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Theory of interacting cavity Rydberg polaritons

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Abstract

Photonic materials are an emerging platform to explore quantum matter (Carusotto and Ciuti 2013 Rev. Mod. Phys. 85 299; Sommer et al 2015 arXiv:1506.00341) and quantum dynamics (Peyronel et al 2012 Nature 488 57). The development of Rydberg electromagnetically induced transparency (Weatherill et al 2008 J. Phys. B: At. Mol. Opt. Phys. 41 201002; Petrosyan et al 2011 Phys. Rev. Lett. 107 213601) provided a clear route to strong interactions between individual optical photons. In conjunction with carefully designed optical resonators, it is now possible to achieve extraordinary control of the properties of individual photons, introducing tunable gauge fields (Schine et al 2016 Nature 534 671) whilst imbuing the photons with mass and embedding them on curved spatial manifolds (Sommel et al 2016 New J. Phys. 18 035008). Building on work formalizing Rydberg mediated interactions between propagating photons (Gorshkov et al 2011 Phys. Rev. Lett. 107 136302; Gullans et al 2016 Phys. Rev. Lett. 117 113601), we develop a theory of interacting Rydberg polaritons in multimode optical resonators, where the strong interactions are married with tunable single particle properties to build and probe exotic matter. In the presence of strong coupling between the resonator field and a Rydberg-dressed atomic ensemble, a quasiparticle called the ‘cavity Rydberg polariton’ emerges. We investigate its properties, finding that it inherits both the fast dynamics of its photonic constituents and the strong interactions of its atomic constituents. We develop tools to properly renormalize the interactions when polaritons approach each other, and investigate the impact of atomic motion on the coherence of multi-mode polaritons, showing that most channels for atom-polariton cross-thermalization are strongly suppressed. Finally, we propose other, and investigate the impact of atomic motion on the coherence of multi-mode polaritons, showing that most channels for atom-polariton cross-thermalization are strongly suppressed. Finally, we propose that most channels for atom-polariton cross-thermalization are strongly suppressed. Finally, we propose that most channels for atom-polariton cross-thermalization are strongly suppressed.

1. Introduction

Current efforts to produce and explore the properties of synthetic quantum materials take numerous forms, from ultracold atoms [1] to superconducting circuits [2–4] and electronic heterostructures [5, 6] and superlattices [7]. Cold atom techniques allow for precise control through lattice tuning [8] and Feshbach resonances [9, 10]. Superconducting quantum circuits present an opportunity to create materials from strongly interacting microwave photons, as they exhibit excellent coherence [11], strong interactions [12], and have recently been shown to be compatible with low disorder lattices [13], low loss lattice gauge fields [14], and interaction and dissipation driven phase transitions [15].

In parallel, there is now growing interest in creating materials from optical photons. Non-interacting photons have been Bose-condensed in a resonator using a dye as a thermalization medium [16]; photons have...
been made to interact weakly and subsequently Bose condense by coupling them to interacting excitons [6].
To explore strongly interacting photonic materials, it has previously been proposed to marry Rydberg electromagnetically induced transparency (EIT) tools developed to induce free-space photons to interact [17–20] with multimode optical resonators [21] to control the properties of individual photons [22], thereby introducing a real mass for 2D photons, and effective magnetic fields [23], in conjunction with Rydberg mediated interactions. It was recently experimentally demonstrated that individual cavity photons do indeed hybridize with Rydberg excitations to form ‘cavity Rydberg polaritons,’ quasiparticles [24–26] that collide with one another with high probability [27].

Formal modeling of these complex systems is incomplete. The properties of interacting free-space Rydberg polaritons have been explored in the dispersive regime [28], as well as the resonant regime for Van der Waals [29, 30] and dipolar interactions [31]. Effective models of strongly interacting two-level cavity polaritons have been developed [32], along with blockade ‘bubble’ approximations that qualitatively reflect the physics of three-level polaritons [33], but to date no effective theories of three-level cavity Rydberg polaritons exist which quantitatively reproduce the observed strong interactions, as a consequence of the intricate renormalization of the two-polariton wavefunction once the polaritons overlap in space.

In this paper, we formalize and extend the theory developed in [21, 27]. We start with formally defining the various Hamiltonians that are relevant to Rydberg polaritons in a resonator and derive the polaritonic creation and annihilation operators and interaction form. We then motivate and develop a single-mode renormalized effective theory, which has excellent agreement with full numerics employed in previous works. We follow this with a discussion on momentum-space interactions; an extension of how familiar position dependent Rydberg interactions change in the Fourier plane. Finally, we explore the impact of atomic motion on polariton coherence.

More specifically, we first show that cavity Rydberg polaritons at large separations are described by the Hamiltonian

\[ H_{\text{pol}} \sim \cos \theta \frac{\theta}{2} H_{\text{phot}} + \sin \theta \frac{\theta}{2} H_{\text{int}}, \]

where \( H_{\text{phot}} \) is the Hamiltonian describing the bare cavity photon dynamics, determined through the resonator geometry; and \( H_{\text{int}} \) is the Hamiltonian describing the Rydberg–Rydberg interactions. The polaritons thus inherit properties from both photonic and atomic constituents, with the proportion of each contribution determined by the dark state rotation angle \( \theta \) [34], providing an interaction tuning knob akin to an atomic Feshbach resonance [9]. In the remainder of the paper we examine the limitations of this model, providing quantitative refinements to various aspects of it.

In section 2 we begin with the Floquet Hamiltonian for non-interacting resonator photons [22] and formally couple these photons to an ensemble of Rydberg-dressed three-level atoms residing in a waist of the resonator [21]. In section 3 we explore the physics of an individual photon in the resonator, discovering one long lived dark polariton (with renormalized mass relative to the bare photon) and two short-lived bright polaritons. In section 4 we generalize to the case of two dark polaritons in the resonator, derive the form of the low-energy polariton–polariton interaction potential, investigate scattering into bright polariton manifolds as well as the regime of validity of the two-polariton picture in the face of interactions and loss, focusing in section 5 on collisional loss of polaritons by dark \( \rightarrow \) bright scattering. Once the interaction energy becomes larger than the dark/bright splitting the simple polaritonic picture breaks down, so in section 6 we explore the maximally challenging case of two dark polaritons in single-mode optical resonator, developing a properly renormalized effective theory of interacting polaritons (with first principles calculable parameters) that we benchmark against a complete (and numerically expensive) microscopic theory. We find excellent agreement in experimentally relevant parameter regimes, pointing the way to a fully renormalized effective field theory of multimode cavity Rydberg polaritons. In section 7 we demonstrate that a properly situated Rydberg-dressed atomic ensemble produces interactions between polaritons that are local in momentum-space. In section 8 we relax the assumption of stationary atoms and investigate the effect of atomic motion on polariton coherence in both single- and multi-mode regimes. Finally, in section 9 we conclude with a discussion of applications of cavity Rydberg polaritons to quantum information processing and strongly-correlated matter.

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2. Coupling the photons to an atomic ensemble

Here we explore how the coupling to an ensemble of three-level atoms impacts the physics of non-interacting 2D photons in multi-mode resonators. We find the emergence of long lived ‘dark’ polaritons, with dynamics similar to those of a resonator photon but renormalized mass and harmonic trapping. Off-resonant, nonadiabatic couplings to ‘bright’ polaritons limit the lifetime of the dark polaritons. We operate in the limit that the light-matter coupling energy scale is much larger than the energy scale of the photonic dynamics within the resonator, making the polaritonic quasiparticles a nearly ‘good’ basis for describing the physics, with corrections that we derive.

The second quantized Hamiltonian for photons within a single longitudinal manifold of a resonator is given by [22]:

\[ H_{\text{phot}} = \int dx\ a^\dagger(x) h_{\text{phot}}(x), \]

where \( a^\dagger(x) \) creates a photon at transverse location \( x \) and \( h_{\text{phot}} \) is the single particle Hamiltonian for a photon within the resonator, typically given by \( h_{\text{phot}}(x) = \frac{\hbar \Omega}{2m_{\text{phot}}} + \frac{1}{2} m_{\text{phot}} \omega_{\text{trap}}^2 k_0^2 - i \kappa \). Here \( x \) runs over the plane transverse to the resonator axis, \( \kappa \) parametrizes the (mode independent) resonator loss, and \( \mathbf{II} \equiv i\hbar \nabla - e\mathbf{A} \) is the mechanical momentum; the parameters of this photonic ‘Floquet’ Hamiltonian are determined by resonator geometry: mirror locations and curvatures, plus the twist of the resonator out of a single plane [22, 23].

We now insert a Rydberg-dressed atomic ensemble into the resonator as a tool to mediate interactions between the photons. To this end, the lower (S \( \rightarrow \) P) transition of this ensemble is coupled to the quantized resonator field, while the upper (P \( \rightarrow \) Rydberg) transition is coupled to a strong coherent field (see figure 1(a)). Before exploring the resulting photon–photon interactions, we must first understand how the Rydberg-dressed atoms impact the linear dynamics of individual photons. The light–matter coupling induced by the introduction of the atomic ensemble takes the form (in the frame rotating with the resonator- and Rydberg-dressing fields):

\[
H_{\text{at}} = \int dx^2dz \left\{ \phi^\dagger_0(x, z) \phi_0(x, z) \left( \delta_{\varphi} - \frac{i\gamma_r}{2} \right) + \phi^\dagger_0(x, z) \phi_0(x, z) \left( \delta_{\varphi} - \frac{i\gamma_\nu}{2} \right) + \phi^\dagger_o(x, z) \phi_o(x, z) \frac{\Omega}{2}(x, z) + \text{h.c.} \phi^\dagger_o(x, z) a(x) G(x, z) + \text{h.c.} \right\}.
\]

Here \( \phi^\dagger_o(x, z) \) and \( \phi_o(x, z) \) are the bosonic creation operators for atomic excitations at 3D location \((x, z)\) in the Rydberg and excited \((P)\) states respectively, from the ‘vacuum’ of ground state atoms. \( \gamma_r \) and \( \gamma_\nu \) are the FWHM of the Rydberg and excited states respectively. \( \delta_{\varphi} \) is the detuning of an untrapped, zero-transverse-momentum resonator photon from the atomic line and \( \delta_{\varphi} \) the two-photon detuning from EIT resonance. \( \Omega(x, z) \) is the laser induced Rabi coupling between excited \((P)\) and Rydberg states, while \( G(x, z) \) is the vacuum-Rabi
coupling strength between a resonator photon localized at transverse location \( x \) and a collective atomic excitation localized at longitudinal location \( z \), and therefore must reflect the atom density. It does not reflect the transverse spatial structure of any particular resonator mode, as \( a^\dagger (x) \) creates a transversely localized photon. Indeed it may be written as \( G(x, z) \approx d_{ge} \frac{L}{\hbar c_0} \rho \frac{\omega_{ge}}{c_0} \), where \( L \) is the length of the atomic ensemble along the resonator axis, \( L_{res} \) is the length of the resonator itself, \( d_{ge} \) is the dipole moment of the atomic transition coupled to the optical resonator, \( \omega_{ge} \) is the angular frequency of this transition, and \( \rho(x) \) is the number density of atoms at location \( x \).

Here and throughout, we incorporate losses through non-Hermitian Hamiltonians rather than through Lindbladian master equations. This allows us to identify the imaginary part of one- and two-particle eigenstates with quasi-particle decay rates ([35] III.B.2).

In this section, we have shown the photonic and atomic Hamiltonians \( H_{\text{phot}} \) and \( H_{\text{at}} \), respectively and defined the photonic and bosonic operators necessary to couple photons to an atomic ensemble. This is a general result that is valid for both single- and multi-mode resonators, evident in the fact that all the operators and Rabi couplings are position dependent, to reflect the different mode shapes and atomic distribution over them.

3. Polariton basis

The atomic density distribution exists in three dimensions, while the manifold of nearly-degenerate resonator modes to which the atoms couple is two-dimensional (here we assume \( G \ll \frac{1}{L_{res}} \), ensuring that the atoms couple to only a single longitudinal manifold of the resonator). In order to develop a formalism of two-dimensional polaritons, we define longitudinally delocalized, transversely localized collective atomic excitation operators (for \( k_{\text{laser}} \) and \( k_{\text{cav}} \) the wavevector magnitudes of the coupling-laser and resonator fields, respectively), normalized to ensure a mode independent bosonic commutation relation. To this end we choose the minimal case of an atomic ensemble of uniform density, and a uniform coupling field that propagates counter to the resonator field:

\[
\phi^\dagger_c(x) \equiv \frac{1}{\sqrt{L}} \int \frac{dz}{\frac{1}{2}} \frac{2}{dz} \phi_\mu^\dagger(x, z) e^{-i k_{\text{cav}} z} \\
\phi_c^\dagger(x) \equiv \frac{1}{\sqrt{L}} \int \frac{dz}{\frac{1}{2}} \frac{2}{dz} \phi_\mu^\dagger(x, z) e^{i k_{\text{cav}} - k_{\text{laser}}) z}.
\]

We can now rewrite the atomic Hamiltonian in the single longitudinal-mode-projected manifold as:

\[
H_{\text{at}} = \int \text{d}x^2 [W^\dagger(x)] [h_{\text{at}}] [W(x)],
\]

where \([W^\dagger] \equiv (a^\dagger(x) \phi^\dagger_c(x) \phi^\dagger_c(x))\) and 
\[
[h_{\text{at}}] = \begin{pmatrix}
0 & G & 0 \\
G & \frac{\delta_c}{2} - i \frac{\gamma_c}{2} & \Omega/2 \\
0 & \Omega/2 & \delta_2 - i \frac{\gamma_2}{2}
\end{pmatrix}.
\]

Here \( G \approx d_{ge} \frac{L}{\hbar c_0} \rho \frac{\omega_{ge}}{c_0} \), where \( L \) is the length of the atomic ensemble along the resonator axis. Note that \([h_{\text{at}}]\) has no position dependence, because we are assuming no atomic motion (covered in section 8), no inhomogeneous electric fields, and that the coupling field is large enough transversely that variations in the control field Rabi frequency \( \Omega \) are negligible. In the basis where \([h_{\text{at}}]\) is diagonal, the resulting creation operators are the generators of three varieties of polaritons: one dark (with little to no excited state participation, depending on \( \kappa \) and \( \gamma_c \)), sandwiched between two bright (with large excited state participation). In the simplest case of no detunings or cavity loss, we can express the polaritonic creation operators in the atomic basis as follows:

\[
d^\dagger = \frac{\Omega}{\sqrt{G^2 + \Omega^2}} a^\dagger(x) - \frac{G}{\sqrt{G^2 + \Omega^2}} \phi^\dagger_c(x),
\]

\[
b^\dagger_\pm = \frac{1}{\sqrt{2}} \left( \frac{G}{\sqrt{G^2 + \Omega^2}} a^\dagger(x) \pm \phi^\dagger_{\mu}(x) + \frac{\Omega}{\sqrt{G^2 + \Omega^2}} \phi^\dagger_{c}(x) \right).
\]

We are primarily concerned with the long lived and strongly interacting dark polaritons, but will include off-resonant couplings to the bright polaritons to accurately model dark polariton lifetime.

More formally, we diagonalize \([h_{\text{at}}]\) = \( \sum_{\mu, m} \hat{\mu}_m \epsilon_m \), where \( m \) is an element of \( \{d, b, \ldots, b\} \), meaning [dark, lower bright, upper bright], \( \epsilon_m \) is the energy of (ket) \( \hat{\mu}_m \) and (bra) \( \hat{\mu}_m \), and the \( \mu_m, \hat{\mu}_m \) satisfy the generalized
orthogonality condition (due to the non-hermicity of $h_{\text{at}}$): $\tilde{\mu}_i \cdot \mu_m = \delta_{im}$. Note that because of the non-hermicity of $h_{\text{at}}$, $\tilde{\mu}_m \neq \mu_i$.

The resulting polariton creation and annihilation operators are thus:

$$\chi_j(x) = [W] \cdot \mu_j,$$
$$\tilde{\chi}_j(x) = \tilde{\mu}_j \cdot [W].$$

For notational convenience only have we named the polariton creation/annihilation operators ‘$\chi_j(x)$’ and ‘$\tilde{\chi}_j(x)$’; these operators are not precisely Hermitian conjugates of one another, but are instead defined to preserve the bosonic commutation relations: $[\chi_j(x), \chi_k(x)] = [\tilde{\chi}_j(x), \tilde{\chi}_k(x)] = \delta_{jk} \delta(x - x')$. We may now write $H_{\text{at}}$ as:

$$H_{\text{at}} = \sum_m \int dx \chi_m(0) \chi_m(x) \epsilon_m.$$

The last step in writing $H_{\text{tot}}$ in the polariton basis is to decompose $a^\dagger(x)$, $a(x)$ into polariton field operators:

$$a^\dagger(x) = \sum_j c_j \chi_j^\dagger(x),$$
$$a(x) = \sum_j \tilde{c}_j \tilde{\chi}_j(x),$$

where the $c_j$, $\tilde{c}_j$ are elements of the inverse $\mu$, $\tilde{\mu}$ matrices: $c_j \equiv \mu^{-1} \chi_j$, $\tilde{c}_j \equiv \tilde{\mu}^{-1} \tilde{\chi}_j$, and the index ‘cav’ denotes the photonic slot of $\mu^{-1}$, $\tilde{\mu}^{-1}$. $H_{\text{tot}}$ can now be written as:

$$H_{\text{tot}} = \sum_j \int dx \epsilon_j \hat{\epsilon}_j \chi_j^\dagger(x) h_{\text{tot}}^{ij}(x) \chi_j(x),$$

where $h_{\text{tot}}^{ij}(x) \equiv \delta_{ij} \epsilon_i + h_{\text{phot}}(x)$.

We will operate in the limit that the difference of the eigenvalues of $h_{\text{at}}$ (the ‘dark’-‘bright’ splitting) is much larger than the spectrum of $h_{\text{phot}}$, making the $[d, b, \gamma, b^\dagger]$ basis that diagonalizes the atomic Hamiltonian a near-diagonal basis for the multimode system. This is equivalent to the statement that a particle oscillating in the trap is more accurately described as a polariton rather than a photon if the light-matter coupling is much greater than the transverse optical mode spacing. To first order in $h_{\text{phot}}/\epsilon$, the Hamiltonian projected into the dark polariton manifold is:

$$H_{\text{tot}}^{\text{pol}} = \int dx \epsilon_d \hat{\epsilon}_d \chi_d^\dagger(x) (\epsilon_d + h_{\text{phot}}(x)) \chi_d(x).$$

For $\gamma_\tau = \bar{\delta}_2 = 0; \epsilon_d = 0$ and $c_d \hat{c}_d = \frac{\Omega^2}{\Omega^2 + \Gamma^2} = \cos^2 \frac{\theta_d}{2}$ where $\theta_d$ is the dark state rotation angle [34]. We then have:

$$H_{\text{tot}}^{\text{pol}} = \cos^2 \frac{\theta_d}{2} \int dx \chi_d^\dagger(x) \left[ \frac{\Gamma^2}{2m_{\text{phot}}} + \frac{1}{2} m_{\text{phot}} \omega_{\text{trap}}^2 |x|^2 - \frac{i \kappa}{2} \right] \chi_d(x).$$

Thus, we see that to lowest-order in ratio of the photonic dynamics energy scale to the atomic coupling energy scale $\frac{\omega_{\text{trap}}}{\Omega^2 + \Gamma^2}$, the atoms simply slow down all photonic dynamics and loss by a factor of $\cos^2 \frac{\theta_d}{2}$. The dominant correction to this story is a second order resonator ($h_{\text{phot}}$)-induced dark $\rightarrow$ bright coupling, producing an effective Hamiltonian [35] (in the above limits):

$$\delta H_{\text{tot}} \approx \sum_{j \in \{b, \gamma, \text{cav}\}} \int dx \frac{H_{\text{phot}} \chi_j^\dagger(x) \chi_j(x) H_{\text{phot}}}{\epsilon_j - \epsilon_4}.$$

To compute this effect for a dark polariton in an eigenstate with energy $E = \frac{\Omega^2}{\Omega^2 + \Gamma^2} \delta \ll \sqrt{\Omega^2 + \Gamma^2}$, where $\delta$ is the two-photon detuning from EIT resonance, we take the first eigenvalue of $h_{\text{at}}$ by solving the equation $\det (h_{\text{at}} - \lambda \Pi) = 0$ and Taylor expanding to first order in $\lambda$ and then to second order in $\delta$ and $\kappa$. The correction is largely imaginary (assumming for simplicity that $\tilde{\epsilon}_c = 0, \gamma, \Omega \gg \gamma_\tau$):

$$\delta \Gamma_{\text{pol}} \equiv -i (\delta H_{\text{tot}}) \approx \frac{E^2}{\Omega^2} \gamma_\tau \approx 2 \delta^2 \cos^4 \frac{\theta_d}{2} \frac{\Omega^2}{\Omega^2 + \Gamma^2} \gamma_\tau.$$

Thus we see that loss is suppressed by the control field.

Additional contributions to polariton loss arise from inhomogeneous broadening of the Rydberg manifold, e.g. atomic motion and electric field gradients (to which Rydberg atoms are susceptible due to their large DC polarizability [36]). Such processes generate a ladder of couplings from the collective Rydberg state, that shares
the spatial configuration of the resonator mode, into collective states orthogonal to it (which therefore are bright). We explore how (random) atomic motion induces polariton decoherence in section 8; for inhomogeneous $E$-fields, the coupling rate to the bright manifold is $\gamma_{d} \approx \alpha E \delta E$, where $\alpha$ is the DC polarizability of the Rydberg state, $E$ is the DC electric field at the atomic sample, and $\delta E$ is the field-variation across it. The resultant broadening of the dark manifold is then (in the limit $\Omega \gg \gamma_{c}$) [26]:

$$\delta \Omega_{\text{pol}} \approx \frac{\gamma_{d}^{2} \sin^{2} \frac{\theta}{2}}{\Omega^{2}} \gamma_{d}$$

(3.3)

It is instructive to compare this result with the loss induced by detuning a resonator mode out of the EIT window (equation (3.2)). Both channels are quadratically suppressed, but the suppression quantity is different, emphasizing the distinction between the underlying physical processes: detuning from the EIT window couples to bright polariton manifolds that live in the resonator and are thus suppressed by the photonic component of the dark polariton, while inhomogeneous broadening couples to non-resonator bright polariton manifolds through the electric field broadening of the Rydberg component of the particle.

Having formalized the notion of a resonator polariton, it is instructive to note some differences between resonator and free-space polaritons. Cavities provide a true 2D system (for a single longitudinal mode and in the limit of small sample length $L$), where the single particle dispersion is controlled through the resonator geometry [22]. On the other hand, free-space polaritons exhibit more rigid dispersion relations. They possess different longitudinal and transverse masses, and the transverse mass may even be complex [37].

To summarize, we have derived the polaritonic creation and annihilation operators and reformulated the polaritonic Hamiltonian in second quantized form for arbitrary multi-mode resonators. With the introduction of the dark state rotation angle, we have a way to quantify the character and behavior of the quasi-particles.

Finally, we have derived a second order correction to the dark polariton lifetime arising from dark to bright polariton coupling and inhomogeneous Stark shifts; both processes result in additional polariton decoherence and are quadratically suppressed.

4. Polariton–polariton interactions

The interaction between Rydberg atoms will result in an interaction between polaritons, much as in the 1D free-space situation [17, 30, 38, 39]. In the limit that the interaction length-scale is comparable to the mode waist of the resonator, there can be a substantial renormalization of the collective atomic excitation which we investigate in section 6. For now, we assume sufficiently weak interactions that photonic—and collective atomic—components of the polariton wavefunction share the same spatial structure; note that these interactions can still dominate over kinetic and potential energies, as well as particle decay rates, so this need not be a ‘weakly interacting’ polaritonic gas in the traditional (mean-field) sense.

The bare 3D interaction between two-Rydberg atoms takes the form $V(x - x') = c_{1}(\theta)c_{2}(\theta') \gamma_{d} \gamma_{d}^{*} e^{i \theta} e^{i \theta'}$ [32], where $c_{j}(\theta)$ is the angular dependence of the interaction. S-Rydberg atoms have radially symmetric wavefunctions and so $c_{j}(\theta) \approx c_{j}(\theta) \equiv 1$. In the second quantized picture, for a thin atomic cloud (thickness $L \ll d$, where $d$ is the cavity analog of the blockade radius [18]; this restriction is needed to ensure the polariton separation is well-defined in the 2D plane) the 2D-projected interaction takes the form (with $\tilde{V}(x) = V(x; z = 0)$):

$$H_{\text{int}} = \frac{1}{2} \iint dx \, dx' \phi_{\mu}^{\dagger}(x) \phi_{\nu}^{\dagger}(x') \tilde{V}(x - x') \phi_{\nu}(x') \phi_{\mu}(x).$$

$\phi_{\mu}(x)$ may be written in the polariton basis in analogy to the way $\phi_{\mu}(x)$ was written in the polariton basis in the preceding section:

$$H_{\text{int}} = \frac{1}{2} \sum_{\mu \neq \nu} \int d \theta d \theta' \, \left[ \int dx \, dx' \chi_{\mu}^{\dagger}(x) \chi_{\nu}(x') \tilde{V}(x - x') \chi_{\nu}(x') \chi_{\mu}(x) \right].$$

(4.1)

Here $d_{\mu}$, $\tilde{d}_{\mu}$ are matrix elements of the inverse $\mu$, $\tilde{\mu}$ matrices: $d_{\mu} = \rho^{-1}_{\mu}, \tilde{d}_{\mu} = \rho^{-1}_{\mu}$, and the index ‘ryd’ denotes the Rydberg slot of $\mu^{-1}, \tilde{\mu}^{-1}$. In the absence of Rydberg loss and 2-photon detuning, $d_{\mu} = \sin \frac{\theta}{2}$.

If the interaction energy $\tilde{V}$ is small compared to the splitting between dark- and bright-polariton branches (section 5 explores the couplings and loss that violate this condition), the diagonal elements of $H_{\text{int}}$ dominate, yielding a lowest-order polariton–projected effective Hamiltonian:
\[ \hat{P}_d (H_{\text{tot}} + H_{\text{int}}) \hat{P}_d = \cos^2 \frac{\theta_d}{2} \left( \int \text{d}x \chi_d^* \left( \frac{\Pi^2}{2m_p} + \frac{1}{2} m_p \omega_0^2 |x|^2 - \frac{\text{i} \hbar}{2} \right) \chi_d \right) + \frac{1}{2} \sin^2 \frac{\theta_d}{2} \left( \int \text{d}x \text{d}x' \hat{n}_d(x) \hat{n}_d(x') \hat{V}(x, x') \right), \tag{4.2} \]

where \( \hat{P}_d \) is the dark polariton projection operator, and we have defined the dark polariton number density operator \( \hat{n}_d(x) = \chi_d^* (x) \chi_d (x) \).

By tuning the dark state rotation angle \( \theta_d \) (via atomic density and control field intensity) it is possible to move from a weakly interacting gas of ‘nearly-photonic’ polaritons (for \( \theta_d \approx 0 \)) to a strongly interacting gas of ‘nearly-Rydberg’ polaritons (for \( \theta_d = \pi \)) and explore the correlations which then develop \cite{ref21}. In this latter limit it is likely that the interactions \( \hat{V} \) become comparable to the dark-bright splitting, so the interaction potential must be renormalized as explored in section 6.

Using the formalism developed in prior sections, we have now expressed the Rydberg polariton interactions in the general context of an arbitrary multi-mode resonator. The new polaritonic interaction term in the Hamiltonian assumes only that the sample is in the general context of an arbitrary multi-mode resonator. The new polaritonic interaction term in the

\[ H_{\text{int}} = -V \hat{N}_d, \]

where \( V \) is the interaction strength. Such an interaction couples the two dark polariton state \( |\delta\rangle \) to a strongly interacting gas of dark-bright polaritons which are predominantly composed of the lossy P-state. Plugging in, the loss rate is:

\[ \Gamma_{dd \rightarrow \delta} = \text{sin}^4 \frac{\theta_d}{2} U_{\text{eff}}^2 G^2 \gamma_e. \tag{5.1} \]

This loss depends heavily on the interaction strength between the Rydberg atoms and the dark state rotation angle. Making the particles more Rydberg-like creates stronger interactions and increases the loss rate; similarly, using a higher principle quantum number increases \( U_{\text{eff}} \), further enhancing the loss.

We now investigate the second scattering process, \( |dd\rangle \rightarrow |db\rangle \) (upper or lower bright polariton). Following the same procedure, the collisional Rabi frequency is:

\[ \Omega_{dd \rightarrow db} = \langle db | \hat{V} | dd \rangle = \sqrt{2} \sin \frac{\theta_d}{2} \cos \frac{\theta_d}{2} U_{\text{eff}}. \]

Since this is an off-resonant process (final and initial state are detuned by \( \Delta = \frac{\sqrt{G^2 + \Omega^2}}{2} \)), the resulting loss rate is \( \Gamma_{dd \rightarrow db} = \frac{\gamma_e}{2} \Gamma_{db} \) with \( \Gamma_{db} = \frac{\gamma_e}{2} \) (we ignore resonator—and Rydberg—loss, as bright-state loss is dominated by P-state loss in alkali-metal atom Rydberg cQED experiments \cite{ref27}). Combining these effects/approximations yields:

\[ \Gamma_{dd \rightarrow db} = \sin^4 \frac{\theta_d}{2} \sin^2 \frac{\theta_d}{2} \frac{U_{\text{eff}}^2 G^2}{G^2} \gamma_e. \tag{5.2} \]
Once again, stronger interactions and a more Rydberg-like character for the polaritons increases the loss from this process but due to the off-resonant nature of this process there is a quadratic suppression from the light-matter coupling which separates the bright—dark polaritons in energy.

We can compare the two loss processes:

\[
\Gamma_{dd\rightarrow bb} \approx \frac{\sin^2 \theta_g}{8} \frac{G^2}{\gamma_i^2}
\]

In the limit \( G \gg \Omega_i \), \( \gamma_i \) (\( \frac{\theta_g}{2} \rightarrow \frac{\theta_g}{2} \rightarrow 1, \sin \theta_g \rightarrow 0 \)), the \( |dd\rangle \rightarrow |db\rangle \) loss channel will dominate, while in the opposite limit, interaction driven loss is dominated by the RESONATING process.

In this section, we explored dark \( \rightarrow \) bright conversion through polariton–polariton scattering. For simplicity, we analyzed this physics in a single-mode resonator with \( \delta \)-interacting polaritons (hard sphere approximation of long-range interactions) and showed that there are in fact two loss channels, with rates exhibiting similar functional forms, each dominating in a region of parameter space.

6. Effective theory for interacting polaritons

The simple polariton-projected interacting theory introduced in section 4 is an accurate description only for polaritons whose interaction energy is less than the EIT linewidth [26] for all pairs of atoms comprising the polaritons. As two polaritons approach one another and their wavepackets begin to spatially overlap, some terms in the interaction energy diverge; full numerics (see SI of [27] for details of the approach) reveal that the two-polariton wavefunction is renormalized to suppress such overlap, at the cost of additional (finite) interaction energy, and loss. Physically, at small separations and strong interactions, the pair distribution of Rydberg excitations no longer follows the wavefunction of the cavity field. We now explore the extreme limit of this physics: a single-mode optical resonator that is moderately-to-strongly blockaded, to develop a low-dimensional effective model in the basis of near-symmetric collective states that produces optical correlations in agreement with full numerics.

The ‘brute force’ numerical approach that we have previously employed accounts for the three-level structure of each atom in the atomic ensemble, and the interactions between atoms. It accurately reproduces observed correlations [27] at the expense of a Hilbert space which grows as \( N^m \), where \( N \) is the number of atoms.
in the atomic ensemble and \( m \) the number of polaritons in the system; and quickly becomes computationally intractable when motional dynamics of the polaritons are included by allowing for multiple resonator modes. The problem of an extremely large Hilbert space is exacerbated since many-body physics \([21, 40]\) demands both multiple resonator modes and significantly more than two excitations, which makes numerically computing the behavior of the system completely untenable without a coarse-grained effective theory.

To the extent that ‘polaritons’ are well-defined collective excitations whose atomic spatial structure reflects the cavity mode functions, it should be possible to develop an effective theory whose Hilbert space size is independent of the atom number, making explorations of multimode/many-body physics tractable. In this section we demonstrate, for a single optical resonator mode, an approach to handle the suppression of short-range double-excitation of the Rydberg manifold, arriving at a coarse-grained effective theory including both dark and bright polaritons whose parameters may be calculated from first principles.

Consider two excitations, either atomic or photonic, in an atomic ensemble coupled to a single-mode resonator. In the absence of interactions, we can explicitly write out the collective states that couple to the resonator field: \(|CC\rangle, |CE\rangle, |CR\rangle, |EE\rangle, |ER\rangle, |RR\rangle\). These states represent two excitations as photons in the resonator, one photon and one p-state excited atom, one photon and one Rydberg atom, two excited p-state atoms, one p-state and one Rydberg atoms and two-Rydberg atoms respectively. The Hamiltonian is closed in this basis, and takes the form \([41]\):

\[
H = \begin{pmatrix}
-\i\kappa & \frac{G}{\sqrt{2}} & 0 & 0 & 0 & 0 \\
\frac{G}{\sqrt{2}} & -\i\kappa + \gamma_p & \frac{\Omega}{2} & \frac{G}{\sqrt{2}} & 0 & 0 \\
0 & \frac{\Omega}{2} & -\i\kappa + \gamma_p & \frac{G}{2} & 0 & 0 \\
0 & \frac{G}{\sqrt{2}} & 0 & -\i\gamma_p & \frac{\Omega}{\sqrt{2}} & 0 \\
0 & 0 & \frac{G}{2} & \frac{\Omega}{\sqrt{2}} & -\i\gamma_p & \frac{\Omega}{\sqrt{2}} \\
0 & 0 & 0 & 0 & -\i\gamma_p & \frac{\Omega}{\sqrt{2}} \\
\end{pmatrix}
\]

where \(\gamma_p \equiv \gamma_\alpha + 2\i\delta_p\) is a complex linewidth incorporating the P-state detuning. The above basis and corresponding Hamiltonian no longer accurately describe the physics once the Rydberg–Rydberg interactions become comparable to the dark–bright splitting: under such conditions, the \(|RR\rangle\) is renormalized due to Zeno suppression of excitation of Rydberg atom-pairs at small separation. We posit that the model can be ‘fixed’ by considering coupling to a new collective ‘two-Rydberg’ state \(|\tilde{RR}\rangle\), where the tilde signifies that the relative two-Rydberg amplitudes are renormalized by interaction; furthermore, the coupling from \(|ER\rangle\) to \(|\tilde{RR}\rangle\) will no longer be \(\frac{\Omega}{\sqrt{2}}\).

To ascertain the form of the state \(|\tilde{RR}\rangle\), we will examine the equations of motion in the frequency domain under the non-Hermitian Hamiltonian in the bare-atomic basis within the two-excitation manifold. We work in a frame that rotates with an energy \(2\Omega_p\), convenient for performing scattering experiments of pairs of photons injected by a probe at energy \(\Omega_p\). We assume that while the state \(|RR\rangle\) is renormalized by the interactions, the state \(|ER\rangle\) is not, and reflects the non-interacting polaritonic wavefunctions of the preceding section; this is the central assumption of this section, and is validated by numerics. Corrections to \(|ER\rangle\) would enlarge the Hilbert space and may be included as higher-order terms in the effective theory.

Following notation from \([27]\) SI L, the equation of motion for the amplitude of two-Rydberg excitations in atoms \(\alpha\) and \(\beta\), \(C_{\alpha\beta}^{R}\), is given by \(i\partial_t C_{\alpha\beta}^{R} = i(U_{\alpha\beta}^{RR} + 2\delta_\beta + \i\gamma_\beta)C_{\alpha\beta}^{R} + i\Omega(C_{\alpha\beta}^{ER} + C_{\alpha\beta}^{C\beta})\), where \(U_{\alpha\beta}^{RR}\) is the interaction between Rydberg-excited atoms \(\alpha\), \(\beta\), \(\delta_\beta\) is the two-photon detuning, \(\gamma_\alpha\) is the Rydberg state linewidth and we have assumed that the control field \(\Omega\) is uniform across the atomic ensemble. Here \(C_{\alpha\beta}^{R}\) and \(C_{\alpha\beta}^{C\beta}\) are the amplitudes to have P- and Rydberg-excitation in atoms \(\alpha\), \(\beta\), \(\gamma\), \(\delta\) respectively, and satisfy \(C_{\alpha\beta}^{R} \equiv C_{\alpha\beta}^{C\beta}\). The assumption that \(|ER\rangle\) is not renormalized by the interactions is equivalent to \(C_{\alpha\beta}^{ER} = \frac{\delta_\alpha \delta_\beta}{\sum \delta_\alpha \delta_\beta}\), where \(g_\chi\) is the vacuum-field light-matter coupling strength of atom \(\chi\). Plugging this expression into the equation of motion for \(C_{\alpha\beta}^{RR}\) yields the un-normalized two-Rydberg state amplitude: \(C_{\alpha\beta}^{RR} = \frac{\delta_\alpha \delta_\beta}{(U_{\alpha\beta}^{RR} + \i\gamma_\alpha)}\), where we defined the complex detuning \(\delta_\beta = \delta_\beta + \i\frac{\Omega}{2} - \Omega_p\). We can now write the normalized collective state \(|\tilde{RR}\rangle\), its effective interaction energy \(\tilde{U}\) and effective coupling \(\frac{\tilde{\Omega}}{\sqrt{2}}\) to \(|ER\rangle\) as:
than one might naïvely anticipate probe strength from residual deviations can be parametrized as corrections to and not with uncorrelated photons escaping at the same average rate; in an experiment where our only access to the Rydberg δ agrees with the full numerical model up to 1. (a) $g(\tau = 0)$ versus probe laser detuning. Scanning the probe laser frequency affects both the detuning to the P-state and the two-photon detuning to the Rydberg state. (b) $g(\tau = 0)$ versus P-state detuning $\delta_e$. The detuning of the P-state $\delta_e$ is varied while keeping the two-photon detuning $\delta_2$ zero. (c) $g(\tau = 0)$ versus light-matter coupling field strength $G$, $g(\tau = 0)$ versus probe detuning $\delta$ (inset). The light-matter coupling strength per atom $g$ is varied from $2\pi \times 0.07$ MHz to $2\pi \times 3$ MHz. For the inset, we scan the probe frequency. (d) $g(\tau = 0)$ versus control field strength $\Omega$. There is good agreement between the full numerics and the renormalized effective theory except for a small region at about $\Omega = 2\pi \times 1-3$ MHz. (e) $g(\tau = 0)$ versus Rydberg–Rydberg interaction coefficient $C_6$. $C_6$ is varied from $2\pi \times 10^6$ MHz m$^6$ to $2\pi \times 10^{10}$ MHz m$^6$, which corresponds to Rydberg states in the range $n \sim 48-1935$. The upper limit on the strength of the blockade is the cavity enhanced optical depth. Once $C_6$ is large enough to fully blockade the sample, the interaction strength, and therefore $g$, saturate. A control field of $\Omega = 2\pi \times 1$ MHz was employed for this plot. The first principles renormalized theory accurately reproduces the blockade from weak—to strong—interactions.

\[
\mathcal{R}(\Omega) = \frac{\sum_{i} \sigma_i^{(1)} |R_i \rangle \langle R_i|}{\sum_{i} |\sigma_i^{(1)}|^{2}}
\]

\[
\mathcal{U} = \frac{\mathcal{R}(\Omega) \mathcal{U}(\Omega)\mathcal{R}(\Omega)}{\sum_{i} |\sigma_i^{(1)}|^{2} \Omega_{RR}^{(0)}}
\]

\[
\frac{\Omega}{\sqrt{2}} = \frac{\mathcal{R}(\Omega) \sum_{i} \sigma_i^{(1)} |E_{R}|}{\mathcal{R}(\Omega) \sum_{i} |\sigma_i^{(1)}|^{2}}
\]

In the extreme limit of strong interactions across all space $U_{RR}^{(0)} \gg \delta$; $\mathcal{U} = \frac{\sum_{i} |\sigma_i^{(1)}|^{2}}{\sum_{i} |\sigma_i^{(1)}|^{2} \Omega_{RR}^{(0)}} = \frac{C_6}{w_c}$, where $w_c$ is the mode waist; the pre-factor makes this interaction substantially weaker than one might naïvely anticipate—the interaction predominantly arises from particles separated by $\sim 2.2 w_c$, and not $w_c$.

We next benchmark the validity of this effective theory against a full microscopic numerical model [27]. As a figure of merit we have chosen the temporal intensity autocorrelation function $g_2(\tau)$, which compares the rate at which pairs of photons escape the resonator with separation in time of $\tau$ to what would be expected for uncorrelated photons escaping at the same average rate; in an experiment where our only access to the Rydberg physics is through photons leaking from the resonator, $g_2(\tau)$ characterizes the strength of polariton interactions, with $g_2(\tau = 0) \ll 1$. $g_2(\tau = 0)$ is independent of probe strength. We compare $g_2(0)$ versus probe detuning $\delta_1$ (figure 3(a)), $g_2(0)$ versus p-state detuning $\delta_e$ (figure 3(b)), light–matter coupling strength $G$ (figure 3(c)), Rydberg control field strength $\Omega$ (figure 3(d)) and van der Waals interaction coefficient $C_6$ (figure 3(e)) between brute force numerics of many individual three-level atoms and the effective theory developed above. It is apparent that our approach largely agrees with the full numerical model up to ‘noise’ arising from randomness in the atom locations. We expect that residual deviations can be parametrized as corrections to $\Omega$ and $\mathcal{U}$ due to coupling to bright polariton manifolds, and a slight enlargement of the Hilbert space to incorporate the additional states coupled to.
We note that there are qualitative differences between the physical behavior of resonator and free-space
interaction correlation functions, as demonstrated by our recent experimental work [27] and [17, 20, 42]. In a single-mode resonator coupled to Rydbergs, we have observed an effectively 0D environment by restricting the cloud thickness. This is equivalent to tunneling in and out of a zero dimensional quantum dot. On the other hand, free-space systems are intrinsically at least 1D; the photons can move longitudinally and avoid each other, experiencing a true position dependent interaction. This difference in system dimension results in very different quantitative g behavior and parameter scaling.

To explore more complex condensed matter models like Laughlin droplets and crystalline phases, multiple particles and resonator modes are needed. Any brute force analytical approach quickly runs into the obstacle of very large computation times due to the rapid increase of the system’s Hilbert space. To circumvent this, we have developed a renormalized single-mode resonator effective field theory that shows very good agreement with numerics and can be extended to the many particle, multi-mode regime, allowing for computationally tractable modeling of larger physical systems.

7. Momentum-space interactions

A resonator which exhibits manifolds of nearly-degenerate modes may be understood as a self-imaging cavity: a localized spot living within such a manifold is re-focused onto itself after a full transit around the cavity. In-between, the localized spot undergoes diffraction, equivalent to the time-of-flight expansion of a free atomic gas [43]. Indeed, what the optics community calls a ‘Fourier plane’ is what a cold atom experimentalist calls ‘momentum-space’: the momentum of the photon in the initial (‘reference’ or ‘image’) plane has been mapped onto its position in the ‘Fourier plane’ [44]. We can formulate \( h_{\text{shot}} \) for any plane we choose to define as ‘real space’. Placing a Rydberg-dressed atomic sample in that plane will produce real space (position dependent) interactions in the system; a sample in any other place will result in interactions that are a mix between real- and momentum-space, depending on the exact plane the atoms are placed. Accordingly, it should be possible to realize interactions which are local in momentum-space by placing a Rydberg-dressed atomic ensemble that mediates these interactions in a Fourier- or nearly-Fourier- plane of the optical resonator.

We explore this idea formally by extending the cavity Floquet Hamiltonian engineering tools of our prior work [22] to the interacting regime. A thin gas of Rydberg-dressed atoms placed in a plane separated from the ‘reference’/‘image’ plane by a ray-propagation matrix \( M = \begin{pmatrix} a & b \\ c & d \end{pmatrix} \) produces interactions of the form:

\[
H_{\text{int}} = \frac{1}{2} \sin^2 \frac{\theta_d}{2} \int dx dx' \chi_d^\dagger(x) \chi_d^\dagger(x') \hat{V} \left( a(x - x') - b \frac{\hbar}{i} (\hat{p} - \hat{p}') \right) \chi_d(x') \chi_d(x).
\]

In the simple case of a delta-interacting gas of atoms (for overlapping polaritons whose characteristic sizes exceed the blockade radius, an accurate but simplified model considers the long-range interacting polaritons as hard spheres; overlapping pieces of the polaritonic wavefunction scatter) placed in such an intermediate plane (a distance \( z \) from the reference plane), we employ this result to transform an expression where the dark polariton creation/annihilation operators and interaction potential are written in the intermediate plane to one where the interaction is transformed and all operators are written in the ‘reference’ plane:

\[
H_{\text{int}} = \frac{1}{2} \sin^2 \frac{\theta_d}{2} \int dx dx' \delta(x - x') \chi_d^\dagger(x') \chi_d(x) \chi_d^\dagger(x') \chi_d(x) = \frac{1}{2} \sin^2 \frac{\theta_d}{2} \int dx d\delta \Delta \chi_d^\dagger(x + \delta) \chi_d(x - \delta) e^{i \frac{\theta_d}{2} (x^2 - z^2)} \chi_d(x + \Delta) \chi_d(x - \Delta).
\]

The resulting polariton interaction is no longer purely local in real space, and indeed can ‘instantaneously’ transport polaritons through space.

The most extreme example of such an interaction occurs if the mediating gas is placed in a Fourier plane of the system, \( a = 0 \), \( b = f \)

\[
H_{\text{int}} = \frac{1}{2} \sin^2 \frac{\theta_d}{2} \int dx dx' \chi_d^\dagger(x) \chi_d^\dagger(x') \hat{V} \left( - \frac{\hbar}{i} (\hat{p} - \hat{p}') \right) \chi_d(x') \chi_d(x),
\]

an interaction that is local in momentum-space.
8. Impact of atomic motion

In this section we investigate the effects of atomic motion on the coherence properties of individual Rydberg polaritons in both single- and multi-mode regimes. We relax the assumption, employed to this point in the manuscript, that the atoms remain spatially fixed, and instead allow them to move ballistically through space. The impact of this motion upon the P-state is ignored because the P-state linewidth of an alkali-metal atom (∼2π × 6 MHz) is typically much larger than any Doppler broadening effect at μK temperatures (∼2π × 100 kHz for Rb), and furthermore, dark polaritons by construction spend very little time in the P-state (they are ‘dark’ to it). In what follows, we will assume the polariton is almost entirely Rydberg-like (the typical experimental situation) [27]; if this is not the case, all doppler-induced broadenings and cross-couplings must be multiplied by a factor reflecting the Rydberg-participation of a polariton sin²θ/2.

We incorporate atomic motion into the Hamiltonian in the bare-atom basis by allowing each atom to have a time-dependent coupling-phase to the probe and control fields resulting from its time-varying position:

\[ H = \omega_c \hat{a}^\dagger \hat{a} |c\rangle \langle c| + \sum_j \omega_{j} |j\rangle \langle j| + \omega_r |r\rangle \langle r| + \sum_j \{ |G_j(x_j + v_j t)|c\rangle \langle j| + \Omega_j(x_j + v_j t)|r\rangle \langle j| \} + \text{h.c.} \],

where \(x_j \) and \(v_j \) are the positions and velocities of the atoms, drawn from a normal distribution reflecting the sample rms size and temperature. The effect of atomic motion, then, is to mix the collective states that couple to the resonator modes with those that, in the absence of atomic motion, do not couple to it. To see this formally, we write the Hamiltonian in the basis of the instantaneous collective eigenstates, resulting in a Hamiltonian of the form:

\[ H = \sum_m E_m(t) |m(t)\rangle \langle m(t)| + |\dot{r}(t)\rangle \langle r(t)|, \quad (8.1) \]

where \(|m(t)\rangle\), \(E_m(t)\) are the instantaneous polaritonic eigenstates and their corresponding energies, and \(|\dot{r}(t)\rangle \langle r(t)|\) is an extra term introduced by this time-dependent change of basis, capturing the effects of atomic motion in the instantaneous collective Rydberg state \(|r(t)\rangle\). In what follows, we examine the form of this final term for the particular case of twisted resonators which produce a Landau level for light [23, 40], so the mode functions are Laguerre–Gauss \( \Psi_l(z = x + iy) = \sqrt{\frac{2^l}{\pi}} z^l e^{-|z|^2} \), with angular momentum \(L = \hbar l\). For a polariton in a mode with angular momentum \(\hbar l\), \(|r(t)\rangle = |\eta(t)\rangle\), with \(|\eta(t)\rangle = \frac{\sum_j e^{i(l+\eta)\frac{k}{\omega_c}} |j\rangle}{\sqrt{\sum_j |\Psi_l\left(\frac{l + \eta}{\omega_c}\right)|^2}} \) is the state where all atoms are in the ground state except for the \(j\)th which is in the Rydberg state, and \(\alpha_l(t) = \sqrt{\sum_j |\Psi_l\left(\frac{l + \eta}{\omega_c}\right)|^2} \) is the normalization factor for mode \(l\). The time derivative \(\frac{d}{dt}|\eta(t)\rangle\) is:

\[
|\dot{\eta}(t)\rangle = \sum_j \frac{\hat{k} \cdot \nabla_{\hat{r}} e^{i(l+\eta)\frac{k}{\omega_c}} \Psi_l\left(\frac{l + \eta}{\omega_c}\right)}{\alpha_l(t)} |j\rangle \\
+ |\eta(t)\rangle \left( \frac{\dot{\alpha}_l(t)}{\alpha_l(t)} - \frac{\eta(t)}{\alpha_l(t)} \right) \\
+ \sum_j \frac{e^{i(l+\eta)\frac{k}{\omega_c}} \Psi_l\left(\frac{l + \eta}{\omega_c}\right)}{\alpha_l(t)} \nabla_{\hat{r}} \hat{r} |j\rangle, \quad (8.2)
\]

where the index \(|j\rangle\) runs over all atoms in the sample, \(\hat{r} = \vec{x} + \vec{v} t\), \(w_c\) is the resonator mode waist, \(k\) is the wavevector defined by the relative orientation of the cavity—and control—fields and \(\nabla_{\hat{r}}\) refers to the gradient with respect to \(\hat{r}\). These terms of \(H\) in the instantaneous eigen–basis have three effects: mixing polaritons in modes of different angular momenta, coupling to bright polariton manifolds orthogonal to the resonator field, and random shifts of the energy of the mode in which the polariton resides. We now investigate the extent to which each of the terms above induce each of these effects. We can rewrite equation (8.2) as \(|\dot{\eta}(t)\rangle = |T_{\eta}\rangle + |T_{\dot{\eta}}\rangle + |T_{\ddot{\eta}}\rangle\), where we have defined the following:
Even for a maximally degenerate concentric cavity, most collective Rydberg states that one can generate (for example through atomic motion, above) are orthogonal to all resonator modes, because their spatial form along the cavity axis does not match the cavity field (equivalently, their longitudinal momentum is not that of a cavity photon). As a consequence, most of the dynamics generated by coupling to \(|T_2\), \(|T_3\), and \(|T_4\) consists of coupling to bright polariton manifolds with no corresponding dark (resonator-like) mode. We bound these effects by assuming, at zeroth order, that all of each coupling is to these bright manifolds. The strength of this coupling is thus the normalization of the corresponding \(T_i\) and in the case of \(|T_1\):

\[
\langle T_1 | T_1 \rangle = \sum_{k} k^2 |w_k| \langle \Psi_k | \Psi_k \rangle,
\]

\[
\langle T_1 | T_{1i} \rangle = \left( k_{ph} \right)^2 \sum_{k} \left| \frac{\langle \Psi_k | \Psi_k \rangle}{\alpha_i(t)} \right|^2 = \left( k_{ph} \right)^2.
\]

We can now write \(|T_1\) \(= k_{ph} |T_1\rangle = k_{ph} \tilde{T}_1\rangle\), where \(\tilde{T}_1\rangle\) is the normalized state-vector corresponding to state \(|T_1\). This corresponds to a Rabi coupling of strength \(\sim k_{ph}\), to a bright polaritonic state which is detuned by \(\Omega\), and a resulting dark \(\rightarrow\) bright loss rate of:

\[
\delta \Gamma = \frac{(k_{ph})^2 \gamma_c}{(c/2)^2} = \frac{2(k_{ph})^2}{\Omega^2} \gamma_c.
\] (8.3)

A small fraction of \(|T_1\) overlaps with other degenerate resonator modes, corresponding to an atomic motion-induced polaritonic motional diffusion:

\[
\langle \gamma_m(t) | T_1 \rangle = \frac{\sum_{k} k_{ph} \Psi_k^m \left( \frac{c}{2} \right) \Psi_k \left( \frac{c}{2} \right)}{\sqrt{\sum_{k} |\Psi_k| \left( \frac{c}{2} \right)|^2 \sum_{m} |\Psi_m| \left( \frac{c}{2} \right)|^2}}.
\]

The expected value of this term is zero since the average atomic velocity is zero: \(\langle \gamma_m \rangle_{t=0} = 0\). The rms coupling, however, is non-zero:

\[
\sqrt{\langle \{\gamma_m, T_1\} \rangle^2} = \frac{k_{ph} \sqrt{N_0}}{c^2} C_{l=1-m},
\] (8.4)

where \(N_0\) is the number of atoms in mode \(l = 0\) and \(C_{l=1-m}= \frac{C_{l=1-m}}{\sqrt{J_{lm}}= \sqrt{\frac{2l+1-2m+1}{\lambda_{lm}}}}\) is a generalized the Doppler coupling matrix element between modes \(l\) and \(m\), incorporating the fact that higher angular momentum modes contain more atoms, and thus provide a smoother atom distribution. We can expand this matrix element for large \(l \approx m\) yielding \(C_{l=1-m} \approx e^{-\lambda_m (2l+2m-1)} \lambda_{2l+2m-1}^2 \sqrt{\frac{2l+1}{(2l+1)!}}\), indicating diffusion only into nearly-adjacent modes.

Last, the rms energy shift (inhomogeneous broadening) of the collective Rydberg state induced by this term is given by:

\[
\sqrt{\langle \{\gamma_m, T_2\} \rangle^2} = \frac{k_{ph} \sqrt{N_0}}{c^2} \sqrt{\frac{2l+1-2m+1}{l(2l)!}}.
\] (8.5)

The second term of equation (8.2) is:

\[
|T_2\rangle = |\gamma_1\rangle \frac{1}{2w_c} \sum_{k} \Psi_k \cdot \Psi_{\gamma_1} \langle |\Psi| \rangle^2 = |\gamma_1\rangle \left( \frac{\alpha_1}{\alpha_i} \right)
\]
Table 1. Atomic motion induced homogeneous/inhomogeneous broadening, as well as rms diffusion/cross-thermalization matrix elements. For broadening terms, the angular momentum of the state under consideration is \( l \); for cross-thermalization we consider nearby angular momentum states with mean \( \langle l \rangle \) and separation \( 2\Delta l \). Computed broadening and cross-thermalization terms are upper bounds on the respective processes. Note that \( \kappa_w \approx 100 \) for typical experiments [27], so \( \kappa_{Wy} \) terms dominate strongly over \( \kappa_{Wl} \) terms until \( l \) becomes large.

| Broadening | Cross-thermalization |
|------------|----------------------|
| \( T_1 \)  | \( \sqrt{\Gamma(l + \frac{1}{2})} \) |
| \( T_2 \)  | \( \sqrt{\Gamma(l + 1)} \) |
| \( T_3 \)  | \( \sqrt{\Gamma(l + 2)} \) |

Again, we examine how this term couples to the lossy manifold of bright polaritons:

\[
\sqrt{(T_2 | T_1)} = \frac{v_{th}}{2w_t} \frac{\Gamma(l + \frac{1}{2})}{l!} \approx \frac{v_{th}}{2 \sqrt{l + \frac{1}{2}} w_t}. \tag{8.6}
\]

This broadening comes from the time-dependent probe field coupling that the atoms experience as they move within the mode; it is much smaller than \( \kappa_{Wy} \). From the functional form of \( |T_1| \) we can also see that it does not couple modes of different angular momenta \( \langle r_{nl} T_2 \rangle = \delta_{l,m} \left( -\frac{\omega}{m} \right) \).

The third term, similar to the first, produces both a broadening and a shift in the dark polariton energy. We can see that this term couples to the bright collective manifold with matrix element:

\[
\sqrt{(T_3 | T_3)} = \frac{v_{th}}{2w_t} \sqrt{3l + 1}. \tag{8.7}
\]

We can similarly evaluate how this term couples to other states in the dark collective state manifold:

\[
|\langle r_{nl} T_3 \rangle|^2 \approx \frac{v_{th}^2}{2w_t^2} \frac{1}{N_0} \frac{1}{(l')^2}, \tag{8.8}
\]

where \( C_{l' \rightarrow l} = \frac{2 \Gamma}{{\pi}^2} 2^{l + \frac{1}{2}} m(m + 1) \frac{\Gamma}{\Gamma} \frac{\Gamma}{\Gamma} \) is the coupling element between modes of the resonator that captures mode spatial overlaps, coupling induced by atomic motion and increasing mode area. This cross-thermalization coupling element converges for large \( l \approx m \) to \( C_{l' \rightarrow l} \approx \frac{6}{\sqrt{l}} l^{-\frac{1}{2}} e^{-\left[l - m - \frac{1}{2}\right]^2 / 8} \). The \( l \)-dependence in both the broadening and energy shift terms arises from the more rapid phase accrual of higher angular momentum modes.

The total broadening of a mode with angular momentum \( l \) is thus \( \Gamma^\text{Doppler} \approx 2 \frac{k_{Wy}^2}{\Gamma^2} \frac{\gamma}{\kappa_{Wy}} \left( 1 + \frac{2\Delta l}{\Gamma} \right) \) the rms Rabi coupling of mode \( l \) to mode \( m \) is \( \Omega_{lm} \approx \frac{\sqrt{2}}{2\pi} \frac{k_{Wy}}{\sqrt{N_0} \kappa_{Wy}} e^{-\gamma / 2 \Delta l} \sqrt{1 + \frac{2\Delta l}{\gamma} \frac{\Omega_{lm}}{2\Delta l}} \), where \( 2\Delta l = l - m, 2\Delta l \equiv l + m \). Noting that \( \kappa_w \approx 100 \) for typical experiments [27], approximating further yields: \( \Gamma^\text{Doppler} \approx 2 \frac{k_{Wy}^2}{\Gamma^2} \frac{\gamma}{\kappa_{Wy}} \), \( \Omega_{lm} \approx \frac{\sqrt{2}}{2\pi} \frac{k_{Wy}}{\sqrt{N_0} \kappa_{Wy}} e^{-\gamma / 2 \Delta l} \). In fact, summing over all final states, the net coupling out of mode \( l \) is \( \Omega_l \approx (16\pi)^{1/4} \frac{k_{Wy}}{\sqrt{N_0} \kappa_{Wy}} \), independent of \( l \).

To summarize, atomic motion results in homogeneous and inhomogeneous broadening of the dark polaritons, along with state diffusion. All effects arise from recoil-induced differential motion of the Rydberg-excited atom, expressed as \( \hbar(t) \): the first couples (equations (8.3), (8.6), (8.7)) the system to modes that decay into free-space due to their spatial symmetry, while the second term (equations (8.4), (8.8)) quantifies thermalization of atomic degrees of freedom into the polaritonic degrees of freedom as a result of atomic motion. The former effect is suppressed by the detuning of the uncoupled (and therefore bright) modes from the dark manifold, while the latter effect is suppressed because atomic motion is random, so it is only the shot-noise in the motion of the ensemble comprising the polariton that leads to polaritonic mode coupling. All atomic motion decoherence effects are summarized in Table 1.

Doppler decoherence fundamentally arises from the relative motion of the atoms comprising the matter-component of a polariton relative to the field comprising the photonic component; as a consequence, the Doppler decoherence is sensitive to the canonical momentum of the optical field, not its mechanical momentum [22]. This distinction is particularly important in cavities whose near-degenerate manifolds represent a particle in a magnetic field, because although the Landau level is translationally invariant in a fundamental sense, the
choice of gauge arising from resonator twist means that polaritons further from the resonator axis are more susceptible to Doppler decoherence, apparent in the $l$-dependence of the loss terms above.

9. Outlook

In this paper we have presented a field theory of interacting cavity polaritons in the strongly interacting regime, including a formal treatment of interaction and atomic motion-induced loss channels (Table B1 presents a summary of all such channels, with numerical values for realistic experimental parameters and potential future improvements), and the development of a renormalized single-mode theory. We also demonstrate that by varying the location of one or more Rydberg-dressed atomic ensembles within the resonator, the interactions can be tuned continuously from local in position-space to local in momentum-space.

A particular non-planar resonator configuration yields a degenerate set of Laguerre–Gaussian eigenmodes which corresponds to a Landau level for photons [23]. Hybridizing these modes with Rydberg atoms to introduce strong interactions between photons makes this a promising platform to study fractional quantum Hall physics [45–48]. Straightforwardly extending our current single-mode effective field theory to include multiple resonator modes and particles, analogous to its free-space counterpart [28], allows us to model this platform and make quantitative predictions for future experimental explorations of fractional quantum Hall physics.

Furthermore, by combining a renormalized cavity Rydberg-polariton field theory with recently demonstrated cavity Rydberg polariton Keldysh-techniques [49], we are now in a position to accurately model the physics of cavity polariton crystals and Laughlin puddles [21], plus quantitative analysis of photonic QIP and quantum repeater protocols [50, 51].

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Appendix A. Resonator degeneracies and photon localization

In sections 2–5, we treat photons as objects that can occupy a completely arbitrary spatial mode in the 2D plane transverse to the resonator axis. In practice, resonator geometry can impose additional symmetries on the allowed photon wavefunctions, and limit the permissible degree of photon localization. In what follows we explore the wavefunction constraints imposed by various resonator configurations.

It is convenient to begin by considering a general non-degenerate spherical mirror Fabry–Perot resonator [52], whose modes are enumerated with three indices $l, m, n$; the first index $l$, is the longitudinal mode number, while $m, n$ index the transverse mode:

The most extreme case is degeneracy of all the transverse modes of a resonator, $\omega_{l=0} = \omega_0$ achieved in planar and concentric cavities [52] (note that we have re-indexed the longitudinal modes of the concentric resonator). Planar resonators are constructed with flat mirrors (radius of curvature $\to \infty$) very close together, while concentric cavities consist of two mirrors separated by the sum of their radii of curvature. In both configurations the space of allowed photon wavefunctions is not constrained by any symmetries, and arbitrarily small spots can be created at any point in 2D space (see figure A1(b)).

The next most extreme case is families of degenerate modes that still require two indices to enumerate, but with restrictions on the indices. An example of this is the confocal resonator; two mirrors with radii of curvature $R$ placed a distance $R$ apart. Such a resonator exhibits $\omega_{l=0} = \omega_{l=0}$. The constraint that $m + n$ is either even or odd imposes a reflection symmetry across the origin; the photon may be arbitrarily well localized in space, but must simultaneously exist at this mirror-image location (see figure A1(c)).

The next case is degenerate families that may be indexed with only a single parameter, which may themselves be further broken down into two sub-categories: (1) families in which the index takes on only a finite number of values; and (2) families in which the index takes on a countably infinite number of values. A spherical mirror Fabry–Perot falls into the first category; $\omega_{l=0} = \omega_{l=0}$ another example is an astigmatic resonator whose length is tuned to enforce degeneracy such as $\omega_{l=0} = \omega_{l=0}$ [26] (see figure A1(d)). The latter category could be achieved in an astigmatic resonator tuned to confocality on only one axis: $\omega_{l=0} = \omega_{l=0}$ or in a non-planar resonator by imposing twist which is a rational fraction of $2\pi$, as in [23]; $\omega_{l=0} = \omega_{l=0}$ (see figure
The reduced degeneracy strongly constrains the wavefunctions which may be represented by the family, to the point that the physical interpretation of these families is often quite unclear. Indeed, the twisted resonator explored in [23] exhibits threefold rotational symmetry, and quantum geometry, meaning that has a minimum spot size; this system may be understood as a Landau level on the surface of a cone, quite an exotic manifold indeed.
Appendix B. Polariton loss channels

Table B1. Summary of polariton loss channels. Functional form of the polaritonic loss mechanisms presented in the paper and their numerical values for realistic experimental parameters. The parameters used are: $G = 2\pi \times 10$ MHz, $\Omega = 2\pi \times 4$ MHz, $\kappa = 2\pi \times 1.5$ MHz, $\gamma = 2\pi \times 6$ MHz, $\gamma_p = 2\pi \times 100$ kHz, $\delta = 2\pi \times 500$ kHz, $w_c = 14$ $\mu$m. For the dark to bright polariton coupling, the dark polariton energy $E_d$ is related to the two-photon detuning $\delta$ by $E = \cos^2(\gamma/2\delta)$. The 100$S$ Rydberg state has a scalar polarizability $\alpha = 6200$ MHz/(V/cm)$^2$. With careful calibration, the background electric field can be reduced to as low as $10$ mV cm$^{-1}$, with a variation of $1$ mV cm$^{-1}$ over the atomic cloud. This results in the broadening $\gamma_{30} = \alpha b E$, which is quadratically suppressed by the control field $U_{\text{control}}$. $U_{\text{control}}$ is a phenomenological interaction strength; we assume the dark polaritons interact weakly which means we need the interaction to be much smaller than the EIT linewidth $\Gamma_{30}$, which is related to the two-photon detuning $\delta$ by $\Gamma_{30} = \gamma_{30} b E$. The parameters used here. For the $100S$ state ($G = -2\pi \times 56.4$ MHz/m$^3$), the interaction energy between two polaritons separated by $\sim 1.5$ $\mu$m (this is equivalent to approximately $0.66$ polaritons per mode waist) $U_{\text{control}} = 2\pi \times 320$ kHz, which satisfies the weak interaction condition. Comparing the $|dd\rangle \rightarrow |bb\rangle$ and $|dd\rangle \rightarrow |db\rangle$ processes, we can see that for our parameters ($G > \Omega$, $|dd\rangle \rightarrow |bb\rangle$ will dominate. The atoms in our system have a temperature of $\sim 30$ $\mu$K; this results in a thermal velocity $v_{\text{th}} = 54$ mm s$^{-1}$. For each of the atomic motion decoherence channels, we have used the worst case scenario for the canonical mode momentum in order to put a realistic upper limit on the broadening: $l = 0$, except for the term $|T\rangle$; broadening, which scales linearly with $l$; for this term, $l = 30$ was used. The number of atoms $N_0$ was calculated by $N_0 = \rho \omega_{\text{laser}}^2 / \omega_{\text{nl}}$. We assumed a thin atomic cloud, $\omega_{\text{nl}} = 5$ $\mu$m with a density $\rho = 10^{13}$ cm$^{-3}$. For these parameters, $N_0 = 30$. Overall, the most dominant effect of atomic motion is the thermal velocity coupling to external manifolds, which is a decoherence channel that is insensitive to the number of modes the resonator supports or to the number of polaritons in the mode but is suppressed by the control field. Some of the loss channels effects can be improved fairly easily; the cavity linewidth $\kappa$ can be reduced by using higher reflectivity mirrors and engineering resonator geometry, the Rydberg linewidth $\gamma_p$ can be improved by optimizing system parameters such as laser linewidths or reducing the number of shifted Rydberg atoms, the electric field broadening $\gamma_E$ can be brought down with clever use of resonator engineering and electronics. Other parameters are much harder to improve on. The light-matter coupling $G$ can be improved with a larger atomic density; a factor of $100$ increase in density $\rho$ is achievable [17]. Similarly, the control field Rabi coupling $\Omega$ can be increased by using D Rydberg states or a build-up cavity. Finally, thermal effects can be reduced by trapping the atoms in a lattice [12].

| Cavity loss $\kappa \cos^2 \frac{\gamma}{2} / 2 \pi$ | $\approx 2 \pi \times 200$ kHz |
| Rydberg loss $\gamma_p \sin^2 \frac{\gamma}{2} / 2 \pi$ | $\approx 2 \pi \times 86$ kHz |
| Dark → bright coupling $\gamma d_d b_b$ | $\approx 2 \pi \times 3.5$ kHz |
| Electric fields $\gamma_{\text{nl}} / \gamma_{\text{nl}}$ | $\approx 2 \pi \times 1.5$ kHz |
| $|dd\rangle \rightarrow |bb\rangle$ scattering $\gamma_{\text{nl}} / \gamma_{\text{nl}}$ | $\approx 2 \pi \times 0.5$ kHz |
| $|dd\rangle \rightarrow |db\rangle$ scattering | $\approx 2 \pi \times 7$ kHz |
| $|T\rangle$ broadening $\gamma_{\text{nl}} / \gamma_{\text{nl}}$ | $\approx 2 \pi \times 9$ kHz |
| $|T\rangle$ cross-thermalization $\left( \frac{1}{\sigma^2} - \frac{\alpha b E}{\sqrt{N_0}} \right) / \gamma_{\text{nl}}$ | $\approx 2 \pi \times 8$ kHz |
| $|T\rangle$ broadening $\gamma_{\text{nl}} / \gamma_{\text{nl}}$ | $\approx 2 \pi \times 27$ Hz |
| $|T\rangle$ cross-thermalization | $\approx 2 \pi \times 5$ kHz |
| $|T\rangle$ broadening $\gamma_{\text{nl}} / \gamma_{\text{nl}}$ | $\approx 2 \pi \times 200$ Hz |

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