Optical properties of an atomic ensemble coupled to a band edge of a photonic crystal waveguide

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Abstract

We study the optical properties of an ensemble of two-level atoms coupled to a 1D photonic crystal waveguide (PCW), which mediates long-range coherent dipole–dipole interactions between the atoms. We show that the long-range interactions can dramatically alter the linear and nonlinear optical behavior, as compared to a typical atomic ensemble. In particular, in the linear regime, we find that the transmission spectrum contains multiple transmission dips, whose properties we characterize. Moreover, we show how the linear spectrum may be used to infer the number of atoms present in the system, constituting an important experimental tool in a regime where techniques for conventional ensembles break down. We also show that some of the transmission dips are associated with an effective ‘two-level’ resonance that forms due to the long-range interactions. In particular, under strong global driving and appropriate conditions, we find that the atomic ensemble is only capable of absorbing and emitting single collective excitations at a time. Our results are of direct relevance to atom–PCW experiments that should soon be realizable.

1. Introduction

Photonic crystal waveguides (PCWs)—dielectric media with a periodically varying refractive index—have attracted significant interest recently as a platform for realizing novel quantum light–matter interfaces. The ability to engineer PCW properties permits control of the electromagnetic environment experienced by atoms, which may be leveraged to achieve strongly enhanced atom–photon coupling efficiencies [1–5] compared to more traditional approaches [6–8], as well as for exploring new regimes of quantum optics.

An exciting example of the latter [9–15] is the ability to engineer long-range coherent interactions between atoms. The emission properties of an atom coupled to a PCW differ markedly from those in free space when the resonant transition frequency of the atom, \( \omega_0 \), is inside a band gap, i.e. a frequency domain where guided modes are absent, as depicted in figure 1(a) for the blue-colored band. Rather than radiating away, the field emitted into these modes is localized around the atom by Bragg reflection, giving rise to an atom–photon bound state [16–18], shown in figure 1(b). The photonic component of this state constitutes an effective cavity mode that can mediate coupling to other atoms, resulting in effective dipole–dipole interactions which acquire the spatial features of the cavity mode. These interactions can in turn be exploited to realize novel regimes of quantum many-body physics and nonlinear optics [12, 14, 19].

Motivated by remarkable advances to interface atoms or solid-state emitters with PCWs [2, 4, 15, 20], we study the fundamental optical properties of a system of two-level atoms with long-range interactions, finding rich behavior that differs markedly from an ensemble of independent atoms. For example, as is evident from figure 1(c), even the linear transmission spectra can be dramatically modified compared to a typical atomic ensemble, which is characterized by a single transmission dip. We will describe the origin of the multiple transmission dips that arise due to the long-range interactions, focusing in particular on the one of highest...
energy, which corresponds to a collective symmetric spin excitation in the atoms. We show how to characterize its key features in the linear regime, and explain how its properties may be used to infer the number of atoms in the system, providing a key tool for future experiments. We then demonstrate that it can exhibit large optical nonlinearities that yield strong photon anti-bunching in the scattered light.

Intuitively, the nonlinear response is a result of anharmonicity in the spectrum of the collective excitations between different number manifolds. In particular, for an interaction range comparable to the system size, the bandgap (BG)-mediated interaction defines a set of symmetric collective excitations, whose anharmonicity scales with the strength $V$ of the atom–atom coupling. For sufficiently large values of $V$, a weak probe beam cannot efficiently populate a second collective excitation in the presence of a first excitation, thereby giving rise to an effective two-level structure. Indeed, we will show that it is possible to generate a single collective excitation deterministically under a global Rabi pulse applied to the entire atomic ensemble, with high fidelity. Interestingly, such a state constitutes a W or Dicke state, whose preparation in atomic ensembles is of active interest [21, 22]. Our results are a step towards developing a comprehensive understanding of the linear and nonlinear optical properties of such systems of interacting atoms.

2. Model and methods

We model an ensemble of two-level atoms with ground and excited states $|g\rangle$, $|e\rangle$, coupled to two guided bands of a 1D PCW, with dispersion relations (frequency $\omega$ versus Bloch wavevector $k$) as depicted schematically in figure 1(a). A realistic platform for implementing such a model is the ‘alligator’ PCW (APCW) described in [2]. The atomic transition frequency, $\omega_2$, is in a band gap of one set of modes, which in practice may have, e.g., transverse-electric (TE) polarization. As described in the introduction, the absence of propagating states at frequency $\omega_2$ prevents an atom from radiating into the TE modes. However, an exponentially decaying photonic cloud of length $l$ forms around each excited atom, facilitating coherent excitation exchange with proximal atoms in their ground state with characteristic strength $V$. The quantities $V$ and $L$ depend on the curvature of the dispersion relation at the lower band edge and the frequency separation $\delta$ between the band edge and atomic transition, as discussed further in [13] and in appendix A. We assume the detuning between $\omega_2$ and the upper TE band edge to be sufficiently large that coupling to this upper band may be neglected.

While the above mechanism produces novel long-range interactions, it does not allow the atoms to be efficiently probed using the same TE modes, since it is not possible to input propagating fields at frequency $\omega_2$. We therefore introduce a second band of modes, which may have transverse-magnetic (TM) polarization, whose own band gap is situated far from $\omega_2$, thereby allowing it to be used as a conventional waveguide to excite the atoms and collect the resulting scattered fields. Our model is then a minimal description that produces long-range interactions, and allows their effect to be probed. In particular, the level structure is considerably simplified compared to previous schemes to realize optical nonlinearities in PCWs [12, 14]. The single–atom coupling strength to the TM band is denoted $\Gamma_{1D}$ and the associated wavevector $k_a$ is shown in figure 1(a). We assume the atoms are trapped along the crystal axis in a lattice of period $d$, equal to the period of the crystal itself.

Figure 1. (a) Band structure of the PCW, showing guided-mode frequency $\omega$ versus Bloch wavevector $k$. The atomic frequency $\omega_2$ is in a band gap (gray region) of one mode (blue), and in a linear-dispersion region of a second mode (green). (b) An atom in its excited state $|e\rangle$, coupled to a PCW (in blue). An atom–photon bound state forms when $\omega_2$ is in a band gap: the photonic component (light red) associated with an excited atom mediates excitation exchange with a second atom in its ground state $|g\rangle$. (c) Blue: representative single-shot transmission spectrum for $n = 10$ atoms placed randomly over $N = 200$ sites, with $V/\Gamma^*= 4$, $L/d = 100$, $\Gamma_{1D}/\Gamma^* = 0.3$ and $k_a d = \pi/2$. Red: spectrum for the same configuration without long-range interactions ($V/\Gamma^* = 0$), i.e., a normal atomic ensemble. (d) Spectrum for the same parameters as (c), averaged over 1000 samples of atomic positions.
The dynamics of the full system may be described by an effective non-Hermitian Hamiltonian for the atoms alone [13, 23]:

$$
H = -\sum_{j} \left[ \left( \Delta + i\Gamma' / 2 \right) \sigma_{j}^{\dagger} + \Omega \sigma_{j} e^{i(kz - \omega_{j} t)} \right] + \sum_{j,k} \left( V(-1)^{j} e^{-|z_{j} - z_{k}|/L} - \frac{i\Gamma_{1D}}{2} e^{ik|z_{j} - z_{k}|} \right) \sigma_{j}^{\dagger} \sigma_{k}^{\dagger}.
$$

(1)

Here $\Delta = \omega_{L} - \omega_{j}$ denotes the detuning of the probe field (incident from the left with wavevector $k_{L}$, frequency $\omega_{L}$, and Rabi frequency $\Omega$) from the atomic resonance frequency, $\Gamma'$ is the decay rate into free space and all other modes, and the operator $\sigma_{j}^{\dagger} = |\mu\rangle \langle \nu|$, where $\mu, \nu = g, e$ are energy eigenstates of atom $j$, which has position $z_{j}$. The TE mode has Bloch wavefunction $E(z) \sim \cos(kz)$, and we assume the atoms are trapped at its anti-nodes, which maximizes the interaction, and results in the integer phase factor $\theta_{jk} = (z_{j} + z_{k}) / d$, where $z_{j} / d = \text{integer}$. In all simulations we set $\Gamma_{1D} / \Gamma' = 0.3$, however our conclusions will be independent of specific parameter choices.

The BG interaction term (proportional to $V \rho_{g}^{\dagger}$) may be diagonalized in the single excitation manifold to give a set of $n$ collective resonances. When the strength and range of the interactions are sufficiently large, one of the modes is shifted considerably further from the bare single-atom resonance frequency than any of the other $n - 1$ modes. This is most evident in the limit $L / d \rightarrow \infty$, when diagonalization gives a single collective mode whose energy is shifted by $nV$, and $n - 1$ modes whose energies are unshifted. In section 3 we show how the properties of this maximally shifted resonance may be characterized, while in section 4 we show that the system exhibits strong optical nonlinearities when driven at the frequency of this highest resonance.

To obtain the fields scattered into the waveguide by the atoms, we time-evolve the initial atomic wavevector $k_{j}$ field correlation functions is provided in [24]. To preserve the norm, the dynamics must in principle be supplemented with quantum jumps, however we will primarily be interested in field correlations under weak driving ($\Omega / \Gamma' \ll 1$), where—as we show in appendix B—quantum jumps may be neglected. The transmitted ($T$) and reflected ($R$) fields are then reconstructed using the input–output relations [24]

$$
\begin{align*}
    a_{\text{out},T}(z) &= \Omega e^{ikz} + \frac{i\Gamma_{1D}}{2} \sum_{j} \sigma_{j}^{\dagger} e^{ikz_{j}} + F_{T}, \\
    a_{\text{out},R}(z) &= \frac{i\Gamma_{1D}}{2} \sum_{j} \sigma_{j}^{\dagger} e^{-ikz_{j}} + F_{R},
\end{align*}
$$

(2)

(3)

where the transmitted (reflected) field is evaluated at position $z$ beyond the last (first) atom (henceforth we drop the subscript out and the argument $z$). Physically, equations (2) and (3) state that the output field properties are completely encoded in the input field and the correlations of the atomic scatterers. The vacuum input noise operators $F_{T,R}$ may be neglected for our correlations of interest. The steady-state transmittance $T$ and two-photon correlation function $g_{\text{eg}}^{(2)}(0)$ of the transmitted field are given by

$$
T = \frac{\langle \psi | a_{j}^{\dagger} a_{j}^{\dagger} a_{j} a_{j} | \psi \rangle}{\Omega^{2}},
$$

(4)

$$
\begin{align*}
    g_{\text{eg}}^{(2)}(0) &= \frac{\langle \psi | a_{j}^{\dagger} a_{j}^{\dagger} a_{j} a_{j} | \psi \rangle}{\langle \psi | a_{j}^{\dagger} a_{j} | \psi \rangle^{2}},
\end{align*}
$$

(5)

and similarly for the reflected field, where $|\psi\rangle$ is the steady-state wavevector. In the cases where we consider stronger driving fields in section 4, we solve the full atomic density matrix equations, thus accounting for quantum jumps. Further discussion of our methods for computing field correlation functions is provided in appendix B.

### 3. Linear optical properties

#### 3.1. Transmission spectrum

Figure 1(c) shows a representative single-shot (atoms fixed in position) linear transmission spectrum for one configuration of $n = 10$ atoms randomly placed in a lattice of $N = 200$ sites. We choose the lattice constant $d$ such that $k_{d}d = \pi / 2$, which for unit filling factors (i.e. for $n / N = 1$) minimizes the probability of reflection from the array, and thus most closely corresponds to a free-space atomic ensemble. However, different choices of $k_{d}d$ (excluding those very close to integer multiples of $\pi$, where cooperative emission from the atoms into the TM modes becomes significant) do not qualitatively change the results.

For the case $V = 0$ (red curve in figure 1(c)), the transmission spectrum is identical to that of a free-space ensemble, i.e. a broadened Lorentzian centered at the single-atom resonance frequency [25]. The resonant
atom, with all other atoms in their ground state, and $G' = n$ and length $t$ scales linearly with the number of atoms $n$, with a slope proportional to $1/N$, as shown in figure 2. This represents a highly non-standard situation in quantum optics, and acquires additional richness when fields can freely propagate and is unaffected by the cavity mirrors, or to running an optical nano
deld through a separate TM waveguide mode.

To understand the structure of the spectrum, note that in the limit $L/d \to \infty$, the BG interaction term in equation (1) is described in the one-excitation manifold by an $n \times n$ matrix with entries $\pm V$, with the phase of a given element determined by the corresponding atomic positions. This matrix has $n - 1$ degenerate resonances at the bare single-atom energy, and one resonance of energy $\omega_{\text{max}} = nV$, whose associated collective excitation we denote $|\phi_{\text{max}}\rangle$. In the limit $L/d \to \infty$, it is easy to show that $|\phi_{\text{max}}\rangle = (1/\sqrt{n})\sum_j(-1)^j|\epsilon_j\rangle$, where $|\epsilon_j\rangle$ denotes the presence of an excitation in the $j$th atom, with all other atoms in their ground state, and $\epsilon_j = z_j/d$.

We note here that for the case $L/d \to \infty$, the band gap interaction is identical in form to the effective interaction between atoms coupled to a single-mode detuned cavity, such that the cavity mode can be effectively eliminated [26]. Even in this limit, however, the spectra and optical properties that we describe here are qualitatively different. In particular, a cavity is conventionally probed via the cavity output, which is directly related to the intra-cavity properties via the cavity input--output relation [27]. Here, we instead consider the effect of the band gap (or ‘cavity’) interaction on the field propagation in a separate TM waveguide mode.

Conceptually, this would be similar to probing an atomic gas coupled to a cavity, but along an axis where the field can freely propagate and is unaffected by the cavity mirrors, or to running an optical nanofiber through a cavity. This represents a highly non-standard situation in quantum optics, and acquires additional richness beyond single-mode cavity physics due to the continuum associated with propagating fields. In different contexts, such a configuration is known to enable novel phenomena such as vacuum induced transparency [28, 29], where pulses propagate with a photon number-dependent group velocity, or in correlated transparency windows, where only pulses of a given number and shape propagate through the medium [14].

For finite-range band gap interactions, the $(n - 1)$-fold degeneracy at the bare single-atom transition frequency is lifted to yield additional observable resonances, as shown in figure 1(c), while the maximum resonance energy, $\omega_{\text{max}}$, is correspondingly reduced. However, as we explain in this section, the key properties of this maximum resonance can still be well quantified.

Current techniques for interfacing atoms with PCWs do not allow precise control of the atomic positions $z_j$, and moreover, thus far experiments do not allow for single-shot spectroscopy. We therefore seek the average optical properties, taking the mean of the appropriate quantities over many random spatial configurations. In this section, we focus on the idealized case where the number of atoms $n$ is identical in each single-shot realization, and only the atomic positions change. While this assumption may not necessarily correspond to a realistic experimental scenario, it will nonetheless allow us to develop a comprehensive picture of the key facets of the system’s optical properties. In section 3.3, we consider the more realistic case where only the average number of atoms $\langle n \rangle$ remains constant.

Figure 1(d) shows the average linear transmission spectrum for the same conditions as figure 1(c), taken over 1000 single-shot realizations. While each of these has a unique maximum resonance frequency $\omega_{\text{max}}$, we find that for $L/d \gg 1$ the average energy of the highest single-excitation resonance $\omega_{\text{max}}$ scales linearly with the number of atoms $n$ in the system, with a slope proportional to $L/Nd$, as shown in figure 2(a). This observation motivates the definition of an effective interaction strength $V_{\text{eff}}(L)$, through the equation

$$\omega_{\text{max}}(n, L) = V + (n - 1)V_{\text{eff}}(L).$$
Here, the first factor of \( V \) is the energy of a single atom (the Stark shift it experiences due to its own bound photon), and its average interaction energy with each subsequent atom is \( V_{\text{eff}}(L) \). We now give a simple model for estimating \( V_{\text{eff}} \) for a photonic crystal of given \( V,L \), and a lattice of \( N \) sites separated by distance \( d \). The band-gap interaction Hamiltonian is

\[
H_{\text{BG}} = V \sum_{j,k} e^{-|z_j-z_k|/\mu_f} \sigma_j^x \sigma_k^x
\]

where, for simplicity, we neglect the phase factor resulting from the product of Bloch wavefunctions. When the scaling of \( \sigma_{\text{max}} \) is linear in \( n \), the coupling between distant atoms can become negligible, and consequently the total energy can scale sub-linearly in the number of atoms. Similarly, for \( L \) the validity is also limited to low densities \( (n \ll N) \), where all atoms have approximately the same excited state amplitude \( 1/\sqrt{n} \).

It follows from the above discussion that provided the conditions \( L \gg d \) and \( n \ll N \) are satisfied, which encompasses a wide range of regimes of interest, equations (6) and (8) may be used to determine \( \sigma_{\text{max}} \) for a PCW with known \( V,L,N \), and \( d \). In principle, this then allows the number of atoms \( n \) in the ensemble to be inferred from the frequency of the highest-energy transmission minima. This would constitute an important system characterization tool, given that existing techniques based on measuring the resonant optical depth break down in the presence of BG interactions. However, our analysis has thus far assumed the number of atoms to be identical in each single-shot realization, a requirement that in practice would itself presumably require prior knowledge of \( n \) by other means. We return to this question in section 3.3, where we show that, even when the number of atoms in a given realization is non-deterministic, the average spectra can nonetheless yield information on the mean number of atoms in the system.

### 3.2. Transmittance at highest-energy resonance and a simple effective model

Another important feature of the maximum resonance to characterize is the associated transmittance \( T_{\text{dip}} \), as illustrated in figure 1(d). Intuitively, this should be a function of the probability of creating the corresponding collective excitation \( |\phi_{\text{max}}\rangle \), i.e. the maximum-energy single-excitation resonance of \( H_{\text{BG}} \). This probability is, in turn, proportional to the overlap \( \langle \phi_{\text{max}} | \psi_{\text{in}} \rangle \), where \( |\psi_{\text{in}}\rangle \) is the normalized state to which the input TM probe field naturally couples. This state is dictated by the relative phase with which the probe field drives the atoms (see equation (1)), and is given by \( |\psi_{\text{in}}\rangle = \frac{1}{\sqrt{n}} \sum_k e^{i\theta_j} |e_k\rangle \), where \( |e_j\rangle \) denotes an excitation at the \( j \)th atom, with all others in the ground state. For the choice \( k_1d = \pi/2 \) this gives \( |\psi_{\text{in}}\rangle = \frac{1}{\sqrt{N}} \sum_k (i)^k |e_k\rangle \), with \( \theta_j = z_j/d \).

Meanwhile, in the limit \( L \to \infty \), the maximum-energy resonance of the BG interaction has the form \( |\phi_{\text{max}}\rangle = \frac{1}{\sqrt{n}} \sum_j (-1)^j |e_j\rangle \). The overlap of the two states is then

\[
\langle \phi_{\text{max}} | \psi_{\text{in}} \rangle = \left( \frac{1}{\sqrt{N}} \sum_i^n (i)^k |e_i\rangle \right) \left( \frac{1}{\sqrt{n}} \sum_j^n (-1)^j |e_j\rangle \right) = \frac{1}{n} \sum_{i,j} (-1)^{i-j}
\]

Since the atomic positions are non-deterministic for any given realization, the latter equality shows that the overlap is proportional to a sum of random phases along the real and imaginary directions. As we are interested in the mean value \( \langle \phi_{\text{max}} | \psi_{\text{in}} \rangle \), we may draw analogy to a 2D random walk to conclude that statistically the magnitude of the mean overlap, which we denote \( \sqrt{N}\text{eff} \), scales as \( 1/\sqrt{2} n \). Only in the special case \( k_1d \approx \pi \) does the summation of phases become coherent, where the \( n \) dependence cancels and \( \sqrt{N}\text{eff} \to 1 \).

The above reasoning breaks down as the number of atoms approaches the total number of lattice sites, since then the sum contains an equal number of terms of opposite phases and thus tends to zero. For example, for \( n = N = 200 \), there are 50 terms of each of the phases 1, i, -1, -i, which when summed give a vanishing overlap. In figure 3(a) we show how the simple formula \( \sqrt{N}\text{eff} = \sqrt{(N-n)/\sqrt{2} nN} \) agrees very well with the numerically obtained scaling for all \( n \).

While the above analysis assumes an infinite-range interaction, in figure 4 we show how the mean overlap \( \sqrt{N}\text{eff} \) scales with \( n \) for a lattice of \( N = 200 \) sites and several finite interaction lengths, where the eigenstate \( |\phi_{\text{max}}\rangle \)
is obtained by numerical diagonalization. It can be seen that the same scaling behavior holds for interactions
with \( L \leq 1 \); below this limit, the BG interaction is no longer dominant and thus no longer determines the
maximum-energy resonance.

As a result of the decrease in the overlap \( k_n \) of the input
field and the maximum-energy resonance, the transmittance
\( T_{\text{dip}} \) increases as a function of \( n \), as shown in
figure 3(b) (dots). In order to characterize quantitatively this effect, we now construct a simplified effective model for the mean properties of
the system around the maximum resonance, valid in the regime
\( L \leq 1 \), which brings together the key
features from the linear regime that we have explained above. Since we are interested in driving the state \( \phi_{\text{max}} \)
close to its resonance frequency, it is convenient to define a shifted laser detuning
\( \Delta_{\text{max}} = \omega_L - \bar{\omega}_{\text{max}} \) relative to
the resonance of this collective state. Our model then assumes that the system has only two relevant degrees of
freedom: (1) an ensemble of \( n \) independent atoms at the frequency
\( \Delta_{\text{max}} \), and; (2) the maximum-energy single-photon excitation \( \phi_{\text{max}} \), at frequency \( \Delta_{\text{max}} = 0 \).

In our regime of interest, the independent ensemble—which we will refer to as the ‘background’ atomic ensemble—will be driven far off resonance by the TM probe field, and provides a small attenuation of the incident field on top of the larger transmission dip arising from the collective state. Far off resonance, an ensemble has a
transmission coefficient for the probe field of approximately
\( t_n^0 \), where \( t_n = (\Delta + i\Gamma' / 2) / (\Delta + i\Gamma / 2) \) is the
transmission coefficient of a single atom [23]. Since the atoms are acting independently, for low attenuation the
contribution of the ensemble to the output field equation can be written as

\[
a_T = \Omega + \frac{1}{2} \sum_{j=1}^{n} \sigma_{j\sigma}^{\prime}
\]

where \( \sigma_{j\sigma}^{\prime} \) denotes the response of a single atom to the (far-detuned) field. Importantly, the background
ensemble is then effectively captured by \( n \) identical copies of a single degree of freedom.

The second degree of freedom in our effective model is the maximum-energy single-photon excitation
\( \phi_{\text{max}} \), which is driven close to its resonance frequency situated at \( \Delta_{\text{max}} = 0 \). For later convenience when we
move on to consider the nonlinear properties of this resonance (section 4), here we shall use the Fock state
notation \( |1\rangle \) to denote \( |\phi_{\text{max}}\rangle \). In order to determine the coupling of the probe field to this state, we note that the
TM probe first excites the state \( |\psi_{\text{in}}\rangle \), defined above, with matrix element \( \sim \sqrt{n} \), while the subsequent overlap
with the state \( |1\rangle \) is given by \( \sim \sqrt{k_n} \), as described in the previous section. Overall, therefore, the matrix element

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![Figure 3](image3.png)

**Figure 3.** (a) Mean overlap \( \sqrt{k_n} \) (blue) of initial excitation and maximum-energy resonance versus \( n \), for \( L/d = 10^6 \) and \( k_d = \pi / 2 \); red line shows a simple fit. (b) \( T_{\text{dip}} \) (see figure 1(d)) versus \( n \) for \( L/d = 10^6 \) (blue) and \( L/d = 50 \) (black) for the full (equation (1)) and effective (equation (11)) models.

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![Figure 4](image4.png)

**Figure 4.** Mean overlap \( \sqrt{k_n} = |\langle \phi_{\text{max}} | \psi_{\text{in}} \rangle| \) for different interaction lengths, and a lattice of \( N = 200 \) sites with \( V / \Gamma = 4 \), \( \Gamma_{\text{in}} / \Gamma = 0.3 \).

---

\[ a_T = \Omega + \frac{1}{2} \sum_{j=1}^{n} \sigma_{j\sigma}^{\prime} \]
coupling the input field to the state \( |1\rangle \) is \( \sqrt{n}\hbar \kappa \Omega \). By similar arguments, we take the total decay rate of this mode to be \( \gamma_e = \Gamma' + n\kappa_0\Gamma_D \).

The level scheme for our effective model, along with the couplings and decay rates for the two degrees of freedom, is shown in figure 5(a). For consistency with the extension of the model to the nonlinear regime in section 4, we take the background ensemble and collective states to be fully independent of each other. In particular, each has its own ground state, which we denote \( |g_b\rangle \) and \( |g_c\rangle \) respectively. The corresponding Hamiltonian is \( H_{\text{eff}} = H_B + H_C \), where the background \((B)\) and collective \((C)\) components are

\[
H_B = -\left( \Delta_{\text{max}} + n V_{\text{eff}}(L) + \frac{\Omega^2}{2} \right) |\sigma_{ee}\rangle \langle \sigma_{ee}| + \hbar c.
\]

\[
H_C = -\left( \Delta_{\text{max}} + \eta + \frac{\Gamma_0}{2} \right) |\sigma_{11}\rangle \langle \sigma_{11}| - \sqrt{n}\hbar \kappa \Omega (\sigma_{eL} + \hbar c),
\]

where \( |e\rangle \) refers to the excited state of the background atom. We emphasize that we do not rigorously derive the effective Hamiltonian from the full Hamiltonian \((1)\), but rather it represents a minimal model meant to capture the most important characteristics of the system around the maximum resonance frequency \( \omega_{\text{max}} \). In order to account for the variations in \( \omega_{\text{max}} \) for finite \( L \), we introduce a random number \( \eta \) to our model, over which we take many samples, and whose statistics we determine by numerical analysis of the spread of \( \omega_{\text{max}} \). In all cases, the mean \( \bar{\eta} = 0 \), since the average frequency of the maximum resonance is accounted for through the quantity \( V_{\text{eff}}(L) \).

The full output operator for the transmitted field comprises contributions from the input field, the background ensemble of \( n \) atoms, and the collective excitation \( |1\rangle \):

\[
a_T = \Omega + \frac{i\Gamma_D}{2} \sum_{j=1}^n |\sigma_{eL,j}\rangle \langle \sigma_{eL,j}| + i\sqrt{n}\hbar \kappa \Omega (|\sigma_{eL,1}\rangle \langle \sigma_{eL,1}| + F_T).
\]

(12)

Again, the noise operator \( F_T \) here is not relevant for our discussion. We shall demonstrate the utility of our effective model for both finite and infinite interaction lengths. In the latter case, \( L/d \to \infty \), the frequency of the maximum resonance is independent of the atomic positions, whereas in the former case, \( 1 \ll L/d \ll \infty \), each configuration has a different value of \( \omega_{\text{max}} \). As a first observation, it follows from this fact that when we average over many spectra, for any given frequency one finds less attenuation (higher transmittance) in the finite \( L \) case than for infinite \( L \). For the case \( N = 200 \), \( L/d = 50 \) presented here, we find the spread of the maximum resonance frequency to be well approximated by a Gaussian distribution, so that for given values of \( L \) and \( n \) we can obtain the standard deviation \( \sigma \) (\( n, L \)), thus giving the required statistics of the parameter \( \eta \).

Figure 3(b) (crosses) shows the effective model predictions for \( T_{\text{dip}} \), taking 1000 samples of different \( \eta \) in the case \( L/d = 50 \), demonstrating that the model reproduces well the results obtained using the full system Hamiltonian \((1)\) and input–output operators \((2)\) and \((3)\) for both values of \( L \) considered. In the case \( L \to \infty \), we can also use the effective model to analytically obtain a simple formula for the transmittance \( T_{\text{dip}} \). Assuming \( \sqrt{n} V_{\text{eff}} \gg \sqrt{\Gamma_D} \), the background ensemble may be neglected and the problem is reduced to that of a two-level system comprising the levels \( |g_b\rangle, |1\rangle \). Solving the corresponding optical Bloch equations in the steady state \((SS)\), we find that the transmittance is given by

![Figure 5](image-url)
through a single atom coupled to the system, and in particular if it may be used to infer the mean number of atoms.

We assume that each atom is trapped independently of the others, in which case the variable \( n \) may be inferred from the frequency of the maximum-energy transmission dip, providing a key tool for system characterization.

\[
\tilde{T}_{\text{dip}} = \frac{\langle a_1 ^+ a_1 \rangle_{SS}}{\Omega^2} = \frac{\Gamma'^2}{(\Gamma' + n \kappa_{1D})^2},
\]

which is analogous to the resonant transmission \( T = \Gamma'^2/(\Gamma' + \Gamma_{1D})^2 \) through a single atom coupled to the probe band alone [23].

### 3.3. Transmission spectrum for random number of atoms

The analyses of the preceding sections assume that the number of atoms \( n \) in each single-shot experiment is constant, with only the atomic positions being random in each case. We have shown how under these conditions the average spectrum may be used to infer \( n \) from the frequency of the highest-energy transmission dip.

However, a more realistic experimental scenario is that the mean number of atoms \( \langle n \rangle \) in the system is constant, with the actual number of atoms \( m \) present in any given single-shot realization being a random parameter. An important question is then whether the average transmission spectrum still serves as a characterization tool for the system, and in particular if it may be used to infer the mean number of atoms \( \langle n \rangle \).

We assume that each atom is trapped independently of the others, in which case the variable \( m \) will be Poisson-distributed about the mean value \( \langle n \rangle \). In each single-shot sample we draw a random number of atoms \( m \) from the corresponding Poisson distribution, generate a random set of positions for these atoms, and then compute the transmission spectrum. Focusing on the region around the maximum resonance, we first consider the limit in which the interaction length \( L \) far exceeds the length of the system, in which case, as we have described above, the maximum resonance frequency corresponding to each \( m \) is simply \( \approx m V \). For sufficiently large \( V \), the transmission dips for each \( m \) are then clearly resolvable and separated by \( V \), as we show in figure 6(a) for the cases \( \langle n \rangle = 4 \) and \( \langle n \rangle = 10 \), with \( L/d = 10^4 \) and \( V/\Gamma' = 4 \). The number of atoms can then be inferred from the combination of the number of attenuation dips and their relative depths.

When the interaction length \( L \) is of the order of the system size, the dips corresponding to different numbers of atoms \( m \) are instead separated by \( V_{\text{eff}}(L) \), and it may no longer be possible to resolve the individual features.

Provided that \( \langle n \rangle \) is sufficiently large, one instead observes a broad dip, as we show in figure 6(b) for three representative cases, with \( L/d = N = 200 \). Analogous to the analysis of section 3.1, where we denoted the frequency of the dip as \( \omega_{\text{max}} \), here we shall denote the dip location \( \omega_{\text{max}} \), reflecting the average over both atom number and atomic positions. In figure 6(c) (solid dots) we plot \( \omega_{\text{max}} \) versus \( \langle n \rangle \) for three different interaction lengths. Similar to the case considered in section 3.1, the dip location scales linearly in the mean number of atoms in the system. For comparison with the previous analysis, figure 6(c) also shows the frequency \( \omega_{\text{max}} \) (crosses) computed in the case where the atom number is fixed at \( \langle n \rangle \) in all single-shot cases. It is apparent that \( \omega_{\text{max}} \approx \omega_{\text{max}} \), so that the conclusions of section 3.1 remain valid even when only the mean number of atoms is fixed. In particular, \( \langle n \rangle \) may be inferred from the frequency of the maximum-energy transmission dip, providing a key tool for system characterization.

![Figure 6](image-url)

Figure 6. (a) Transmittance spectra for the cases \( \langle n \rangle = 4 \) and \( \langle n \rangle = 10 \) for an interaction length \( L/d = 10^{0} \). The two curves are obtained by averaging the spectra of many single-shot realizations, where each realization consists of a random number of atoms \( m \) and a random set of positions thereof. (b) Representative transmission spectra around the maximum resonance frequency, for the mean atom numbers \( \langle n \rangle \) indicated, and \( L/d = 200 \). (c) Location \( \omega_{\text{max}} \) of the maximum resonance dip as a function of \( \langle n \rangle \) (solid dots) for different interaction lengths \( L/d \). The crosses show the corresponding dip location \( \omega_{\text{max}} \) for the case where \( n \) is constant in all realizations, as in figure 6(a). In all three figures, \( V/\Gamma' = 4 \), \( N = 200 \), \( \Gamma_{1D}/\Gamma' = 0.3 \), \( \Omega/\Gamma' = 0.01 \), and the averaging is performed over 30 000 single-shot realizations.
4. Nonlinear optical properties

4.1. Photon statistics of scattered fields

We now consider the nonlinear optical properties of the system, beginning with the second-order correlation function of the reflected field, $g^{(2)}_R(0)$, when the system is driven at its maximum single-excitation energy $\omega_{\text{max}}$. The reflected field arises purely due to scattering from the ensemble, whereas the transmitted field results from interference between the forward-scattered and incident fields (equation (2)). Thus, $g^{(2)}_R(0)$ yields information on the system’s ability to scatter two photons simultaneously. The limiting case of a single two-level atom produces perfect anti-bunching, $g^{(2)}_R(0) = 0$, indicating that it can only absorb and re-emit single excitations at a time.

For a given value of $V$, using equation (1) we compute the correlation function $g^{(2)}_R(0)$ for each of 1000 random samples of atomic positions, keeping the number of atoms $n$ fixed in all cases. In each realization, the system is weakly driven at a frequency corresponding to the largest eigenvalue $\omega_{\text{max}}$ of the Hamiltonian in the single-excitation manifold. While this situation may not necessarily be realistic experimentally, since it would require knowledge of $\omega_{\text{max}}$ for a single shot, the results are nonetheless elucidating. The number of occurrences of each value of $g^{(2)}_R(0)$ from the sample set is shown in figure 7(a): evidently, the stronger the interaction, the more prevalent strong anti-bunching becomes. In particular, as figure 7(b) shows, for $V/V' = 6$ the proportion of configurations with $g^{(2)}_R(0) < 0.1$ approaches 90%, suggesting that in the vicinity of the maximum resonance frequency $\omega_{\text{max}}$ the ensemble typically behaves as an effective two-level system. The origin of the observed antibunching may easily be understood in the $L \rightarrow \infty$ limit: diagonalizing the BG interaction, we find the maximum single-photon eigenenergy to be $\omega_{\text{max}}^{(1)} = nV$, and the maximum two-photon eigenenergy to be $\omega_{\text{max}}^{(2)} = 2(n - 1)V$. Defining the anharmonicity $A = \omega_{\text{max}}^{(2)} - 2\omega_{\text{max}}^{(1)}$, we find $A(L = \infty) = -2V$.

We now consider the more relevant experimental scenario where only the mean number of atoms $\langle n \rangle$ in the system is known in advance. We recall from section 3.3 that, when $\langle n \rangle$ is sufficiently large, the transmission dip corresponding to the maximum resonance is approximately centered on the frequency $\omega_{\text{max}}$ given by equation (6). One may then adopt the strategy of driving the system at this average frequency. Intuitively, since the average and the actual single-shot resonance frequencies will in general not coincide, the prevalence of the strong anti-bunching observed in figure 7(b) should be reduced.

We investigate this question in figure 7(c), where for the case $V/V' = 6$ we compute the photon correlation function $g^{(2)}_R(0)$ for different mean numbers of atoms $\langle n \rangle$, with the driving frequency taken to be the
corresponding average $\bar{\omega}_m(n)$. For each $\langle n \rangle$ we take 1000 single-shot samples, in each case drawing a number of atoms $m$ from the corresponding Poisson distribution, then generating a random set of positions for the atoms. In the figure, we plot the number of configurations with a value of $g^{(2)}_R(0) < \bar{\omega}$, for the values $x = 0.1$, 0.3, and 0.8. The downward trend in each case may be explained by the fact that, for finite $L/d$ and $n < N/2$ as we have considered, the number of possible configurations is an increasing function of $n$, resulting in a larger spread in the maximum resonance frequency, so that on average the drive is more off-resonant with respect to the single-shot frequency $\omega_{\text{max}}$. Clearly, the anti-bunching is typically not as strong as we observed in figure 7(b), however there can still be a significant fraction of configurations that give rise to sub-Poissonian statistics in the reflected field. For instance, in almost the entire range of $\langle n \rangle$ considered, around 50% of the configurations have $g^{(2)}_R(0) < 0.8$.

Returning now to the subject of the anharmonicity $\Lambda$, recall that for $L \gg d$ and $n \ll N$, as discussed in section 3, we find that $\omega^{(1)}_{\text{max}}$ is characterized by an effective interaction energy $V_{\text{eff}}(L)$. It is then natural to ask whether in the finite-range interaction regime, the anharmonicity can also be characterized by $V_{\text{eff}}(L)$. We investigate this question numerically, via diagonalization of the system within the two-excitation manifold. In figure 8(a), we plot the mean anharmonicity $\Lambda$ (solid dots, with averaging over 1000 samples of atomic positions), normalized to the interaction strength $V_{\text{eff}}(L)$, as a function of $L$ for two different system sizes, $N = 50$ and $N = 200$, with $n = 20$ atoms. In addition, we plot (dashed lines) a hypothesized anharmonicity $-2V_{\text{eff}}(L)$. In both cases we see that for large $L/d \gtrsim N/2$, the approximation based on $V_{\text{eff}}(L)$ is very accurate, while for small $L$ there is a significant difference between the two models, which clearly increases for larger systems.

It is interesting to note that the anharmonicity deviates from $-2V_{\text{eff}}(L)$ for $L/d \lesssim N$, despite the fact that equation (8) for $V_{\text{eff}}(L)$ works well to describe the maximum resonance in the linear regime, even for $L/d \ll N$. We believe that for $L/d \lesssim N$, the anharmonicity is reduced as there are an increased number of states that have resonant frequencies between the bare atomic frequency and the maximum resonance, which allow for less of an energy cost to produce a second excitation. This is in contrast to the limit of infinite-range interactions, where only a single collective state with large shift exists.

A second natural question is whether the same strong anti-bunching can be observed when the system is instead driven at the frequencies of other resonances in the single-photon manifold. To investigate this, we diagonalize the interaction Hamiltonian $H_{\text{BG}}$ in the single-excitation manifold to obtain the full set of (in general non-degenerate) resonances, which we index by $q = 1, \ldots, n$ from highest to lowest energy (i.e. $q = 1$ is the maximum-energy eigenstate). We then weakly drive the system at each frequency $\omega_q$, and compute the correlation function $g^{(2)}_R(q)$. Again, we consider 1000 samples of atomic positions, for fixed atom number $n = 20$, and in figure 8(b) show a histogram of the occurrences of given values of $g^{(2)}_R(q)$ (along the $y$-axis) for each $q$.

Evidently, only the $q = 1$ resonance gives rise to strong anti-bunching in a large proportion of cases. This result is easily understood in the conceptual limit $L/d \to \infty$, when only a single eigenstate of non-zero energy results from the band-gap interactions. For finite-range interactions, it is possible for a given parameter set that other resonances also display nonlinear behavior, however as we will now argue, the maximum-energy resonance is particularly well-suited for observing optical nonlinearities.

In the single-excitation manifold, the total trace of the Hamiltonian $H_{\text{BG}}$ is simply the sum of the Stark shifts on each atom due to the interaction with their individual effective cavity modes, $\text{Tr}(H_{\text{BG}}) = nV$. Meanwhile, for finite-range interactions we have shown that the average energy of the maximum resonance is given by equation (6), from which it follows that the average energy $\bar{\omega}_{\text{res}}$ of the remaining $n-1$ resonances is $(n-1)(V - V_{\text{eff}}(L))$. Choosing, for example, the representative case where $L/d = N/2$, we find that

---

**Figure 8.** (a) Mean anharmonicity $\Lambda = \frac{\omega^{(1)}_{\text{max}} - 2\omega^{(1)}_{\text{res}}}{L_{\text{max}}}$ from direct diagonalization (dots, 1000 samples per $L$), and from the formula $\Lambda = -2V_{\text{eff}}(L)$ (dashed lines), both normalized to $V_{\text{eff}}$ as a function of $L$ for $N = 200$ (blue) and $N = 50$ (black), with $n = 20$ in both cases. (b) Colormap histogram (from 1000 samples of atomic positions) of $g^{(2)}_R(q)$ for driving at the different resonance frequencies of the single-photon manifold (q labels these resonances). Other parameters: $V/V' = 4$, number of atoms $n$ (sites) $N = 20$ (200), $L/d = 100$. 
is given by $G = G(W) \propto p$ as predicted by the effective model, for the same conditions as achieved in time, as a function of $D$ versus anharmonicity $\omega$.

Enabling the generation of only Gaussian quantum states in conventional atomic ensembles involve probabilistic or measurement-based schemes [33, 34]. Consequently, techniques for preparing non-Gaussian states in conventional atomic ensembles involve probabilistic or measurement-based schemes [33, 34], whereas in our system the strong nonlinearities derived from the BG interaction allow the deterministic generation of Fock states under strong external driving. This mechanism is then reminiscent of previous proposals to exploit Rydberg nonlinearities for the generation of collective atomic excitations [35, 36].

We now study the dynamics under equation (1) with a driving field $\Omega$ that is sufficiently large to produce Rabi oscillations, solving the master equation for the atomic density matrix when the system is initialized in its ground state $|g\rangle^{\text{opt}}$. Figure 9(a) shows a representative sample of the time evolution of the total single-excitation manifold probability, $p_1$, for an ensemble of 6 atoms driven at its maximum resonance frequency with different $\Omega$, and $V/\Gamma' = 10$.

Clearly, depending on the driving strength, $p_1$ can be very large. Intuitively, to maximize $p_1$, $\Omega$ must be sufficiently large to overcome dissipation, yet sufficiently small to minimize subsequent population of the doubly excited manifold, either through exciting the maximum-energy, doubly excited resonance, or through off-resonant excitation of the ‘background’ ensemble of modes of lower energy. We observe this ‘dissipation versus anharmonicity’ trade-off in figure 9(a), where $\Omega/\Gamma' = 5$ gives a larger maximum value of $p_1$ than the cases $\Omega/\Gamma' = 1$ and $\Omega/\Gamma' = 10$. In general, for a given set of system parameters, there will be some optimal choice of Rabi frequency and laser detuning that maximizes the probability of creating a single excitation.

Figure 9(b) shows the maximum value of $p_1$ (denoted $p_1^{\text{max}}$) during the evolution time, as a function of both $\Omega$ and the detuning $\Delta_{\text{max}} = \omega_1 - \omega_{\text{max}}$. Since the anharmonicity is negative, to minimize the probability of two-photon absorption the optimal choice of $\Delta_{\text{max}}$ is positive. The optimal probability $p_1^{\text{opt}} \approx 0.81$ occurs for the parameters $\Omega^{\text{opt}}/\Gamma' = 6.75$, $\Delta_{\text{max}}^{\text{opt}}/\Gamma' = 0.4$.

$\Xi_{\text{rem}} \approx 0.3(n - 1)V$, which in particular places an upper bound on the average energy of the second highest resonance. At the same time, the average energy of the highest resonance is more than twice as large, $\Xi_{\text{max}} \approx V + 0.7(n - 1)V$, demonstrating why it is particularly interesting for few-photon nonlinear optics.

4.2. Effective two-level dynamics in the multi-photon regime

Motivated by the observation that the fields scattered by the system can be strongly anti-bunched when the system is driven close to the frequency $\omega_{\text{max}}$, an interesting question is whether a global Rabi pulse of frequency $\approx \omega_{\text{max}}$ applied to the whole ensemble can produce only a single excitation in the atoms, as well as the associated fidelity for doing so as a function of the various system and laser parameters. Before addressing this question, we note that the eigenstate corresponding to the maximum resonance frequency, which for $k_s d = \pi/2$ and $L \to \infty$ is given by

$$|\phi_{\text{max}}\rangle = \frac{1}{\sqrt{n}} \sum_j (-1)^j |e_j\rangle,$$

has the form of a Dicke or W state, which is of considerable interest for quantum information processing and metrology [30, 31]. Conventional atomic ensembles are typically highly linear, and when strongly driven they enable the generation of only Gaussian quantum states [32]. Consequently, techniques for preparing non-Gaussian states in conventional atomic ensembles involve probabilistic or measurement-based schemes [33, 34], whereas in our system the strong nonlinearities derived from the BG interaction allow the deterministic generation of Fock states under strong external driving. This mechanism is then reminiscent of previous proposals to exploit Rydberg nonlinearities for the generation of collective atomic excitations [35, 36].

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Figure 9(b) shows the maximum value of $p_1$ (denoted $p_1^{\text{max}}$) during the evolution time, as a function of both $\Omega$ and the detuning $\Delta_{\text{max}} = \omega_1 - \omega_{\text{max}}$. Since the anharmonicity is negative, to minimize the probability of two-photon absorption the optimal choice of $\Delta_{\text{max}}$ is positive. The optimal probability $p_1^{\text{opt}} \approx 0.81$ occurs for the parameters $\Omega^{\text{opt}}/\Gamma' = 6.75$, $\Delta_{\text{max}}^{\text{opt}}/\Gamma' = 0.4$.

4.3. Effective model for multi-photon dynamics

While figures 9(a) and (b) are calculated with full density matrix dynamics, we now introduce a simplified effective model for the system response in the nonlinear regime. The model enables both a simple understanding of the maximum single excitation probability $p_1$ for different system parameters, and predictions when the atom
number becomes too large for exact density matrix solutions to be feasible. We extend the simple model introduced in the linear regime (section 3.2), again distinguishing independent and collective degrees of freedom of the system. The former are described by an ordinary ‘background’ ensemble of \( n \) single two-level atoms driven far off resonance, and the latter consist of the maximum-energy resonances in the one- and two-photon manifolds, denoted \([1]\) and \([2]\) respectively.

Similar to before, the level scheme for the effective model is shown in figure 5(b), while the Hamiltonian is

\[
H = H_b + H_c, \quad \text{with} \quad H_b = -[(\Delta_{\text{max}} + nV_{\text{eff}}(L))\sigma_+ + \Omega(\sigma_3 + \text{h.c.})]
\]

\[
H_c = -[(\Delta_{\text{max}}\sigma_1 + 2(\Delta_{\text{max}} + V)\sigma_{22} + \sqrt{n}\bar{\nu}_n(\sigma_{11} + \sqrt{2}\sigma_{21} + \text{h.c.})].
\]

For simplicity, in demonstrating the utility of our effective model we shall consider the limiting case \( L \to \infty \), avoiding the need to average over many realizations with different maximum resonance frequencies, as we did in the linear regime (see equation (11) and subsequent discussion). However, the method of section 3.2, describing the use of the effective model in the linear regime for finite \( L \), may straightforwardly be generalized to the multi-photon case considered in this section.

Dissipation is described in the effective model by the Lindblad operators

\[
\mathcal{L}_b[\rho_B] = \frac{\Gamma}{2}(\sigma_+\rho_B + \rho_B\sigma_+ - 2\sigma_+\rho_B\sigma_+),
\]

\[
\mathcal{L}_c[\rho_C] = \frac{\gamma_n}{2}(\sigma_{11}\rho_C + \rho_C\sigma_{11} - 2\sigma_{11}\rho_C\sigma_{11}) + \gamma_0(\sigma_{22}\rho_C + \rho_C\sigma_{22} - 2\sigma_{22}\rho_C\sigma_{22}),
\]

where \( \rho_B \) and \( \rho_C \) respectively denote the density matrices of the background and collective degrees of freedom, and the full system density matrix \( \rho \) is given by \( \rho = \rho_B \otimes \rho_C \). In this multi-photon version of the effective model, the response of the background ensemble of \( n \) independent atoms is again captured as \( n \) identical copies of a single two-level atom. We will be interested in computing the probabilities of different excitation numbers when the system is driven close to the resonance frequency of the collective state \([1]\), and where the probability of background excitations in the background ensemble remains small. As the atoms in an off-resonant ensemble behave in the same way, the actual number of background excitations created follows from taking a binomial distribution based upon the excited state population \( p_n \) of a single atom. In the results presented below, we assume that there are at most two background excitations.

We expect that the effective model introduced above provides a good description of the full system in the regime of weak driving, or to calculate the fidelity of generating a single collective excitation \([1]\). It should break down, however, when quantum jumps from higher excitation manifolds constitute an important part of the system dynamics. In particular, as the background ensemble and collective maximum-energy resonances occupy completely separate Hilbert spaces, our model cannot predict the effect of quantum jumps between them, as are possible in the actual system.

Before applying the effective model to compute excitation probabilities under strong driving, we note that the model may also in principle be used to predict photon bunching in the transmitted field when \( \Gamma_{1D}/T' \gg 1 \), similar to the case of a single emitter coupled to a 1D waveguide where one finds \( g_2^{(2)}(0) \propto (\Gamma_{1D}/T')^4 \) for a resonant field \([37]\). On the basis of figure 3(a) and equation (13), which show that the collective state \([1]\) couples to the waveguide with an average rate \( n\bar{\nu}_n\Gamma_{1D} \), one may naively expect that the simple formula \( g_2^{(2)}(0) \propto (n\bar{\nu}_n\Gamma_{1D}/T')^4 \) should suffice to predict the degree of bunching. However, to obtain good agreement with the full model of equation (1), one must also account for fluctuations in the matrix element \( \kappa_n \) from shot to shot, \( g_2^{(2)}(0) \propto (n(\kappa_n + \epsilon)\Gamma_{1D}/T')^4 \), where \( \epsilon \) is a zero-mean random variable whose statistics may be calculated separately.

Figure 9(c) shows the effective-model prediction for the maximum single-excitation probability \( p_1^{\text{max}} \) reached during the time evolution, as a function of \( \Omega \) and \( \Delta_{\text{max}} \), under the same conditions as figure 9(b). While the laser parameters corresponding to the optimal value \( p_1^{\text{opt}} \) (the global maximum of \( p_1^{\text{max}} \)) do not coincide with those of the full model \( \Omega^{\text{opt}}/T' = 7.2 \) and \( \Delta_{\text{max}}^{\text{opt}}/T' = 2.8 \), the optimal value itself is accurate to within two percent, giving \( p_1^{\text{opt}} \approx 0.83 \). We may then consider the fidelity of introducing only a single excitation for given physical resources \( n \) and \( V \). Figure 10(a) shows \( p_1^{\text{max}} \) for the case \( n = 6 \) as a function of \( V \), optimized over the driving strength \( \Omega \) and the detuning \( \Delta_{\text{max}} \), with both models predicting that \( p_1^{\text{max}} \) approaches unity for large \( V \). Figure 10(b) shows the effective model prediction for \( p_1^{\text{max}} \) as a function of interaction strength \( V \) and number of atoms \( n \), again optimized over the laser parameters. We see that as the interaction strength increases, the fidelity of introducing a single excitation is very high regardless of the number of atoms, since by increasing the strength of the driving one can overcome the weaker coupling of the input field to the spin wave.

Using the effective model, it is straightforward to analytically obtain an estimate of the error in deterministically creating a single excitation. The two different sources of error are (1) dissipation via emission into free space and into the waveguide, and (2) the presence of a second excitation. We begin by considering the
latter case, showing that under appropriate conditions the contribution of the background ensemble to this type of error is negligible. The optimum conditions are then reached by balancing the errors of type (1) and (2) for the anharmonic, collective excitations.

The principal mechanisms by which the system can be doubly excited in the regime of interest are by acquiring population in the collective state [2], or by acquiring a single background excitation in addition to an excitation in state [1]. For off-resonant driving, the probability of exciting a single atom is

\[ p_e = \frac{\Omega^2}{(\Delta_{\text{max}} + n\Omega_{\text{eff}})^2}. \]

It follows that for an ensemble of \( n \) atoms, the total probability \( p^{(1)} \) of introducing a background excitation is (taking for simplicity \( L/d \to \infty \), such that \( \Omega_{\text{eff}} \to V )

\[ p^{(1)} = \frac{n\Omega^2}{(\Delta_{\text{max}} + nV)^2}. \] (17)

Next we consider the population \( p_2 \) of the collective state [2]. We recall that the transition \([1] \to [2]\) has a transition energy different from that of \([g_2] \to [1]\) by an amount \( 2V \). For large \( V \), and near-resonant driving of \([g_2] \to [1]\), the population in state [2] adiabatically follows that of state [1], and is suppressed by an amount

\[ p_2 \approx (\frac{\Omega_n^2}{4V^2}) p_1, \]

where \( \Omega_n = \sqrt{n}\bar{\Omega} \). For large \( n \), we see that this error dominates over the population in the background ensemble \( p^{(1)} \approx \Omega^2/nV^2 \).

Where population of the background ensemble is negligible, the total error in achieving perfect inversion to the collective excited state [1] is then the sum of the error due to intrinsic decay of this state into free space and the waveguide (errors of type (1), as defined above), and the error resulting from the small population acquired in the doubly excited state [2]. As we have shown, the latter may quantified using the simple arguments given above. The former, meanwhile, may be quantified by calculating the maximum population that can be inverted in an ideal two-level system with the same detuning, drive strength, and decay rate. While there is no simple closed-form expression for this, one can nonetheless find the optimal error numerically by solving the simple two-level system master equation

\[ \dot{\rho} = -i[H, \rho] + \mathcal{L}[\rho], \]

where

\[ H = \Delta_{\text{max}} \sigma_1 + \Omega_n (\sigma_1 x + \sigma_1 y) \]

\[ \mathcal{L}[\rho] = \frac{\gamma_n}{2} (\sigma_{11} \rho + \rho \sigma_{11} - 2\sigma_{11} \rho \sigma_{11}). \] (18)

As an example of how this simplified form of the effective model may be applied, we consider the configuration corresponding to \( n = 20 \) and \( V = 100 \) in figure 10(b), where the optimal detuning and laser strength are found to be \( \Delta_{\text{max}}/V = 5.5 \) and \( \Omega_{\text{opt}}/V = 36.9 \) respectively. Solving equation (18) with these parameters, we find the error in achieving perfect inversion of an ideal two level atom to be \( \approx 0.0337 \). Meanwhile, following our analytical analysis, we find the error associated with the population of the state [2] to be \( \approx 0.0188 \), and the error due to the introduction of a background excitation to be \( 0.0068 \). The total error due to the three effects is then 0.0593, predicting an optimal inversion of \( p_1^{\text{max}} = 0.9407 \), while the value predicted by the complete effective model of equations (15) and (16) is 0.9528. In this case, therefore, simply summing the errors of the two different loss mechanisms gives a value \( p_1^{\text{max}} \) that agrees with the prediction of the effective model to within approximately one percent.

The above discussion has focused on the probability of preparing the first Dicke state [1], however due to the strong anharmonicity for large \( V \) in our system, it is also possible in principle to prepare higher-order Dicke states, as we now briefly outline. To maximize the preparation fidelity, one should follow a multi-step procedure where each Dicke state up to the desired order is prepared in succession, under the corresponding optimal
conditions. For example, to reach the second Dicke state [2], the first Dicke state [1] is prepared using the optimal laser strength and detuning as we have described in the preceding sections. At a time corresponding to the maximum excitation probability of state [1], the external field is then modified to have the corresponding optimal frequency and strength for the transition from [1] to [2]. In the simple case $L \to \infty$, the energy $E_m$ of the maximum-energy $m$-excitation eigenstate of equation (7), i.e. the $m$th Dicke state $|m\rangle$, is given by $E_m = m(n + 1 - m) V$. It follows that the energy difference between successive Dicke states $|m\rangle$ and $|m + 1\rangle$ is $E_{m+1} - E_m = (n - 2m) V$, which then corresponds to the detuning to be applied to the driving frequency each time an additional excitation is created.

While the scheme we have outlined would in principle allow for the efficient preparation of higher-order Dicke states, we note that the higher the desired excitation number, the more limited the method is by the errors we have described above. In particular, for large $m$ the energy spectrum of the Dicke states becomes increasingly linear, and moreover each successive state is located closer to the collection of non-shifted modes in the same manifold. Given the limitations on the number of atoms $n$ and dipole–dipole interaction strength $V$ that can be achieved in PCW systems (appendix A), we expect their potential utility in generating such collective entangled states to be restricted to low excitation numbers.

5. Summary and outlook

In summary, we have shown that an ensemble of two-level atoms with long-range interactions has rich optical properties in both the single- and many-photon regimes. In order to understand and inform experiments that are within reach, the model we have considered is conceptually the simplest that produces long-range coupling and allows the atoms to be probed. We have explained how the resulting physics may be described in terms of yet simpler, and more intuitive models for a wide range of parameter regimes of interest. Moreover, we have shown how spectroscopic measurements may be used to determine vital system parameters, such as the number of atoms, in a regime where conventional techniques break down. Moving forward, it may be possible to exploit strong nonlinearities to produce a blockade effect, where within some region of the system only a single excitation is supported. If the full system is large enough to support several such regions, one would expect the scattered light to exhibit rich spatiotemporal correlations [19].

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Appendix A. Realistic experimental parameters and losses

In our model, the photonic crystal is fully characterized by the parameters $V$ and $L$, respectively describing the coupling strength of an atom to the TE modes and the characteristic length of the resulting photonic cloud. The values of these parameters are principally determined by: (1) the band curvature $\alpha$ at the band edge, where in the effective mass approximation the dispersion relation is given by $\omega_k \approx \omega_{k_0}(1 - \alpha (k - k_0)^2/k_0^2)$, with $\omega_{k_0}$ and $k_0$ respectively denoting the band edge frequency and wavevector [13], and; (2) the detuning of the atomic resonance frequency from the band edge frequency, $\delta = \omega_a - \omega_{k_0}$, as shown in figure 1(b) of the main text. Appropriate design of the photonic crystal allows these parameters, $\alpha$ and $\delta$, to be tuned to desired values, in turn allowing control of the parameters $V$ and $L$.

In particular, it can be shown that $L = \sqrt{\alpha \omega_{k_0}/k_0^2 \delta}$, i.e. the interaction length decreases for flatter bands (smaller $\alpha$) and larger detuning from the band edge [13]. Given this length $L$, it can then be shown that the interaction strength $V$ is identical to what atoms would experience in a real photonic crystal cavity of the same mode volume, and with an effective atom–cavity detuning given by $2\delta$. Specifically, one finds that $V = \tilde{g}_d^2/2\delta$, where $\tilde{g}_d = g_d \sqrt{d/L}$, and $g_d$ is the vacuum Rabi splitting that would be achieved in a photonic crystal cavity of length $d$.

A realistic platform in which this may be realized is the APCW, described in [2, 15, 20]. A comprehensive analysis of realistic experimental values for our parameters of interest was provided in [13], which we now summarize. For the APCW described in [20], the band curvature $\alpha = 10.6$ at the band edge, and the atom–cavity coupling $g_d/2\pi = \sqrt{d/L} \times 12.2$ GHz. Taking a detuning of $\delta/2\pi = 400$ GHz, band edge wavevector
\[ k_b = \pi/d, \]  
and the free space emission rate for Caesium to be \( \Gamma' / 2\pi \approx 5 \text{ MHz} \), one finds \( L/d \approx 30 \), and \( V / \Gamma' \approx 1.25 \). Stronger and longer-range interactions may be obtained by reducing the detuning \( \delta \) of the atoms from the band edge.

A necessary condition to observe the linear and nonlinear behavior we describe is that the interaction strength \( V \) must exceed the total rate of dissipation. The dominant dissipative processes are: (1) free-space emission from the atomic excited state, at characteristic rate \( \Gamma' \); (2) emission into the TM waveguide modes, at rate \( \Gamma_{\text{ID}} \); and (3) photon loss in the crystal, resulting from material absorption and scattering, as well as a small residual photon transmittance in the TE mode inside the band gap, with a combined rate \( \kappa \).

It can be shown that the total effective dissipation rate for an excited atom is given by
\[ \Gamma_{\text{tot}} = \Gamma' + \Gamma_{\text{ID}} + \kappa (\bar{g} / \delta)^2, \]
where the term proportional to \( \kappa \) represents the Purcell enhancement arising from the atom being off-resonantly coupled to an effective cavity mode. The ratio of interaction strength to total losses \( V / \Gamma_{\text{tot}} \) can be optimized with respect to detuning \( \delta \). Doing so, one finds that the theoretical maximum is given by \( V / \Gamma_{\text{tot}} \approx \sqrt{C} \), where \( C = \bar{g}^2 / (\Gamma \kappa) \) is the cooperativity factor, with \( \Gamma = \Gamma' + \Gamma_{\text{ID}} \). It should be noted that in order to optimize \( \delta \) but maintain a fixed interaction length \( L = \sqrt{\alpha \omega_b / \kappa \delta^2} \), the band curvature of the structure must be optimized simultaneously. Thus, the numbers provided for the APCW above are far from the theoretical limit, as the curvature parameter has not been optimized.

In fact, if one assumes that the minimum photon loss rate \( \kappa \) is comparable to what can be achieved in a real photonic crystal cavity (i.e. the fabrication—see below—and material imperfections are comparable), then one can re-write the cooperativity as \( C \approx Q d / L \) \([13]\). Here \( Q \) represents the quality factor of an actual photonic crystal cavity of length \( d \), which can readily exceed \( 10^5 \). This implies, for example, that the coherent rate of interaction \( V \) can exceed the total rate of dissipation by an order of magnitude over distances of up to 1000 lattice sites. The choices of \( V \) and \( L \) in our numerical calculations thus fall well within the realistically achievable bounds, except for the single case where \( L \to \infty \). This case, however, merely serves as a conceptual limit and idealized point of comparison, which in practice is well-approximated when the interaction length approaches the total system size, \( L/Nd \gtrsim 1 \).

Finally, we consider the effect of disorder in the APCW due to fabrication imperfections. Defects in the crystal may arise from, for example, imperfect periodicity in the refractive index variation, and/or differences in the dielectric contrast from one unit cell to another. Such defects result in Anderson localization, where a propagating mode in a perfect waveguide is trapped over some characteristic localization length \( \xi \). The role of Anderson localization on the atom–atom interactions becomes significant when \( \xi \) is shorter than the ideal interaction length \( L \).

A theoretical analysis of the effects of disorder near a photonic crystal band edge was given in \([13]\). As discussed therein, one finds that localization lengths of \( \xi / d > 100 \) are possible with state-of-the-art photonic crystal fabrication. Moreover, the role of disorder in an APCW was investigated experimentally in \([2]\), revealing a localization length due to fabrication imperfections of at least the length of the structure itself, i.e. \( \xi / d > 200 \).

**Appendix B. Computation of field correlations**

One main objective of our work is to compute correlation functions of the output field, i.e. of the field in the waveguide (TM mode) after scattering from the atomic ensemble has taken place. The technique we employ to achieve this is described in \([23, 24]\), and may be summarized as follows.

Starting from a full description of the atoms and fields, one can formally integrate out the field degrees of freedom to obtain a master equation for the atomic density matrix. With any master equation, an equivalent description can also be obtained in the ‘quantum jump’ picture. Here the wavefunction of the atoms evolves under the non-Hermitian Hamiltonian given by equation (1) in the main text, which describes the atomic ‘spins’ interacting by photon exchange. This deterministic evolution must be supplemented with stochastic quantum ‘jumps’.

The jump processes that may occur in our system are single-atom emission into free space, described by the operator \( O_j = \sqrt{\Gamma / 2} \sigma_p^j \) for the \( j \)th atom, and collective decay into the right- and left-propagating modes of the waveguide, described by the operators \( O_{R,L} = \sqrt{\Gamma_{\text{ID}} / 4} \sum_k \exp(\mp ik \xi) \sigma_p^k \) (for \( R \) + for \( L \)). Once the atomic correlation functions have been obtained (either by the master equation or quantum jump approach), one can use the formally integrated expressions for the fields to calculate field correlations in terms of the atomic ones, making use of equations (4) and (5) in the main text.

According to the quantum jump formalism, during evolution of the atomic wavefunction under \( H \), stochastic quantum jumps should be applied for each decay process at a frequency \( f_{ij}(t) = \langle \psi(t)|O^O|\psi(t) \rangle \), where \( |\psi(t)\rangle \) is the time-evolving wavefunction. However, we will now argue that under the appropriate circumstances, \( |\psi(t)\rangle \) can be replaced with the quasi-steady state (quasi-SS) wavefunction, where jumps occur sufficiently infrequently that they may be neglected altogether.
Formally, after a jump, evolution under the effective Hamiltonian $H$ will cause the system to lose memory of the jump and return to the quasi-SS in a maximum time of $\tau_{eq} \approx 1/f'$, since $f'$ represents the minimum decay rate that any excited state can have. The expectation values of correlations will then be dominated by the values associated with the quasi-SS wavefunction provided that the fraction of time spent out of equilibrium is negligible, i.e. provided $\tau_{eq}/f_{jump} \ll 1$, where $f_{jump} = f_{Out} + f_{In} + \sum f_{jO}$ is the total jump frequency incorporating all decay processes.

These general considerations can now be applied to our regime of interest. We consider the lattice spacing $k_d = \pi/2$, with a weak coherent state input on resonance in the absence of the BG interactions. In this case, the transmitted intensity decays exponentially in the number of atoms, $T \sim \exp(-\Omega D)$, where the optical depth is given by $OD \sim 2n\Gamma D/\Gamma'$ [24]. For large optical depth, the strong attenuation results from a near cancellation of the incident field and the field re-emitted in the forward direction by the atoms. Thus, from equation (2) in the main text we have

$$\langle \psi | a_{out,t} | \psi \rangle = \langle \psi | O a | \psi \rangle + \frac{\Gamma_D}{2} \sum_j e^{-ik_{D}z_j} \langle \psi | \sigma_{jL}^{\dagger} | \psi \rangle \approx 0. \quad \text{(B.1)}$$

The operator in the second term of equation (B.1) is simply the jump operator for the right-propagating field, $O_{R}$, divided by a factor of $\sqrt{\Gamma_D}$, so we immediately have that $\langle \psi | O_{R} a | \psi \rangle \approx -i\sqrt{\Gamma}/\Gamma_D \Omega$, while in the weak driving limit we moreover have $\langle \psi | O_{R} O_{R} a | \psi \rangle \approx \Omega^2/\Gamma_D$, giving the jump frequency for this decay mode.

For our choice of lattice spacing, the reflectance $R = (\Gamma_D/\Gamma)^{2n}$, where $\Gamma = \Gamma_D + \Gamma'$, is more strongly suppressed in the number of atoms than is the transmittance, from which it follows that for large optical depth $\langle \psi | O_{R} a | \psi \rangle \approx 0$. Having concluded that the majority of incident photons in the driving field $\Omega$ are neither transmitted nor reflected, they must be scattered into free space. In order to maintain the SS, the rate at which photons are emitted into free space must equal the rate at which they are input to the system, which also implies $\sum_j \langle \psi | O_{i} a | \psi \rangle \approx 2\Omega^2/\Gamma_D$.

Using $\tau_{eq}/f_{jump} \ll 1$, the condition for quantum jumps to be negligible is equivalent to a condition on the strength of the incident field:

$$\Omega \ll \sqrt{\frac{\Gamma_D \Gamma'}{3}}. \quad \text{(B.2)}$$

While this specific condition results from an analysis of a resonant input field, an analogous inequality may be derived for weak input fields of other frequencies, provided that the average time between quantum jumps $(1/f_{jump})$ becomes large compared to the memory time of the system. As an important point, generically for sufficiently weak intensities quantum jumps can be ignored, since the jump rate scales like $\Omega^2$.

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