

## Dynamical Crystallization in the Dipole Blockade of Ultracold Atoms

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We describe a method for controlling many-body states in extended ensembles of Rydberg atoms, forming crystalline structures during laser excitation of a frozen atomic gas. Specifically, we predict the existence of an excitation-number staircase in laser excitation of atomic ensembles into Rydberg states. It is shown that such ordered states can be selectively excited by chirped laser pulses, and, via quantum state transfer from atoms to light, be used to create crystalline photonic states.

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When cold atoms are excited into high-lying Rydberg states, the resulting interactions give rise to energies that exceed the translational energy by many orders of magnitude and strongly modify the excitation dynamics. In very small ensembles confined to a few micrometers, a single Rydberg atom can entirely block any further excitation. This, recently observed, “dipole blockade” [1–4], enables the production of highly entangled collective states with potential applications for fast quantum information processing [2,5,6], and as single-atom as well as single-photon sources [2]. A number of theoretical and experimental studies have addressed the evolution of Rydberg states in large atomic ensembles [7–16]. While strong interactions are critical for many of the predicted and observed phenomena, they also cause a very complex excitation dynamics, thus far preventing controlled quantum state preparation in large ensembles.

In this work, we describe a method that overcomes this obstacle and permits coherent manipulation of strongly correlated, many-body states of large ensembles. The approach is based on a dynamical crystallization of localized collective excitations. The crystalline states can be created with chirped laser pulses, and have a widely tunable lattice spacing. By extending the dipole-blockade mechanism [2] to the regime of multiple Rydberg atoms, such excitation crystals provide a suitable initial state to realize quantum random walks [17]. We show that the crystalline correlations can be mapped onto a propagating light beam and be detected via photon correlation measurements as a pulse train of single photons.

The key idea is illustrated in Fig. 1. For repulsive van der Waals interactions and for a given Rydberg atom number  $n$ , the states with the lowest interaction energy maximize the spacing between Rydberg atoms, which, hence, arrange on regular crystals [Fig. 1(b)]. If initially all atoms are in their ground states, for large negative detunings  $\Delta$  of the laser field, the initial state coincides with the many-body ground state in the rotating frame of reference. As  $\Delta$  is increased to positive values, it effec-

tively lowers the energy levels of excited many-body states [Fig. 1(c)]. They ultimately cross at critical detunings  $\Delta_{n+1}^n$ , marking discrete jumps in the Rydberg atom number from  $n$  to  $n + 1$  [Fig. 1(d)]. As a result, larger and larger Rydberg atom crystals are consecutively populated during evolution of the chirped pulse. In essence, the detuning  $\Delta$  acts as a control parameter that decreases the energy difference between adjacent number states  $|n\rangle$  and  $|n + 1\rangle$ , allowing further Rydberg atoms to “enter” the excitation volume. A similar blockade effect has been observed recently for interacting atoms in an optical lattice [18]. The

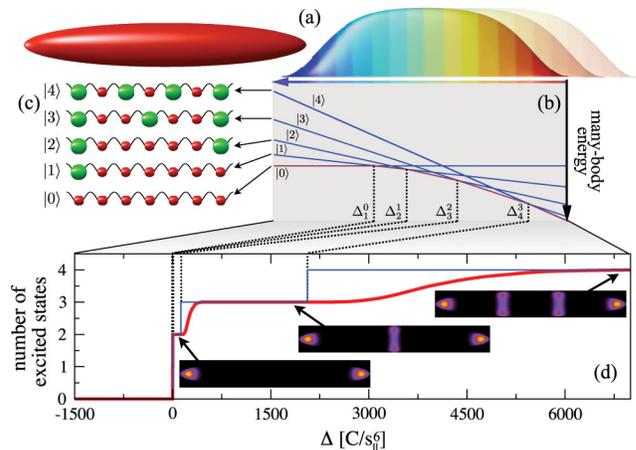


FIG. 1 (color online). (a) A cold atomic gas is illuminated by a chirped laser pulse, whose frequency changes from the blue ( $\Delta < 0$ ) to the red ( $\Delta > 0$ ) side of the driven Rydberg transition. (b) Schematics of the many-body energy spectrum as a function of the laser detuning  $\Delta$  and for a Rabi frequency  $\Omega = 0$ . Shown are the lowest-energy states  $|n\rangle$  for a given Rydberg atom number  $n$ . These states correspond to Rydberg atom (large spheres) lattices sketched in (c). Their energy crossings give rise to an excitation-number staircase shown in (d) for a chain of  $N = 31$  atoms (thin solid line) and a cigar-shaped condensate (thick solid line) with the same length  $s_{||}$  and 500 atoms. The insets show the Rydberg atom densities resulting in the latter at the detunings marked by the arrows.

resulting “dipole-blockade staircase” also bears resemblance to the Coulomb blockade staircase observed in nanoscale solid-state devices [19], where the number of electrons increases as a function of external lead voltage. In contrast to such solid-state devices, the present system permits dynamical manipulation of strongly interacting particles in a highly coherent manner.

The Rydberg gas is described as an ensemble of two-level systems at fixed positions  $\mathbf{r}_i$ , each possessing a ground state  $|g_i\rangle$  and an excited state  $|e_i\rangle$  [20], laser-coupled with a Rabi frequency  $\Omega(t)$ . Within the frozen gas limit, one obtains the following Hamiltonian

$$\hat{H} = -\Delta \sum_i \hat{\sigma}_{ee}^{(i)} + \sum_{i<j} V_{ij} \hat{\sigma}_{ee}^{(i)} \hat{\sigma}_{ee}^{(j)} + \frac{\Omega}{2} \sum_i (\hat{\sigma}_{eg}^{(i)} + \hat{\sigma}_{ge}^{(i)}),$$

where  $\hat{\sigma}_{\alpha\beta}^{(i)} = |\alpha_i\rangle\langle\beta_i|$  ( $\alpha_i, \beta_i = e, g$ ), and  $V_{ij} = C/r_{ij}^6$  describes the repulsive ( $C > 0$ ) van der Waals interaction between Rydberg atom pairs at a distance  $r_{ij}$ .

We first consider the simplest case of atoms in a one-dimensional optical lattice, modeled as a chain of length  $s_{\parallel}$ , that is composed of  $N$  atoms equally separated by  $a = s_{\parallel}/(N - 1)$ . At zero laser intensity, the general structure of the resulting many-body energy spectrum can be easily understood [see Fig. 1(c)]. The  $2^N$  many-body states are composed of  $\binom{N}{n}$  states  $|n, k\rangle$ , where  $k$  labels the spatial configuration of  $n$  excited Rydberg atoms. Their partially degenerate interaction energies  $\epsilon_n^{(k)}$  are shifted by  $-n\Delta$ . Within each  $n$  manifold, the state  $|n, 0\rangle \equiv |n\rangle$  with the lowest energy  $E_n^{(0)} \equiv E_n = \epsilon_n^{(0)} - n\Delta$  maximizes the Rydberg atom spacing at  $z_{ij} = s_{\parallel}/(n - 1) = a_n$ , forming a crystal with lattice spacing  $a_n$ . For small excitation fractions ( $a \ll a_n$ ), the corresponding energies  $E_n \approx -n\Delta + C/s_{\parallel}^6(n - 1)^7$  yield successive level crossings between regular  $n$ - and  $(n + 1)$ -Rydberg atom chains at

$$\Delta_{n+1}^n = \frac{C}{s_{\parallel}^6} [n^7 - (n - 1)^7] \xrightarrow{n \gg 1} 7 \frac{C}{s_{\parallel}^6} n^6. \quad (1)$$

Tuning  $\Delta$  from negative to increasingly positive values, thus, induces stepwise changes in the many-body ground state character from the initial state  $|0\rangle$  to regular chains of  $n$  excited Rydberg atoms ( $|n\rangle$ ). Equation (1) yields a well-defined large- $N$  limit, where the detuning  $\Delta_{\nu} = 7C\rho^6\nu^6 = 7C/a_{n+1}^6$  determines the fraction  $\nu = n/N$  of excited atoms and the Rydberg atom lattice spacing  $a_n = s_{\parallel}/(n - 1)$ .

Transitions between the crystal states are induced by the excitation laser, which couples adjacent  $n$  manifolds, such that the degeneracies at  $\Delta_{n+1}^n$  are lifted and replaced by avoided crossings of separation  $\delta_{n+1}^n$ . Adiabatic preparation of a given crystal state is possible provided that the pulse-evolution is slow compared to  $1/\delta_{n+1}^n$ .

At the first three crossings, adjacent lattice states are directly coupled by the laser field [Figs. 2(a)–2(c)], such

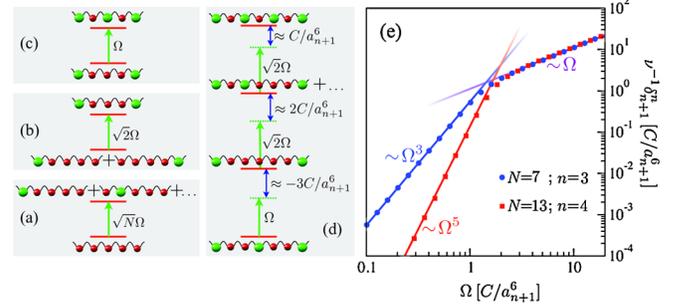


FIG. 2 (color online). (a)–(c) Schematics of single-photon coupling between adjacent Rydberg lattice states  $|n\rangle$  and  $|n + 1\rangle$  for  $n < 3$ . (d) Schematics of the three-photon coupling for  $n = 3$ . The intermediate states are off-resonant by  $\sim C/a_{n+1}^6$ . (e) Resulting energy gap for different  $N$  and  $n$  as a function of  $\Omega$ . The fitted power-laws demonstrate the transition from multi-photon to sequential one-photon coupling, discussed in the text.

that the energy gap  $\delta_{n+1}^n \sim \Omega$ . For  $n \geq 3$ , however, there is no direct laser-coupling between adjacent levels [Fig. 2(d)]. Instead, the gap arises from  $(2n - 3)$ -photon transitions via off-resonant intermediate states detuned by  $\sim C/a_{n+1}^6$ .

If the Rabi frequency  $\omega_n = \Omega(t)/(Ca_{n+1}^{-6}) \ll 1$ , the intermediate states, that couple  $|n\rangle$  and  $|n + 1\rangle$  are far off-resonant and act as virtual levels for resonant multi-photon transitions. For a given  $n$ , these transitions involve  $2n - 2$  intermediate states such that the gap scales as  $\delta_{n+1}^n \sim \omega_n^{2n-3} [n/(N - 1)]^6 C/a_n^6$ . This exponential drop with increasing  $n$ , prevents the preparation of large crystals. However, at larger Rabi frequencies,  $\omega_n > 1$ , power broadening exceeds the intermediate state detunings. In this regime, consecutive crystal states are coupled by sequential one-photon transitions, which results in a linear  $\Omega$ -scaling of  $\delta_{n+1}^n$ . Our numerical calculations confirm this transition from multi- to single-photon coupling at  $\Omega \sim C/a_n^6$  [see Fig. 2(e)]. Beyond this critical Rabi frequency, the gap scales as  $\delta_{n+1}^n \sim \nu\Omega$ , ensuring a finite gap in the large- $n$  limit. Since the lattice spacing  $a_n \approx 1/(\nu\rho)$  is also independent of  $s_{\parallel}$ , very large Rydberg crystals should indeed be producible with finite peak Rabi frequencies  $\Omega > C/a_n^6$ .

We now focus on the dynamics in frozen gases, of randomly spaced atoms. Specifically, we consider a Bose-Einstein condensate, whose initial state  $\prod_i \phi(\mathbf{r}_i) \otimes |g_i\rangle$  is described by a Thomas-Fermi profile  $\phi(\mathbf{r}) \propto \sqrt{1/4 - (x^2 + y^2)/s_{\perp}^2 - z^2/s_{\parallel}^2}$ . Observables are now determined by quantum average over the  $N$ -body spatial wave function and evaluated via Monte Carlo integration. The correlated  $N$ -body wave function is obtained via a Hilbert space truncation approach that exploits the excitation blockade of closely spaced atoms [11].

The simple chain model, developed above, provides a qualitative picture for the excitation dynamics in disor-

dered systems. An important difference is that a disordered gas accommodates Rydberg atom configurations with continuously distributed interaction energies, leading to unavoidable nonadiabatic transitions within a given  $n$  manifold at the end of the laser pulse. The resulting crystals are thus composed of localized collective excitations.

To demonstrate the experimental feasibility of the proposed scheme, we have simulated the dynamics of a cigar-shaped Rubidium condensate excited to  $65s$  Rydberg states with realistic laser pulses [3,10] [Fig. 3(a)]. For  $s_{\parallel} = 15 \mu\text{m}$ , two excitations, localized at the edges of the cloud, are excited. Increasing  $s_{\parallel}$  gives smaller critical detunings  $\Delta_{n+1}^n$  [see Eq. (1)], such that consecutively larger lattices are produced [Figs. 3(b)–3(e)]. Upon further increase of the system size  $s_{\parallel}$ , we find a linearly increasing correlation length (see [21]). The depicted time evolution of the Rydberg atom number populations  $P_n$  demonstrates that—despite the disordered atom positions—the gas is transferred to a state with precisely  $n$  excitations, arranged on perfectly filled Rydberg crystals.

The described mechanism equivalently applies to higher dimensions and different geometries. Exemplary, we show

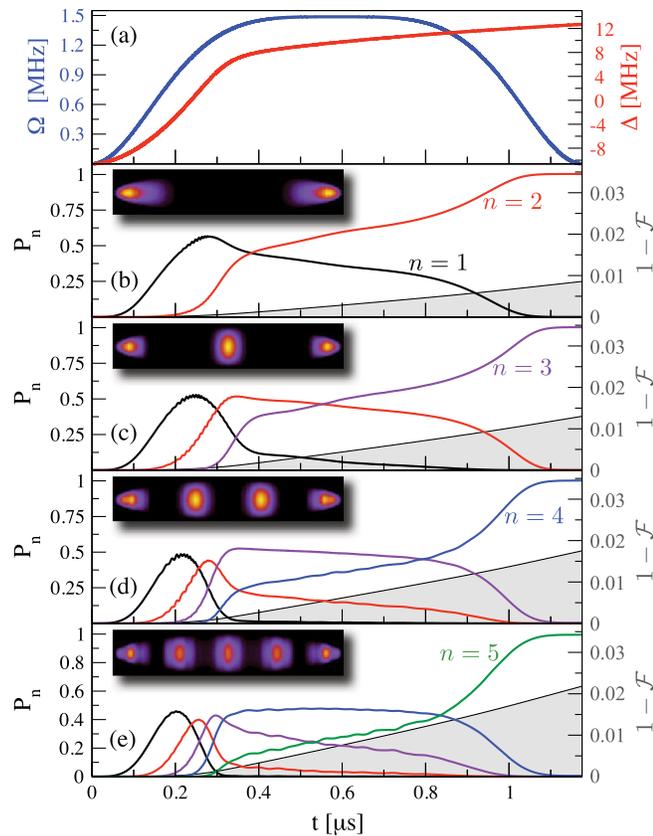


FIG. 3 (color online). (a) Pulse envelope and chirp of the applied laser field, that excites a cigar-shaped Rb condensate to  $65s$  states. (b)–(e) Resulting time dependence of the probabilities  $P_n$  to excite  $n$  atoms in a gas of 300 atoms and with a length of  $s_{\parallel} = 15 \mu\text{m}$  (b),  $22.5 \mu\text{m}$  (c),  $35 \mu\text{m}$  (d),  $45 \mu\text{m}$  (e). The insets show the resulting final Rydberg atom densities.

in Fig. 4 the excitation-number staircase of a pancake-shaped condensate ( $s_{\perp} \gg s_{\parallel}$ ). The depicted densities are obtained via a Monte Carlo average over randomly sampled atom positions, from which we obtain the lowest-energy excited state configuration for a given  $n$ . Analogously to the one-dimensional case, atoms are excited on regular lattice structures, which undergo a structural transition to a single-atom centered hexagon, with increasing  $\Delta$  or  $s_{\perp}$ . Increasing the system size  $s_{\perp}$ , thus, yields radial growth of triangular crystals, with the lattice constant controllable by the detuning  $\Delta$ .

Single-atom decay processes, such as black body radiation induced transitions and spontaneous decay typically occur on very long time scales of several  $100 \mu\text{s}$ . In the limit of small final decay probabilities, we can estimate its importance by following the coherent dynamics and determining the decay probability from the time-dependent number of excited atoms. As shown in Fig. 3, the non-fidelity due to such processes is as small as  $\sim 0.02$ , even for the largest considered Rydberg chain. As this value increases linearly with  $n$ , the preparation of rather large chains of up to 25 atoms has a decay-limited fidelity of  $\mathcal{F} \approx 0.9$ . Cryogenic environments [22] reduce the value of  $1 - \mathcal{F}$  by a factor of  $\sim 3$  for the present parameters, and, thus, permit high fidelity preparation of considerably larger crystals. Effects of interaction-induced atomic motion are estimated the classical displacement  $\Delta r \approx Cr^{-7}M^{-1}\tau^2$  of an atom pair, initially separated by  $r$ , where  $M$  is the mass of the atoms. For the parameters of Fig. 3 ( $r > 10 \mu\text{m}$ ) and  $\tau = 4.0 \mu\text{s}$ , this gives a change in atom distances by less than a fraction of  $10^{-3}$ , leaving the excitation dynamics practically unaffected.

A number of techniques can be used to experimentally probe the Rydberg crystals. Absorption imaging of ground state atoms in optical lattices [23] would permit detection of excited atoms as defect spots at the corresponding lattice sites. Field ionization of Rydberg atoms combined with position sensitive MCP detectors and ion optics magnifi-

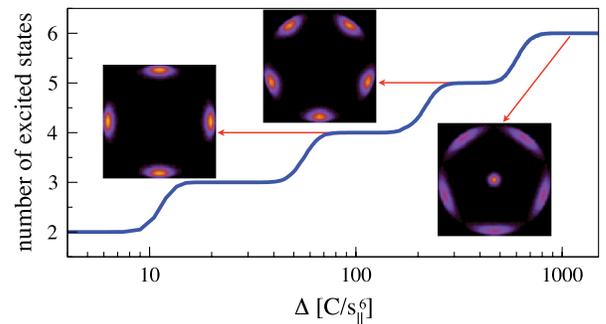


FIG. 4 (color online). Dipole-blockade staircase in a 2D condensate, along with Rydberg atom densities of the corresponding many-body ground states. To visualize the angular correlations, each individual configuration is rotated such that one excitation is centered at the bottom of the image and the horizontal component of the Rydberg atoms' center of mass vanishes.

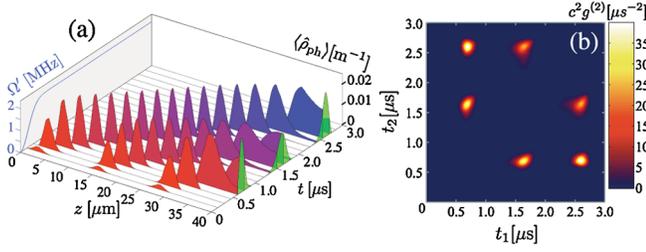


FIG. 5 (color online). Light mapping of a three-Rydberg atom crystal with  $s_{\parallel} = 30 \mu\text{m}$  and  $s_{\perp} = 4 \mu\text{m}$ , embedded in a condensate with a longitudinal and transverse size of 50 and  $10 \mu\text{m}$ , respectively. (a) Pulse propagation dynamics, as described by the expectation value of the photon density operator  $\hat{\rho}_{\text{ph}} \propto \hat{\mathbf{E}}^{\dagger} \hat{\mathbf{E}}$ . The outer-left blue curve shows the time evolution of the control Rabi frequency  $\Omega'$ , while the right green area gives the resulting photon signal, detectable outside the atom cloud. (b) Corresponding two-photon correlation function  $g^{(2)} = \langle \hat{\rho}_{\text{ph}}(t_1) \hat{\rho}_{\text{ph}}(t_2) \rangle$ . Multiplication by the squared speed of light,  $c$ , yields a temporal density and ensures the correct normalization  $c^2 \int g^{(2)} dt_1 dt_2 = n(n-1)$ .

ation [24] should permit to image the predicted  $\mu\text{m}$ -scale structures.

Alternatively, the crystalline excitations can be mapped onto correlated states of light and probed via photon correlation measurements. Here, a strong  $\pi$  pulse is used to coherently transfer the Rydberg lattice to a crystal state of atoms in another hyperfine ground state  $|g'\rangle$ . A second laser with Rabi frequency  $\Omega'(t)$  drives the transition between  $|g'\rangle$  and a low-lying excited state  $|e'\rangle$ , which is coupled to  $|g\rangle$  by a quantum field  $\hat{\mathbf{E}}(\mathbf{r}, t)$ . Analogous to light storage schemes in an EIT medium [25], the initially prepared quantum state of atomic excitations can be transferred to the radiation field  $\hat{\mathbf{E}}$  by adiabatically turning on the control field  $\Omega'$ . Figure 5 shows the resulting transfer dynamics for a three-atom chain, embedded in a larger condensate [26]. The collective nature of the excitations not only provides enhanced coupling to light, but also leads to highly directed photon emission along the total wave vector of the applied laser fields. The Rydberg chain is mapped onto a clear triple-peak structure, with well-resolvable delay times of  $\sim 1 \mu\text{s}$ . The corresponding second order correlations, shown in Fig. 5(b), clearly reveal the single-photon nature of each pulse, as characterized by the absence of equal-time photon signals. Since there is a one-to-one mapping between the photon density  $\hat{\rho}_{\text{ph}}$  and the initial Rydberg state density operator [25], the depicted time correlations directly reflect the strongly correlated nature of the prepared many-body state. In this way, correlation measurements, provide a probe for (im)perfections of the underlying Rydberg lattice.

In summary, we have shown that frequency-chirped excitation of cold gases permits to form ordered structures of Rydberg atoms out of otherwise disordered ensembles. Such crystals naturally realize a network of collective excitations without the need of extended microtrap arrays. Chirped excitation in optical lattices would provide an experimental scenario to study magnetism of long-range interacting spins and explore their quench dynamics through quantum phase transitions. The described quantum optical imaging technique could be used as a real-space probe of spatial correlations between the effective spins, and, at the same time, provide a novel source of non-classical light.

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