Two-dimensional crystals of Rydberg excitations in a resonantly driven lattice gas

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The competition between resonant optical excitation of Rydberg states of atoms and their strong, long-range van der Waals interaction results in spatial ordering of Rydberg excitations in a two-dimensional lattice gas, as observed in a recent experiment of Schauß et al. [Nature 491, 87 (2012)]. Here we use semiclassical Monte Carlo simulations to obtain stationary states for hundreds of atoms in finite-size lattices. We show the formation of regular spatial structures of Rydberg excitations in a system of increasing size, and find highly sub-Poissonian distribution of the number of Rydberg excitations characterized by a large negative value of the Mandel Q parameter which is nearly independent of the system size.

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I. INTRODUCTION

Cold atoms in optical lattices represent a remarkably clean and controllable systems to simulate and study many-body physics [1]. A crucial ingredient for realizing various phases of matter is availability of interactions with different strength and range. The interaction between the ground-state atoms in a lattice is typically short range (on-site) and can be tuned relative to the (inter-site) tunneling energy to realize the Hubbard’s model transition from the superfluid to the Mott insulator phase with precisely one atom per lattice site [2]. Second order tunneling [3] can translate into an effective nearest-neighbor interaction which allows the realization of the Heisenberg spin or extended Hubbard models [4, 5]. Dipolar interactions [6] between atoms with large magnetic moment or between polar molecules have still longer range. But atoms excited to the Rydberg states exhibit unprecedented magnitude and range of dipole-dipole or van der Waals interactions [7–9]. Dressing ground state atoms by the Rydberg state with an off-resonant laser field can thus lead to an effective long-range interaction [10–13].

An alternative approach is to explore resonant coupling of the ground state atoms to the highly excited Rydberg states [14]. The interaction between the atoms in Rydberg states translates into the level shifts of multiple Rydberg excitations which are therefore suppressed in dense atomic ensembles [15–19]. Thus, within a small volume, where the interatomic interaction energies exceed the excitation linewidth of the Rydberg state, a single Rydberg atom blocks the excitation of all the other atoms [20–22]. Larger atomic ensembles can accommodate more Rydberg excitations which effectively repel each other so that no two Rydberg atoms can appear closer than the blockade distance [27–29]. Simultaneously, the total number of excitations within the ensemble exhibits reduced fluctuations [30–32].

Spatial correlations and crystallization of Rydberg excitations have recently invigorated many theoretical investigations [12, 13, 33–49]. A salient experiment of Schauß et al. [24] have demonstrated spatial ordering of Rydberg excitations of resonantly driven atoms in a two-dimensional (2D) optical lattice. In an atomic cloud with the diameter of about two blockade distances, up to five Rydberg excitations were observed. In that experiment, the lifetime of the Rydberg state and the coherence time of the atomic polarization were much longer than the duration (∼ µs) of the optical excitation pulses. Hence, neglecting the relaxations, numerical simulations of the coherent excitation dynamics of many-body system well reproduced the experimental observations [24].

The purpose of the present study is to obtain and characterize the many-body stationary state of a strongly interacting dissipative system subject to continuous (long-time) excitation. To that end, we employ an iterative Monte Carlo sampling algorithm [49] to simulate the steady-state distribution of Rydberg excitations of the van der Waals interacting atoms in a 2D lattice of variable size. As shown in [50], however, the state of the system resulting after only a few µs driving is already close to the steady state. Similarly to [29], we obtain spatially ordered patterns formed by definite numbers of Rydberg excitations, while the probability distribution of the excitation numbers is very narrow (highly sub-Poissonian) characterized by a large negative value of the Mandel Q parameter [30–32]. In a moderately sized atomic cloud, a few mutually repelling Rydberg excitations dwell near the circular boundary of the system, as was also observed in [29]. With increasing the size (diameter d) of the cloud, while keeping the atomic density fixed, the mean number of Rydberg excitations grows (∝ d^{6/3}), but its distribution remains sub-Poissonian with nearly constant Q ≈ −0.84. Concurrently, the Rydberg excitations tend to arrange in regular spatial shell structures, which are, however, progressively smeared for larger number of excitations. The corresponding density-density correlations decay on the length scale of the blockade distance.

The paper is structured as follows: After describing the Monte Carlo procedure in Sec. I, we present the results of numerical simulations in Sec. II, followed by the discussion and conclusions in Sec. IV.
II. THE MONTE CARLO ALGORITHM

Strongly interacting many-body systems are rarely amenable to exact numerical treatment due to the exponential scaling of the corresponding Hilbert space with the system size. For an ensemble of $N$ two-level atoms interacting via a long-range potential, truncating the Hilbert space by limiting the number of Rydberg excitations can greatly reduce the dimension of the problem and allow approximate solutions for tens or hundreds of atoms sharing several excitations [24, 51, 52]. As an alternative technique, semiclassical Monte Carlo simulations [32, 49, 51, 53] can efficiently deal with many more atoms and excitations. The essence of the semiclassical methods is to treat each atom separately in a potential interaction via a long-range potential, truncating the Hilbert space by limiting the number of Rydberg excitations, while neglecting the inter-atomic coherences (entanglement). This approximation is valid when the dephasing rate of the atomic polarization is larger than the Rabi frequency of the driving field [49].

Let us recall the properties of a single two-level atom driven by a laser field with Rabi frequency $\Omega$ and detuning $\delta$ on the transition from the ground state $|g\rangle$ to the excited (Rydberg) state $|r\rangle$. We denote by $\Gamma_r$ the decay rate of the excited state population $\langle \hat{\sigma}_{rr} \rangle$, and by $\Gamma_z$ the relaxation rate of atomic polarization $\langle \hat{\sigma}_{zz} \rangle$, where $\hat{\sigma}_\mu \equiv |\mu\rangle \langle \nu| \hat{\sigma} \langle \nu |\mu\rangle$ is the projection ($\mu = \nu$) or transition ($\mu \neq \nu$) operator of the atom. The steady-state population of the Rydberg state is then a Lorentzian function of detuning $\delta$ [54],

$$\langle \hat{\sigma}_{rr} \rangle = \frac{\Omega^2}{2\Omega^2 + 2\gamma_r^2 (\gamma_r^2 + \delta^2)},$$

with the width $w \simeq 2\Omega \sqrt{\gamma_r^2/\Gamma_r}$, where $\gamma_r \equiv \frac{1}{2} \Gamma_r + 2\Gamma_z$ and we assume strong driving $\Omega^2 > \Gamma_r \gamma_r$ (or weak decay $\Gamma_r < \Omega^2/\gamma_r$). On resonance, $\delta \ll w$, the population saturates to $\langle \hat{\sigma}_{rr} \rangle \rightarrow \frac{1}{2}$.

Atoms in the Rydberg state $|r\rangle$ interact pairwise via the van der Waals (vdW) potential, $\hbar \Delta(z) = \hbar C_6 / z^6$, where $C_6$ is the vdW coefficient and $z$ is the interatomic distance [55]. Hence, given an atom $i$ in state $|r\rangle$, it will induce a level shift $\Delta(z_{ij})$ of another atom $j$, which effectively translates into the detuning $\delta$. When $\Delta(z_{ij}) \gtrsim w$, the transition $|g\rangle \rightarrow |r\rangle$ of atom $j$ is non-resonant and its Rydberg excitation is blocked by atom $i$ [3, 20]. We can therefore define the blockade distance $d_b$ via $\Delta(d_b) = w$ which yields $d_b = \sqrt[3]{C_6 / w}$.

Our aim is to obtain the stationary distribution of Rydberg excitations of $N \gg 1$ atoms in a 2D lattice. We make a semiclassical approximation which discards quantum correlations between the atoms but preserves classical $N$-body correlations. Its validity hinges on the assumption of strong dephasing $\Gamma_z \gtrsim \Omega$ which suppresses intra- and interatomic coherences and disentangles the atoms [49]. Each atom $j$ then behaves as a driven two-level system of Eq. (1) but with the detuning $\delta$ determined by operator $\hat{S}_j = \sum_{i \neq j} \hat{\sigma}_{rr} \Delta(z_{ij})$ which describes the total interaction-induced shift of its level $|r\rangle$ involving the contributions of all the other Rydberg atoms $\hat{\sigma}_{rr}$.

We employ the procedure described in [49] to generate the stationary distribution of Rydberg excitations in an ensemble of $N \lesssim 10^3$ atoms. The algorithm performs an iterative Monte Carlo sampling of $\{\hat{\sigma}_j\}$ for $N$ atoms, in the spirit of the Hartree-Fock method. We first initialize all the atoms in, e.g., the ground state, $\hat{\sigma}_{ij}^g \rightarrow 1 \forall j \in [1, N]$. Then, at every step, for each atom $j$, we set $\hat{\sigma}_{rr}^j \rightarrow 0$ or 1 with the probability determined by its current Rydberg state population $\langle \hat{\sigma}_{rr} \rangle$. In turn, the thus constructed binary configuration of Rydberg excitations $\{\hat{\sigma}_{rr}^j\} \rightarrow \{0, 1, 0, 0, \ldots\}$ determines the level shift $\hat{S}_j$ (equivalent to detuning $\delta$) of atom $j$ when evaluating $\langle \hat{\sigma}_{rr} \rangle$. We continuously iterate this procedure, sifted repeatedly through every atom in the potential generated by all the other atoms. We thus generate a large number ($\sim 10^6$) of configurations $\{\hat{\sigma}_j\}$ from which we obtain the averaged probability distribution $\langle \hat{\sigma}_{rr} \rangle$ of Rydberg excitations and their correlations $\langle \hat{\sigma}_{rr} \hat{\sigma}_{rr} \rangle$, as well as the probabilities $p_k(n)$ of $n$ Rydberg excitations and their spatial patterns.

Note that in the experiment, after preparing the atoms in the ground state $|g\rangle$ and exciting them with a laser, one performs projective measurements $\{\hat{\sigma}_{rr}\}$ of Rydberg excitations [29]. Every experimental sequence then results in a particular configuration $\{\hat{\sigma}_{rr}\} \rightarrow \{1, 0, 0, 1, \ldots\}$, and averaging over many such experimental sequences yields the Rydberg excitation probabilities for individual atoms or the whole ensemble. Hence, the Monte Carlo sampling of the excitation configurations closely imitates the experiment.

III. RESULTS OF NUMERICAL SIMULATIONS

In our simulations, we use the parameters similar to those in the experiment [29], with the exception of a larger relaxation rate $\Gamma_z$ compatible with the semiclassical approximation. We thus assume a 2D lattice of cold $^{87}$Rb atoms whose ground state $|g\rangle \equiv 5S_1/2 \left| F = 2, m_F = -2 \right\rangle$ is coupled to the Rydberg state $|r\rangle \equiv 43S_1/2$ by a two-photon transition with the Rabi frequency $\Omega/2\pi = 100$ kHz. The decay rate of $|r\rangle$ is $\Gamma_r \approx 0.065\Omega \approx 40$ kHz and we take large enough coherence relaxation rate $\Gamma_z \approx \Omega \approx 630$ kHz (in the experiment $\Gamma_z \gtrsim 160$ kHz, stemming from the two-photon laser linewidth $\sim 70$ kHz and residual decay $\sim 90$ kHz of the atomic intermediate state to the ground state [29]). The resulting excitation linewidth of $|r\rangle$ is $w/2\pi \approx 0.8$ MHz. With the vdW coefficient $C_6/2\pi \approx 2.45$ GHz µm$^6$ [53] the corresponding blockade distance is $d_b \approx 3.81$ µm. We performed numerical simulations for various diameters $d$ of the circular boundaries enclosing the 2D lattice of atoms at constant density (fixed lattice spacing $a = 532$ nm [29]).

In Fig. 1 we present the results of simulations for
FIG. 1. (Color online) Rydberg excitations of a resonantly-driven ensemble of $N = (44, 188, 401, 688)$ atoms in a 2D lattice of diameter $d = (1, 2, 3, 4)d_0$, respectively. (a1-a4) Average spatial distribution of Rydberg excitation probabilities $\tilde{\sigma}_r$; each pixel corresponds to a single atom/lattice site. (b1-b4) Probabilities $p_R(n)$ of $n$ Rydberg excitations (open bars) and the mean number of excitations $\langle n_R \rangle$. Also shown is the Poisson distribution $p_{\text{Poisson}}(n)$ for the same $\langle n_R \rangle$ (solid brown circles). (c1-c4) Average spatial distributions of $n$ Rydberg excitations, corresponding to two largest $p_R(n)$ in (b1-b4), after centering and aligning each configuration $\{\tilde{\sigma}_r\}_e$, and the corresponding axial $P(\rho)$ and angular $P(\phi)$ probability distributions $[P(\rho) \text{ and } P(\phi)]$ are binned over the lattice spacing $a$ and $P(\phi)$ are binned over the interval $\pi a/d$.

In Figs. 1(b1-b4), we show the mean number of Rydberg excitations in the cloud $\langle n_R \rangle = \sum_n np_R(n)$ and the probabilities $p_R(n)$ to find $n = 0, 1, 2, \ldots$ excitations. Interestingly, $\langle n_R \rangle$ grows with the size of the system as $\langle n_R \rangle \propto (d/d_0)^{2/3}$ [Fig. 2(a)], rather than $(d/d_0)^2$ as one might expect for a 2D bulk system. This is a finite-size effect, due to the larger concentration of the mutually repelling Rydberg excitations at the cloud boundary whose circumference grows linearly with $(d/d_0)$. [We may conjecture that in a 3D system $\langle n_R \rangle \propto (d/d_0)^{3\alpha}$ with $2 < \alpha < 3$ due to the concentration of Rydberg excitations on the surface of the sphere.]

To quantify the number distribution of Rydberg excitations, we use the Mandel $Q$ parameter \[ Q = \frac{\langle n_R^2 \rangle - \langle n_R \rangle^2}{\langle n_R \rangle} - 1, \] where $\langle n_R^2 \rangle = \sum_n n^2 p_R(n)$. A Poissonian distribution $p_{\text{Poisson}}(n) = \langle n_R \rangle^ne^{-\langle n_R \rangle}/n!$ would lead to $Q = 0$, while $Q < 0$ corresponds to sub-Poissonian distribution, with $Q = -1$ attained for a definite number $n$ of excitations. We find highly sub-Poissonian distribution of $p_R(n)$, Figs. 1(b1-b4), and nearly constant $Q \simeq -0.84 \forall d > d_0$, Fig. 2(b). Note that in the absence of coherence realization, $\Gamma_z = 0$, the (classical) correlations of the Rydberg excitations are considerably smaller, $Q \sim -0.5$ [54] (see Sec. IV).

Next, for each $d$, we select the values of $n$ with the largest probabilities $p_R(n)$ (frequent occurrence) and analyze the average spatial distribution of the correspond-
we observe reduced contrast of the angular distribution for ability of Rydberg excitation at the cloud center, while the blockade distance $d_b$ boundary, and their angular separation is aligned configurations are shown in Figs. 1(c1-c4). Be-

Fig. 2. (Color online) (a) Mean number of Rydberg excitations $\langle n_R \rangle$, and (b) the corresponding $Q$ parameter vs the system size $d/d_b$. In (a) the the black dashed line for $d < d_b$ is $\langle n_R \rangle = 1$, and the dotted line for $d \geq d_b$ is $\langle n_R \rangle = 0.2 + (d/d_b)^{1.66}$.

We also show the corresponding axial $P(\rho)$ and angular $P(\phi)$ probability distributions of the excitations. We then determine the mean angle of the position vectors of all the excited atoms with respect to a reference (x) axis and rotate the configuration about the origin by that angle. The averages over many thus centered and aligned configurations are shown in Figs. 1(c1-c4). Below each 2D density plot (apart from that for $n = 1$) we also show the corresponding axial $P(\rho)$ and angular $P(\phi)$ probability distributions of the excitations. We observe that the Rydberg excitations tend to arrange in regular spatial patterns, i.e., they crystallize. In a cloud of diameter $d \lesssim 2d_b$, the $n$ excitations are pushed to the boundary, and their angular separation is $\delta \phi \sim 2\pi/n$. But already for $d \sim 3d_b$ we encounter a significant probability of Rydberg excitation at the cloud center, while for $d \sim 4d_b$ we clearly observe a double peak structure of $P(\rho)$, with the largest peak close to the boundary of the cloud and the smaller peak corresponding to the inside ring of the corresponding 2D density plot. The distance between the two peaks of $P(\rho)$ is somewhat larger than the blockade distance $d_b$ (see below). Simultaneously, we observe reduced contrast of the angular distribution $P(\phi)$.

In Fig. 3 we plot the normalized spatial correlations

$$g^{(2)}(z) \equiv \frac{\sum_{i \neq j} \delta_{z,z_{ij}} \sigma^x_{iτ} \sigma^x_{jτ}}{\sum_{i \neq j} \delta_{z,z_{ij}} \sigma^x_{iτ} \sigma^x_{jτ}},$$

where $\delta_{z,z_{ij}}$ is the Kronecker delta which selects the pairs of atoms $i,j$ at distance $z_{ij} = z$. Clearly, at short distances $z < d_b$ the Rydberg excitations avoid each other [27,28], while at $z_{max}$ slightly larger than $d_b$ there is a pronounced maximum of $g^{(3)}(z)$, which implies higher probability of finding pairs of Rydberg excitations separated by distance $z_{max}$. Precisely this suppression, or blockade, of Rydberg excitations at shorter distances, and the elevated probability of excitations at distance $z_{max}$, are responsible for the observed quasi-crystallization and inner shell structure of the spatial distribution of $\sigma^x_{iτ}$. At longer distances $z > d_b$, the correlation function $g^{(3)}(z)$ exhibits strongly damped spatial oscillations, signifying the absence of long-range order. Similar results were obtained for a 1D system in [49], where it was found that the correlation length $\xi$ exceeding the oscillation period $\lambda > d_b$ can only be attained at very high atomic densities (thirty or more atoms per blockade distance $d_b$).

IV. DISCUSSION

Above we have analyzed the steady-state distribution of Rydberg excitations in a 2D cloud of atoms subject to continuous resonant driving. We have observed very narrow (sub-Poissonian) distribution of the number of excitations and the formation of regular spatial structures of definite numbers of Rydberg excitations. These quasi-crystals are effected by the boundary of the system and the effective repulsion between the Rydberg excitations which avoid each other at distances smaller than the blockade distance $d_b$. A cloud of diameter $d \lesssim 2d_b$ can then accommodate only a few Rydberg excitations which dwell near the boundary. With increasing the cloud size $d$, the Rydberg quasi-crystals grow, forming spatial shell structures, but their contrast decreases and beyond $d \sim 4d_b$ they are hardly discernible. The absence of the long-range order in Rydberg quasi-crystals, also attested by the rapid decay of the density-density correlation function $g^{(2)}(z)$, is due to the “softness” of the van der Waals potential and the finite excitation linewidth $\nu$ of the Rydberg state. Yet, the Mandel $Q$ parameter, which quantifies the counting statistics of the Rydberg excitations, remains negative and nearly independent of the system size.

Sub-Poissonian statistics of the number of Rydberg excitations and the corresponding negative values of the Mandel $Q$ parameter were observed in several experiment [30,32] which dealt with the transient unitary dynamics of the system. It is interesting to note that for a coherently driven system $Q \sim -0.5$ [50] while incoher-
ent excitation studied here results in \( Q < -0.5 \) and the Rydberg quasi-crystal exhibits sharper contrast than in \[29\]. In other words, dephasing leads to stronger classical (density-density) correlations of Rydberg excitation. This effect is inherited \[29, 50\] from a single superatom—collection of atoms within the blockade distance: Without dephasing, the Rydberg population of a superatom saturates to \((n_R) \to \frac{1}{2} (Q = -0.5)\); with strong dephasing and many atoms, \((n_R)\) approaches 1 \((Q = -1)\), as seen in Fig. 2 for \( d < d_0 \).

In our simulations, we assumed an experimental setup similar to that in \[29\], i.e., square lattice with circular boundary, obtaining, for a moderately sized atomic cloud, qualitatively similar results. Taking, instead, a different lattice geometry would lead to different spatial ordering of Rydberg excitations, but the more fundamental quantities, \( Q \) and \( g^{(2)} \), would behave invariantly for a large cloud of the same atom density.

Let us finally discuss the limitations of our approach and the obtained results. The two key questions are whether the steady state can be attained in a real cold-atom experiment, especially for a large ensemble of atoms \[29\], at least for a moderate number of atoms \( N \lesssim 50 \), and here we briefly summarize the conclusions. In an experiment with typical relaxation rates of the atoms \[29\], the time scale for attaining the true many-body steady state is tens of \( \mu s \)—more for larger \( N \)—which can be prohibitively long due to the loss of atoms and the need of continuous laser irradiation. However, already after several \( \mu s \) of laser driving, the spatial probability distribution of Rydberg excitations ceases to change appreciably \[29\] being close to the steady-state distribution \[50\]. Moreover, stronger coherence relaxation \( \Gamma_z \gtrsim \Omega \) will quickly dephase the atoms rendering the system essentially classical but still strongly correlated \[50\]. The results of the semiclassical Monte Carlo simulations are then trustworthy, as was shown in \[49\].

\[\begin{align*}
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