



Wigner Crystallization of Single Photons in Cold Rydberg Ensembles

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The coupling of weak light fields to Rydberg states of atoms under conditions of electromagnetically induced transparency leads to the formation of Rydberg polaritons which are quasiparticles with tunable effective mass and nonlocal interactions. Confined to one spatial dimension their low energy physics is that of a moving-frame Luttinger liquid which, due to the nonlocal character of the repulsive interaction, can form a Wigner crystal of individual photons. We calculate the Luttinger K parameter using density-matrix renormalization group simulations and find that under typical slow-light conditions kinetic energy contributions are too strong for crystal formation. However, adiabatically increasing the polariton mass by turning a light pulse into stationary spin excitations allows us to generate true crystalline order over a finite length. The dynamics of this process and asymptotic correlations are analyzed in terms of a time-dependent Luttinger theory.

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The extraordinary properties of Rydberg atoms [1], such as large dipole-dipole interactions and long lifetimes, are currently attracting much attention. The interest ranges from quantum information [2–4] to many-body phenomena [5–15]. So far only few works considered the effect of interactions onto the light fields [16–21]. In recent experiments [16] it was shown that under conditions of electromagnetically induced transparency (EIT) the Rydberg interaction leads to a nonlocal, and strongly nonlinear behavior of the probe field [22,23]. This gives rise to, e.g., the formation of a small avoided volume which contains at most one excitation [20,21]. In the present Letter we want to explore the many-body properties on larger length scales. One of the simplest but most dramatic effects resulting from a nonlocal repulsive interaction is the formation of a Wigner crystal, predicted for electrons in the early days of quantum mechanics [24]. We will show that a similar phenomenon can be observed in a dilute one-dimensional gas of photons coupled to Rydberg atoms. The resulting quantum state is highly nonclassical and cannot be created in conventional Kerr-type point-interacting systems [25,26]. This has potential applications in photon based quantum communication and information. E.g., the regularity of the photon train can provide high bit rates in quantum repeater protocols and multiplexing.

Under conditions of EIT and small excitation densities, the coupling between photons and Rydberg atoms leads to the formation of light-matter quasiparticles, the so-called dark-state polaritons (DSP) [27,28]. The DSP follow a Schrödinger-equation with an externally tunable mass and additional strong repulsive and nonlocal interactions. We analyze the formation of a quasicrystalline state of polaritons in one dimension using DMRG simulations and time-dependent Luttinger-liquid (LL) theory. We show that under typical time-independent slow-light conditions the moving-frame ground-state displays

density-wave correlations that decay fast in propagation direction due to the small polariton mass. However, using the external control and making the DSP more massive, i.e., converting them into stationary spin excitations, increases the effect of interactions. Consequently, decelerating a light pulse to a full stop inside a gas of Rydberg atoms [29,30] can lead to perfect crystalline order over the length of the medium. We note that Wigner crystallization of solitons, representing coherent light pulses, has recently been proposed in [31]. In contrast, our approach leads to the crystallization of single excitations, forming a regular train of single photon states upon readout.

To be specific, we consider an ensemble of N atoms with a three-level linkage pattern [cf. Fig. 1(b)], composed of a ground state $|g\rangle$, intermediate state $|e\rangle$, and metastable Rydberg-state $|r\rangle$. The transition $|g\rangle - |e\rangle$ is driven by a quantized probe field $\hat{E} = \sqrt{(\hbar\omega_p/2\epsilon_0)}\hat{\mathcal{E}}(\mathbf{r}, t)e^{-i(\omega_p t - \mathbf{q}_p \mathbf{r})} + \text{H.a.}$, with carrier frequency ω_p and wave vector \mathbf{q}_p . $\hat{\mathcal{E}}(\hat{\mathcal{E}}^\dagger)$ are normalized field amplitudes corresponding to annihilation (creation) of a photon and are slowly varying in space

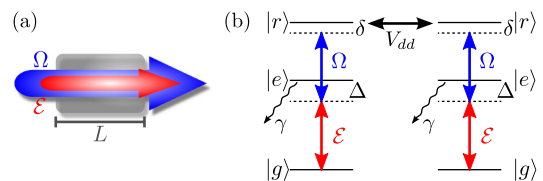


FIG. 1 (color online). (a) Schematic setup for the creation of dark-state polaritons in a medium on length L . (b) Effective atomic linkage pattern for EIT in Rydberg gases. The weak quantized field $\hat{\mathcal{E}}$ is off-resonantly driving the $|g\rangle - |e\rangle$ transition with a one-photon detuning Δ , whereas the strong control field Ω is driving the $|e\rangle - |r\rangle$ transition with a final two-photon detuning δ .

and time. The transition $|e\rangle - |r\rangle$ is coupled via an external control field with Rabi frequency Ω , carrier frequency ω_c , and wave vector \mathbf{q}_c . We chose the z axis as the common propagation direction of the fields and define the one- and two-photon detunings as $\Delta = \omega_e - \omega_g - \omega_p$, $\delta = \omega_r - \omega_g - \omega_c - \omega_p$, where $\omega_{g,e,r}$ are the energies of the atomic states ($\hbar = 1$).

In the absence of Rydberg interactions the Hamiltonian can be diagonalized using adiabatic eigensolutions, the dark- and bright-state polaritons (BSP), which fulfill approximate bosonic commutation relations [27,28]. Following [32] we define the DSPs as $\hat{\Psi} = \cos\theta\hat{E} - \sin\theta\hat{\Sigma}_{\text{gr}}$, and BSPs as $\hat{\Phi} = \sin\theta\hat{E} + \cos\theta\hat{\Sigma}_{\text{gr}}$ where $\tan^2\theta = g^2n/\Omega^2$. Here $\hat{\Sigma}_{\mu\nu}$ are continuous atomic spin flip operators, n is the atomic density and $g = \wp\sqrt{\omega_p/2\hbar\epsilon_0}$, with the $|g\rangle - |e\rangle$ dipole moment \wp . The DSP propagates lossless with group velocity $v_g = c\cos^2\theta$, while the BSP has a velocity $c\sin^2\theta$ and is subject to losses with rate $(g^2n + \Omega^2)/\Gamma$, where $\Gamma = \gamma + i\Delta$, with 2γ being the spontaneous decay rate of $|e\rangle$. Near single-photon resonance $|\Delta| \lesssim \gamma$, and for an optically thick medium $L \gg L_{\text{abs}}$, where $L_{\text{abs}} = c\gamma/g^2n$ is the resonant absorption length in the absence of EIT and L the medium length, an input bright polariton will quickly be damped out. In the following we will consider $|\Delta| \gg \gamma$, where absorption is irrelevant. However, for $\cos^2\theta \ll \sin^2\theta \approx 1$ and light pulses of finite length, an input bright polariton can still be disregarded as it will quickly escape the medium ($c \gg v_g$). This allows us to eliminate the BSP and after a short transient the free dynamics is governed by [33]

$$\hat{H}_0 = \int d^3\mathbf{r} \hat{\Psi}^\dagger(\mathbf{r}) \left[\frac{\hat{p}_z^2}{2m_{\parallel}} + \frac{\hat{\mathbf{p}}_{\perp}^2}{2m_{\perp}} - v_g \hat{p}_z + \delta(\mathbf{r}) \right] \hat{\Psi}(\mathbf{r}), \quad (1)$$

where $\hat{p}_z = -i\partial_z$, $\hat{\mathbf{p}}_{\perp} = -i\nabla_{\perp}$, and $\sin^2\theta \approx 1$ was used. This corresponds to an effective Schrödinger equation for particles with tensorial mass and additional drift terms, moving in an external potential $\delta(\mathbf{r})$. The drift is determined by the EIT group velocity v_g , and the masses are $m_{\parallel}^{-1} = v_g L_{\text{abs}}(\Delta/\gamma)$ and $m_{\perp}^{-1} = v_g/2q_p$ [32,34]. The above model is valid as long as the BSP amplitude is negligible and [33]

$$|\delta| \ll \frac{g^2n}{|\Delta|}, \quad \frac{L_{\text{abs}}}{L_{\text{DSP}}} \leq \frac{\gamma}{|\Delta|}. \quad (2)$$

The first condition describes the regime of perturbative coupling between DSP and BSP [35]. The second denotes the region of slow-light dispersion [28,34], where L_{DSP} is a characteristic length scale of the DSP. Let us now take into account interactions between the atoms in their Rydberg state $|r\rangle$, with van der Waals interaction potential $V(\mathbf{r}) = C_6/|\mathbf{r}|^6$. In the continuum limit and transforming to polaritons, we find to lowest order in $\cos\theta$

$$\hat{H}_{\text{int}} = \frac{C_6}{2} \int d^3\mathbf{r} d^3\mathbf{r}' \frac{\hat{\Psi}^\dagger(\mathbf{r}) \hat{\Psi}^\dagger(\mathbf{r}') \hat{\Psi}(\mathbf{r}') \hat{\Psi}(\mathbf{r})}{a^6 + |\mathbf{r} - \mathbf{r}'|^6}, \quad (3)$$

where we introduced a cutoff a to account for a possible regularization at short distances [20]. However, as we will

show later, for strong interactions or heavy particles the results become independent of the cutoff and we are allowed to set $a = 0$. The effect of the interaction is equivalent to a two-photon detuning. Consequently, the interaction shift has to be smaller than $g^2n/|\Delta|$ which can be translated into a minimal distance $a_c = (C_6|\Delta|/g^2n)^{1/6}$, which the DSPs have to keep to ensure the validity of the model. As shown in [20] for the case of a resonant interaction (i.e., $\Delta = 0$) and large optical depth, an incoming coherent light pulse will quickly develop strong antibunching with a minimum separation length along the propagation direction corresponding to the EIT blockade radius $a_b = (C_6\gamma/\Omega^2)^{1/6} \gg a_c$. A similar effect happens for $\Delta \neq 0$ due to the fast escape of the BSP. Since under slow-light conditions, $\cos\theta \ll 1$, the initial preparation produces DSPs with a mutual distance larger than the critical value a_c and a vacuum of BSPs, the system is well described by $\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}$.

To address the question whether the interaction leads to Wigner-crystallization of polaritons we restrict ourselves to one dimension (1D). This can be achieved, e.g., by using elongated cigar-shaped atomic ensembles with transverse extent smaller than the blockade radius [21], or atoms in hollow-core fibers [36,37] or trapped in the evanescent field of ultrathin optical fibers [38,39]. The low-energy physics can be described in terms of a Luttinger liquid [40]. The LL model allows for an exact treatment also in the case of bosons [41] with $1/|x|^\alpha$ interactions, as long as $\alpha > 1$. Transforming to a frame comoving with the EIT group velocity removes the drift term, $\sim v_g \partial_z$, in Eq. (1). Assuming a fixed excitation density ρ_0 and $|\Delta| \gg \gamma$, we follow the standard LL approach [40] to construct an effective low-energy Hamiltonian

$$H_{\text{LL}} = \frac{1}{2\pi} \int dx \left\{ uK(\pi\hat{\Pi})^2 + \frac{u}{K}(\nabla\hat{\phi})^2 \right\}. \quad (4)$$

$\hat{\Pi}$ and $\hat{\phi}$ are conjugate fields with $[\hat{\phi}(x), \hat{\Pi}(y)] = i\delta(x-y)$. u and K are the sound velocity and the Luttinger parameter, respectively. The K parameter governs the asymptotic behavior of the charge-density-wave correlations (CDW) in the ground state. For example, the oscillatory part of the density correlations is given by $\langle \hat{\rho}(z)\hat{\rho}(0) \rangle_{\text{osc}} \sim \rho_0^2 \cos(2\pi\rho_0 z) z^{-2K}$, with $\hat{\rho}(z) = \hat{\Psi}^\dagger(z)\hat{\Psi}(z)$. As first-order correlations decay as $\langle \hat{\Psi}^\dagger(z)\hat{\Psi}(0) \rangle \sim z^{-1/(2K)}$ the point $K = 1/2$ marks the crossover from a regime where superfluid order dominates ($K > 1/2$) to a regime with predominant CDW correlations of period $1/\rho_0$ ($K < 1/2$). We note that technically spoken, the interaction (3) is of short-range character and we will not find any slower-than-power law correlations as for, e.g., unscreened Coulomb interactions [42].

We like to point out that one can create true crystalline order by adding a weak periodic lattice potential $\delta(x) = \delta_0 \sin(2\pi x/d)$, which leads to a sine-Gordon Hamiltonian [40] for commensurate fillings $\rho_0 = 1/(sd)$, $s \in \mathbb{N}$. This model exhibits a quantum phase transition to a gapped ordered phase for arbitrarily small but finite δ_0 , if

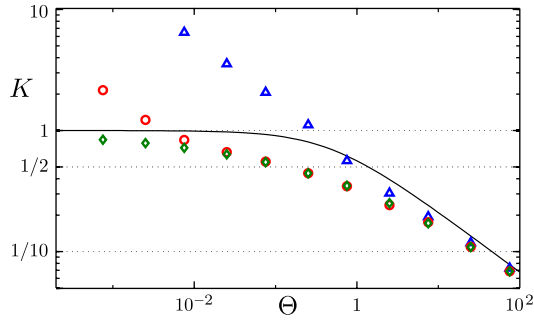


FIG. 2 (color online). Luttinger parameter K as a function of interaction strength. The continuous line shows the analytical approximation [46] for C_6/r^6 . Green diamonds show results from DMRG calculations with periodic boundary conditions (BC) for the unscreened potential. Red circles are for $\rho_0 a = 1/5$, blue triangles for $\rho_0 a = 1/2$. The numerical parameters were $\Delta x = 1/10\rho_0$ with $L = 10/\rho_0$ and $d = 32$ for periodic BC. Quantum Monte Carlo results for C_3/r^3 can be found in [41].

$K < K_s = 2/s^2$ [40,43]. To avoid the necessity of a comoving lattice potential one then should consider stationary-light polaritons [34,44,45].

Although no exact expression for K exists, an approximate closed formula was given in [46]:

$$K = \frac{1}{\sqrt{1+2\Theta}}, \quad \Theta = \frac{\pi^3}{180} \rho_0^4 m C_6, \quad (5)$$

where Θ is a measure for the ratio between interaction and kinetic energy.

To check this expression we determined K numerically using DMRG [47] and made use of the fact that $K/u = \pi\rho_0^2\chi$ is determined by the compressibility $\chi^{-1} = \rho_0^2(\partial\mu/\partial\rho_0) = \rho_0^2 L(\partial^2 E/\partial N^2)$ [40]. Furthermore, $uK = \pi\rho_0/m$, which is true for any Galilean-invariant model [48]. We have validated the numerical procedure for the case of the integrable Lieb-Liniger model, where χ can be calculated exactly as a function of interaction. Using a proper discretization of the model [49] leads to the results for K shown in Fig. 2 for regularized and diverging van der Waals interactions. As expected for large Θ (i.e., small K) the K parameter becomes independent of the cutoff a . Moreover, for $a = 0$, Eq. (5) gives the right order of magnitude over the whole range and we will use this expression in the following. As we are interested in the regime of small K we conclude that the actual form of the potential at short distances is irrelevant and only the asymptotic form is important. Hence we can set $a = 0$, as long as $\rho_0 a \ll 1$.

Let us first discuss the time-independent case $\Omega(t) = \Omega = \text{const}$. We concentrate on the ground state where CDW correlations should be most pronounced. In Fig. 3 we have plotted the normalized two-particle correlation $g^{(2)}(z)$ in the ground state of $\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}$ obtained by DMRG corresponding to different values of K . The large- z behavior follows the LL expression and one recognizes

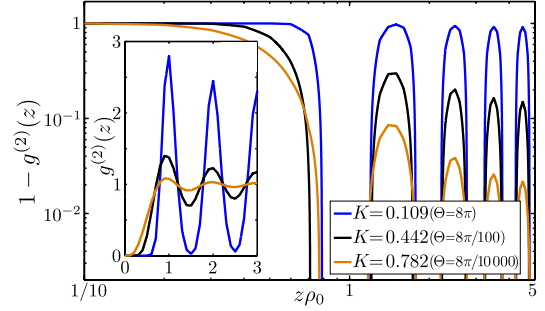


FIG. 3 (color online). Normalized two-particle correlation. Main panel: $1 - g^{(2)}(z)$, in double logarithmic scale. Inset: $g^{(2)}(z)$ in linear scale. Full lines show numerical results for interaction strength increasing from orange over black to blue. The particles are subject to periodic boundary conditions. Note that at $\rho_0 z \gtrsim 5$ finite size effects become noticeable.

well pronounced oscillations for $K \leq 1/2$. For small distances the plots show an extended spatial region over which $g^{(2)}(z)$ vanishes, showing that the CDW is a regular array of single-photon Fock states. The correlations around $z = 0$ become more suppressed than in the case of free fermions, which is the strongest possible for point interactions [25,50].

We can use Eq. (5) to estimate the critical interaction strength required to enter the CDW dominated regime, i.e., $K \leq 1/2$, giving $\Theta_{\text{crit}} = 3/2$. Θ is proportional to the effective mass of the polaritons $m \sim v_g^{-1} \sim g^2 n/\Omega^2$ which is different along longitudinal (m_{\parallel}) and transverse directions (m_{\perp}), and can be tuned via the control field Ω . For $m = m_{\parallel}$ we find

$$\Theta = \frac{\pi^3}{180} \left(\frac{\gamma}{|\Delta|} \right)^2 (\rho_0 L_{\text{abs}})^4 \frac{c}{v_g} \text{OD}_c^6, \quad (6)$$

where $\text{OD}_c = a_c/L_{\text{abs}}$ is the optical depth per critical radius. In the crystalline state the characteristic length scale is $L_{\text{DSP}} \sim 1/\rho_0$ and thus condition (2) translates into $\rho_0 L_{\text{abs}} \leq \gamma/|\Delta|$. Using, e.g., $\rho_0 L_{\text{abs}} = \gamma/|\Delta| = 1/100$ and $v_g/c = 10^{-5}$, we find that the optical depth per critical radius at $\Theta = \Theta_{\text{crit}}$ has to be $\text{OD}_c^{\parallel} \gtrsim 20$. As the mass along the transverse direction is larger, the conditions are more relaxed here and a similar analysis yields $\text{OD}_c^{\perp} \gtrsim 5$. Nevertheless, a crystalline structure will be challenging to prepare along both directions as for typical parameters $\text{OD}_c \lesssim 1$. It should be noted, though, that in a finite-size system the CDW might still be observable, as its amplitude can be quite large [51].

A closer look at Eq. (6) suggests a possibility to overcome this challenge using standard light storage techniques [27,52,53]. Let us consider an initial polariton pulse close to the moving-frame ground state but now with time-dependent control fields. In the absence of interactions, decelerating the DSPs by reducing v_g in time preserves their spatial structure and density ρ_0 [28]. Simultaneously,

their effective mass is increased, which suggests an increasing Θ and hence a decreasing K according to Eq. (5) for interacting DSPs. When the pulse is brought to a complete stop, $K(t)$ approaches zero potentially leading to true long-range order. If v_g is switched off instantaneously, the initial spatial correlations will be frozen. Thus the switching has to be done smoothly on a time scale τ long enough for correlations to propagate through the system. The latter process is determined by the speed of sound $u(t) = \pi\rho_0/[m(t)K(t)]$. For small K we find the scaling $K \sim 1/\sqrt{m} \sim \sqrt{v_g}$; i.e., the sound velocity decreases only with the square root of the group velocity, $u(t) \sim 1/\sqrt{m(t)} \sim \sqrt{v_g}$, allowing the correlations to propagate through the system before being frozen.

In order to describe the adiabatic switch off we consider the LL Hamiltonian (4) with time-dependent parameters $K(t)$ and $u(t)$ [54]. Choosing a special, but generic time dependence $\Omega(t) = g\sqrt{n}/\sqrt{f(t)c/v_g(0) - 1}$, where $f(t) = e^{x(t)} \sinh[x(t)] e^{-\text{arccosh}(C)/\sqrt{C^2 - 1}}$, with $x(t) = \text{arccosh}(t/\tau + C)$ and $C = (K_0^2 + 1)/(2K_0)$, $K_0 = K(t=0)$ and switch-off time τ , the time-dependent LL model can be solved exactly (see [33] for details). Because of the finite speed of sound, the final correlations exhibit a ‘‘crossover’’ as a function of distance from the power-law behavior with adiabatic exponent $K(t)$ to one with the initial exponent K_0 at a length scale $l_0 = (\pi^4/90)C_6\rho_0^5\tau \approx \text{OD}_c(\rho_0 a_c)^5 c\tau|\Delta|/\gamma$, as can be seen in Fig. 4. Obviously, the switch-off time τ should be maximal. On the other hand, τ has to be sufficiently small to bring the pulse to a complete stop within the medium, such that $L \leq \int_0^\infty dt v_g(t)$. Using the above protocol, we find

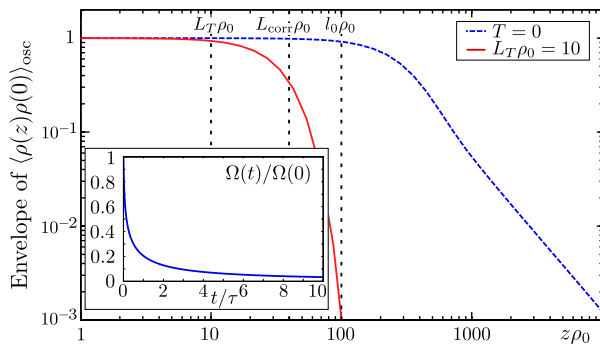


FIG. 4 (color online). Main panel: space dependent amplitude of the oscillatory part of the correlation function in the long-time limit, $K_0 = 0.8$, $K(t) = 5 \times 10^{-5}$. The dashed blue line shows the spatial decay of density-density correlations for zero temperature, which shows a crossover from adiabatic to diabatic algebraic decay at $l_0\rho_0 = 100$ (indicated by the rightmost vertical line). The solid red line shows the modified decay for initial temperature corresponding to a thermal length $L_T\rho_0 = 10$ (leftmost vertical line), which shows a crossover to exponential decay at length scale $L_{\text{corr}}\rho_0 \approx 40$. Inset: $\Omega(t)/\Omega(0)$ for $K_0 = 0.8$ and $v_g(0)/c = 10^{-5}$.

$$\frac{l_0}{L} = \frac{2\pi}{K_0} \rho_0 L_{\text{abs}} \frac{|\Delta|}{\gamma}. \quad (7)$$

It is interesting to note that this expression does not depend on the interaction strength. This is a consequence of the chosen protocol where the temporal change of $v_g(t)$ depends on the interaction strength. Assuming that K_0 is close to unity and that $\rho_0 L_{\text{abs}} \leq \gamma/|\Delta|$, l_0/L can approach unity showing that a crystalline order over the whole medium is possible.

Changing the control-field in time leads to additional couplings between the DSP and BSP [28]. The decay rate due to this coupling is given by $\gamma_\theta = \gamma\dot{\theta}^2/g^2n$. Requiring $\int_0^\tau dt \gamma_\theta(t) \ll 1$ and using the above protocol we find $c\tau/L_{\text{abs}} \gg 4K_0^2/(K_0^2 - 1)^2$. For $K_0 \approx 0.99$ and $L_{\text{abs}} \approx 5 \mu\text{m}$ we have $\tau \gg 0.16$ ns, which is certainly feasible.

So far we have assumed that the initial state for the light storage is the moving-frame ground state of the LL Hamiltonian. Let us now discuss the effects of initial excitations. As the system is nonintegrable it is reasonable to assume that the state of the DSPs after the initial preparation is thermal (we set $k_B = 1$). In a thermal state all correlations decay exponentially with a correlation length $\sim L_T/K = \pi\rho_0/(mTK^2)$ [55]. Since for a noninteracting gas and adiabatic mass changes $T(t) \sim 1/m(t)$ holds, one naïvely expects that the correlation length increases $\sim 1/K^2(t)$. Evaluating the bosonic correlation functions with a thermal distribution we find a slightly different result (cf. Fig. 4). For intermediate length scales, correlations decay as $\exp(-|z|^2/L_{\text{corr}}^2)$ as long as $|z| \leq L_{\text{corr}} = 2\sqrt{l_0 L_T^0}/\pi K_0 \times (\ln[K_0/K(t)])^{1/4}$ which crosses over to an exponential decay for larger distances [33]. Here L_T^0 is the initial thermal correlation length.

To estimate the initial temperature of the DSP we observe that any polariton component with frequency larger than the off-resonant EIT linewidth $\Omega^2/|\Delta|$ will escape [33]. Thus a reasonable estimate for an upper temperature limit is $T \lesssim (1/2)(\Omega^2/|\Delta|)$, and we obtain the final correlation length at finite temperature as

$$\frac{\sqrt{l_0 L_T^0}}{L} = \left(\rho_0 L_{\text{abs}} \frac{|\Delta|}{\gamma} \right) \sqrt{\frac{2\pi}{\text{OD}} \frac{|\Delta|}{\gamma}}. \quad (8)$$

Although the first term on the right side is less than unity, the whole expression can still approach unity and thus even for an initial polariton wave packet with finite energy an almost perfect Wigner crystal can be created.

An important question is how to observe the crystalline structure of the stored DSPs. This is achieved by turning the stationary excitations back into propagating photons, following standard light retrieval techniques [27]. However, this must be done by instantaneously switching on the control field as an adiabatic switch-on would just invert the generation process. The resulting regular train of single-photon pulses that leaves the medium can then be detected using standard correlation measurement techniques [21].

In summary, we showed that the combination of EIT with interacting Rydberg gases leads to strongly interacting light-matter particles, termed Rydberg polaritons. We discussed the experimental requirements needed to obtain a quasi-long-range-ordered ground state corresponding to a moving-frame Wigner crystal of Rydberg excitations in one dimension by mapping the problem to a Luttinger liquid. Numerical and analytic results showed that under slow-light conditions the kinetic energy contributions in the longitudinal direction are too large to enter the density-wave dominated regime. Using a time-dependent Luttinger liquid approach we showed, however, that decelerating a light pulse in a gas of Rydberg atoms to a full stop over a sufficiently long deceleration time can create true crystalline order over a substantial fraction of the medium. Turning the Wigner crystal of spin excitations back into electromagnetic fields by a sudden switch-on of the drive field produces a train of photons with long-range crystalline order.

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