_{rr}^{j}$ for $i=j-1$ and $\mathcal{V}_{\rm NN}^{ij}=0$ otherwise. We have performed exact numerical integrations of density-matrix equations for small systems of several ($N\leqslant 7$) atoms interacting via the $\mathcal{V}_{\rm vdW}$ and $\mathcal{V}_{\rm NN}$ potentials and verified that they yield similar results for both two- and three-level excitation schemes. We therefore employ the $\mathcal{V}_{\rm NN}$ potential henceforth, returning to the corrections due to longer-range interactions of the $\mathcal{V}_{\rm vdW}$ potential later.

We obtain the time evolution and the steady state of the full many-body density matrix employing the t-DMRG method [35,36]. Our implementation follows the original proposal of [37], generalized to open quantum systems [38,39]. Simulations for up to $N\sim 10^2$ atoms are possible, as the relaxation keeps the entanglement inside the system small: for our calculations, bond dimensions of $\chi=20$ turned out to be sufficient in the matrix-product decomposition. As a consequence, the usual limitations on the propagation time do not apply and the t-DMRG integration can be performed for, in principle, arbitrarily long times. Furthermore, since the stationary state is an attractor of the dynamics, accumulated errors self-correct. We verified that for small systems with the truncated potential $\mathcal{V}_{\rm NN}$, the exact and t-DMRG solutions are indistinguishable.

Results of the t-DMRG simulations for two- and three-level atoms in a realistically large lattice of length $30a$ with open boundary conditions are shown in Fig. 2. Since interactions between the atoms suppress Rydberg excitations, the atoms at the boundaries $j=1$ and $j=N$ having only one neighbor acquire the largest population of the Rydberg state $|r\rangle$. Next to the boundary, we observe period 2 spatial oscillations of excitation probabilities $\langle\hat{\sigma}_{rr}^{j}\rangle$. For two-level atoms [Fig. 2(a)], this edge effect decays within a few lattice sites. This is due to the fact that even under strong driving $\Omega_{gr}\gg\Gamma_{rg}$ the excited-state population of a noninteracting two-level atom saturates to $\langle\hat{\sigma}_{rr}\rangle\rightarrow\frac{1}{2}$ [cf. Eq. (1a)] with large fluctuations. In contrast, for a (noninteracting) three-level atom under the CPT excitation scheme, the population of the Rydberg state can be very large, $\langle\hat{\sigma}_{rr}\rangle\rightarrow 1$, when $\Omega_{ge}\gg\Omega_{er}$ [cf. Eq. (1b)]. For a chain of strongly interacting atoms, we then observe a high amplitude of spatial oscillations of $\langle\hat{\sigma}_{rr}^{j}\rangle$ extending over many lattice sites [Fig. 2(b)].

Atoms excited to Rydberg states are typically no longer trapped in an optical lattice. An important question, then, is whether the stationary state can be reached during times short

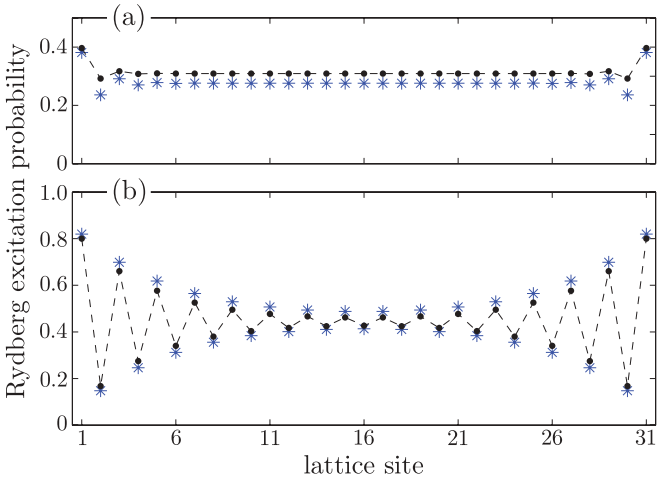

FIG. 2. (Color online) Steady-state Rydberg excitation probabilities $\langle\hat{\sigma}_{rr}^{j}\rangle$ of (a) two-level atoms and (b) three-level atoms in a lattice of length $30a$ ($N=31$ atoms). Filled (black) circles (connected by dashed lines) show the t-DMRG solutions of the density-matrix equation with potential $\mathcal{V}_{\rm NN}^{ij}$, and (blue) asterisks are the RE solutions for the hard-core (infinite-NN) potential. Parameters are (a) $U=2\Omega_{gr}$ and $\Gamma_{rg}=\frac{1}{4}\Omega_{gr}$ [$\langle\hat{\sigma}_{rr}\rangle\simeq 0.496$] and (b) $U=2\Omega_{ge}$, $\Gamma_{eg}=4\Omega_{ge}$, and $\Omega_{er}=\frac{1}{5}\Omega_{ge}$ [$\langle\hat{\sigma}_{rr}\rangle\simeq 0.961$].

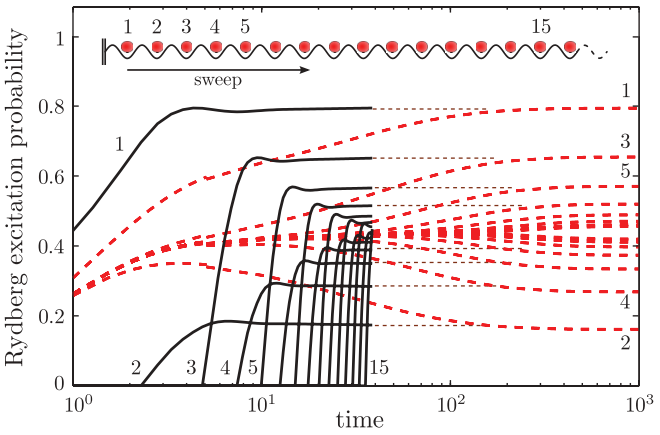

FIG. 3. (Color online) Time evolution of Rydberg excitation probabilities for simultaneous application of driving lasers at time $t = 0$ to all atoms [dashed (red) lines] and for sequential application—"sweep"—of the lasers to atoms $j = 1,2,\ldots,15$ at times $t_j = 2.4(j-1)T$ [thick solid (black) lines]. All parameters are as in Fig. 2(b) and time is in units of $T = \frac{\Gamma_{eg}}{\Omega_{ge}\Omega_{er}}$.

enough for the center-of-mass motion of the excited atoms to be negligible. In Fig. 3 we show the time evolution of Rydberg-state populations of individual atoms in a lattice with open boundary conditions. Initially, the atoms are in the ground state $|g\rangle$. Applying the driving lasers to all atoms at once [dashed (red) lines] yields the global stationary state of the system after times two to three orders of magnitude longer than the steady-state equilibration time $T = \Gamma_{eg}/(\Omega_{ge}\Omega_{er})$ of an isolated atom (note the logarithmic scale of the time axis). The reason for this is the initial formation of small crystalline domains with dislocation defects in between, which require excessive healing times. In the present system, the steady state is unique and therefore independent of the initial conditions and the details of preparation [40], such as, e.g., the way the light fields are switched on. This permits faster preparation of the global stationary state. Similarly to classical crystals, it is indeed much faster to "grow" the Rydberg quasicrystal by first applying the lasers to the atoms at the lattice boundary ($j = 1$) and then successively extending the irradiated region ($j = 2,3,\ldots$) until the full Hamiltonian is realized. In Fig. 3 we verify this intuitive approach [solid (black) lines for atoms $j = 1,2,\ldots,15$]. Since the crystalline order is rooted in the Rydberg blockade of the NN sites, a "sweep" velocity as fast as one lattice period a per single-atom equilibration time T can be applied.

We now describe an effective REs model, which yields an analytic solution for the stationary state of the system. We are interested in the Rydberg excitation probabilities $\langle\hat{\sigma}_{rr}^j\rangle$ of atoms and their steady-state correlations $\langle\hat{\sigma}_{rr}^j\hat{\sigma}_{rr}^k\rangle$, which allows us to disregard the coherences between the atoms. We restrict the interatomic interactions to the complete blockade of Rydberg excitations of NNs, assuming the potential $\mathcal{V}_{\rm NN}^{ij}$ with $U \to \infty$. At each lattice site, we then have two incoherent processes: a state-dependent pump with rate P and a de-excitation with rate D; the corresponding Lindblad generators are $\hat{L}_{\rm p}^j = \sqrt{P}\,(\hat{\sigma}_{rr}^{j-1} - \mathbb{1})\hat{\sigma}_{rg}^j(\hat{\sigma}_{rr}^{j+1} - \mathbb{1})$ and $\hat{L}_{\rm d}^j = \sqrt{D}\hat{\sigma}_{gr}^j$. The ratios of the two rates $\kappa_l \equiv \frac{P}{D} = \frac{\langle\hat{\sigma}_{rr}\rangle}{1-\langle\hat{\sigma}_{rr}\rangle}$ for the $l = 2$- and 3-level atoms are obtained from Eqs. (1) as

$$\kappa_2 = \frac{|\Omega_{gr}|^2}{|\Omega_{gr}|^2 + \gamma_{rg}^2}, \quad \kappa_3 = \frac{|\Omega_{ge}|^2}{|\Omega_{er}|^2}. \tag{2}$$

In this model, the equation of motion for the density matrix is $\dot{\rho} = \sum_j \sum_{\mu={\rm p,d}}(2\hat{L}_\mu^j\rho\hat{L}_\mu^{j\dagger} - \{\hat{L}_\mu^{j\dagger}\hat{L}_\mu^j,\rho\})$. After sufficient relaxation time, the density matrix attains an essentially classical form $\rho = \sum_{\{n_j\}} p(\{n_j\})|\{n_j\}\rangle\langle\{n_j\}|$, where $p(\{n_j\})$ is the probability of configuration $\{n_j\} \in (0,1)^N$ ($0 \equiv g$, $1 \equiv e$). Classical state-space dimension grows exponentially with N, precluding numeric integration for large systems. However, the steady state of the REs fulfills the detailed balance relation,

$$\frac{p(\{m_j\})}{p(\{n_j\})} = \kappa_l^{\sum_j(m_j - n_j)}. \tag{3}$$

States with the same number of excitations have equal weight and the partition function is given by $Z_N = \sum_{M=0}^N \Omega(M,N)\kappa_l^M$, where $\Omega(M,N)$ is the number of possible arrangements of M excitations on a lattice of N sites. In one dimension, we have the analytic expression $\Omega(M,N) = \binom{N-M+1}{M}$, which enables efficient calculation of all the steady-state probabilities $p(\{n_j\})$.

In Fig. 2 we compare the solutions of the REs model with the t-DMRG results, observing reasonable agreement, especially for the three-level excitation scheme. In all cases, however, the decay of correlations is correctly captured by the REs model. This means that, in the present system, the main features of local quantities are essentially classical.

For a large lattice $N \gg 1$, the partition function converges to $Z_N = \cosh\left((2+N)\frac{\sqrt{\kappa_l}}{2}\right)$, with which the Rydberg excitation probabilities at odd sites j (the boundary being at $j = 1$) are given by $\langle\hat{\sigma}_{rr}^j\rangle = e^{-1/(2\sqrt{\kappa_l})}[1 + e^{-(j-1)/\sqrt{\kappa_l}}]/2$. The decay of spatial oscillations of probabilities $\langle\hat{\sigma}_{rr}^j\rangle$ is characterized by the correlation length,

$$\xi(\kappa_l) = \sqrt{\kappa_l}a. \tag{4}$$

Note that in a translationally invariant system (i.e., in the bulk), the excitation probability is uniform in space, but the onset of crystallization is revealed by the density-density correlations; the corresponding correlation length is again $\xi(\kappa)$. Long-range correlations can only exist if the fluctuations $(\Delta N_r)^2 \equiv \langle\hat{N}_r^2\rangle - \langle\hat{N}_r\rangle^2$ of the total number of Rydberg excitations $\hat{N}_r = \sum_{j=1}^N \hat{\sigma}_{rr}^j$ in the system are suppressed. In the thermodynamic limit $N \to \infty$ of the REs model we obtain

$$\frac{(\Delta N_r)^2}{\langle\hat{N}_r\rangle} = \frac{1}{4\sqrt{\kappa}} + O(\kappa^{-1}). \tag{5}$$

For the two-level excitation scheme of the atoms, $\kappa_2 < 1$, we then have the correlation length shorter than the lattice spacing, $\xi < a$, and large fluctuations. For the same system, Lee *et al.* [27] predicted a phase transition to a period 2 density wave using a mean-field approximation, which is, however, inadequate in one dimension. It is interesting to note that the same correlation properties with $\kappa = 1$ arise in the coherently driven ensemble after thermalization [41]. We note that even in a 2D square lattice the crystalline state cannot be reached with

the two-level atoms; the 2D extension of the REs approach leads to the classical hard-square model, for which a phase transition to the Néel ordered (checkerboard) phase occurs at the critical value of $\kappa_{\rm crit}^{2D} \approx 3.7965$ [42]

For the CPT excitation of three-level atoms, one can tune κ_3 by the Rabi frequencies Ω_{ge} and Ω_{er}, attaining large correlation lengths $\xi > a$. Further increase in ξ is, however, constrained by the validity of the NN interaction approximation. We therefore consider the corrections originating from the hitherto neglected interactions of the atom with the next-nearest neighbors (NNNs). To estimate its maximal effect, let us assume a strong driving, resulting in Rydberg excitations at every other lattice site (half-filling). A nonblocked atom is then likely to be detuned by $\Delta' = \Delta/2^6$ due to the interaction with the NNN Rydberg atoms. Expressing the NN detuning $\Delta = \beta w$ in terms of the excitation line width w and using Eq. (1b), we obtain a modified

$$\kappa_3' = \frac{|\Omega_{ge}|^2}{|\Omega_{er}|^2 + (|\Omega_{ge}|^2 + |\Omega_{er}|^2)\left(\frac{\beta}{64}\right)^2} \leqslant \left(\frac{64}{\beta}\right)^2. \qquad (6)$$

To ensure almost-perfect NN blockade, it is reasonable to take $\beta \approx 10$ and $\sim$1% excitation probability for the blocked atom. In one dimension, the correlation length is therefore limited to $\xi \lesssim 7a$, due to the softness of the vdW potential and the long wings of the Lorentzian excitation profile. In two dimensions, the phase transition to Néel order may occur for three-level atoms, $\kappa_3 > \kappa_{\rm crit}^{2D}$. But since the effects of the NNN interactions (along the diagonals of the square lattice) will be more pronounced, precluding a soluble hard-square REs model, the existence of a phase transition in two dimensions remains an open question. We, finally, note that for the dipole-dipole interaction $\mathcal{V}_{\rm DD}^{ij} \propto \frac{C_3}{d_{ij}^3}$, Eq. (6) reduces to $\kappa_3' \leqslant (8/\beta)^2$, leading to a short correlation length, $\xi < a$.

In summary, we have studied the competition between interactions and fluctuations in an open many-body quantum system represented by a 1D lattice of atoms optically excited to the strongly interacting Rydberg states. Although there is no true phase transition in this system, we have shown that the steady state of an ensemble of three-level atoms can exhibit quasicrystallization of Rydberg excitations with a correlation length extending over many lattice periods. In contrast, for two-level atoms even under strong driving, the correlations are only between the neighboring atoms. We have found that in 2D a transition to Néel order may occur for the three-level driving but not the two-level driving. Using the t-DMRG simulations for several tens of atoms in one dimension, we have shown that for uniform optical driving of all the atoms, the steady state is attained only after very long times. A sequential excitation of neighboring atoms by dynamically "sweeping" the lattice with the driving lasers can result in the same state in a much shorter time. We have derived an effective REs model whose exact steady-state solution, being in good agreement with numeric results, yields explicit analytic expressions for the excitation probabilities and the correlation length.

To conclude, optically driven Rydberg gases are convenient model systems to study many-body correlations and phase transitions in open systems. Apart from their relevance to quantum-optical implementations, open (dissipative) systems have the advantage of possessing robust steady states. An important question is how the transition to an ordered state in a dissipative many-body system is reflected in the (complex) spectrum of the corresponding Liouvillian. For example, in a unitary Rydberg lattice gas, the transition to the crystalline phase is associated with the opening of a gap in the energy spectrum; yet, for open, free-fermion models it was shown in [32] that a transition to long-range order is accompanied by a critical slow-down, i.e., closing the gap in the decay spectrum.

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