Unbound-to-bound transition of two-atom polaritons in an optical cavity

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We consider two spin-1/2 fermions inside an optical cavity which supports a single-mode quantized light field. We demonstrate that the atom-light coupling (ALC) gives rise to the two-atom polariton states, where the two atoms are highly entangled with cavity photons. We focus on the case where the cavity light is on resonant with the bare atomic transition. We show that in the absence of interatomic interaction, the polariton is unbound, has finite center-of-mass momentum, and contains no atomic spin-singlet fraction in its ground state. When strong attractive interatomic contact interaction is present, a stable bound polariton state exists when the ALC strength is below a critical value. When the ALC strength exceeds the critical value, a first-order transition is observed and the bound polariton becomes unbound. The first-order transition is characterized by abrupt changes of various quantities associated with the polariton and should be readily detectable.

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I. INTRODUCTION

With the successful realization of cold atoms loaded into optical cavities [1,2], cavity quantum electrodynamics (QEDs) with ultracold atoms [3–5] has attracted enormous attention and provided a unique platform to explore exotic collective quantum behaviors of the hybrid atom-cavity systems [6–28]. For example, the noted Dicke super-radiance transition [29,30] has been realized in cavity Bose-Einstein condensate (BEC) systems [9,10] and then extended to Fermi gases [12–14]. This research reveals the important effects brought by the external center-of-mass (COM) motion of atoms in the dispersive coupling regime [5]. On the other hand, the interatomic interactions may also compete with the atom-light coupling (ALC) dramatically, resulting in the super-radiant solid in a cavity Rydberg system [16].

Another recent breakthrough is the realization of synthetic non-Abelian gauge potentials, which provide a coupling between the atomic COM and its internal degrees of freedom by implementing two Raman beams on ultracold atoms, forming an effective spin-orbit coupling (SOC) [31–36]. This SOC has stimulated numerous studies and underlies a variety of many-body phenomena [37–42]. In recent proposals [43–47], one of the Raman beams is replaced by an optical cavity field. It was shown that a cavity-assisted two-photon Raman transition can give rise to dynamical SOC with many intriguing nonlinear properties.

In this paper, we investigate the two-atom ground-state properties of an attractive two-component Fermi gas inside an optical cavity. In contrast to the classical Raman beams, the cavity field is quantized and causes the nonlocal atom-light coupling. The cavity photons entangle with the atoms and interplay with the interatomic interactions, which results in an exotic two-atom polariton state. Tuning of the ALC strength can induce a first-order phase transition in this exotic polariton, which changes between a bound and an unbound state. Accompanying this transition, the entanglement entropy, the photon population, and the atomic spin-singlet fraction associated with the polariton state exhibit discontinuous jumps across the critical ALC strength. With progress in experimental techniques of the atom-cavity system, our study has interesting implications for future experiments.

II. THE MODEL

We consider a two-component Fermi gas coupled to a single-mode cavity field with wave vector \( \mathbf{k} \), and frequency \( \omega_c \), which can be implemented by a unidirectional optical ring cavity [44,48–50]. Under the rotating-wave approximation, the Hamiltonian of the system is given by

\[
\hat{H} = \hat{H}_{\text{int}} + \hat{H}_{\text{atom}} + \hat{H}_{\text{dc}}. 
\]

Here, \( \hat{H}_{\text{atom}} \) denotes the annihilation operator of the single-mode light field.

\[
\hat{H}_{\text{atom}} = \sum_{\sigma = \uparrow, \downarrow} \int d^3 \mathbf{r} \left[ \hat{\psi}^\dagger_\sigma(\mathbf{r}) \hat{P}_{2m} + \epsilon_\sigma \right] \hat{\psi}_\sigma(\mathbf{r}) + \hat{H}_{\text{int}}. 
\]

\( \hat{H}_{\text{int}} \) describes the atoms moving inside the cavity, where \( \hat{\psi}_\sigma(\mathbf{r}) \) denotes the annihilation operators of the atomic state \( \sigma (\uparrow, \downarrow) \), and \( \hat{P}_{2m} \) is the bare atomic energies, and \( \hat{H}_{\text{dc}} = \frac{1}{2} g_0 \int d\mathbf{r} \nabla \nabla \cdot \mathbf{r} \hat{\psi}^\dagger_\uparrow(\mathbf{r}) \hat{\psi}_\uparrow(\mathbf{r}) + \hat{\psi}^\dagger_\downarrow(\mathbf{r}) \hat{\psi}_\downarrow(\mathbf{r}) \), describes the atomic interaction with \( \hat{n}_\sigma(\mathbf{r}) = \hat{\psi}^\dagger_\sigma(\mathbf{r}) \hat{\psi}_\sigma(\mathbf{r}) \). The ALC is described by

\[
\hat{H}_{\text{dc}} = g_0 \int d\mathbf{r} \hat{\psi}^\dagger_\uparrow(\mathbf{r}) \hat{\psi}_\downarrow(\mathbf{r}) + \text{H.c.}, 
\]

with the coupling strength \( g(\mathbf{r}) = g_0 e^{i \mathbf{k} \cdot \mathbf{r}} \) and \( g_0 \) the single-photon Rabi frequency. Through a gauge transformation,

\[
\hat{\psi}^\dagger_{\uparrow, \downarrow} \rightarrow e^{\mp i \mathbf{k} \cdot \mathbf{r}} / \sqrt{2} \hat{\psi}^\dagger_{\uparrow, \downarrow},
\]

we can eliminate the position-dependent phase factor in \( g(\mathbf{r}) \) [51], and the total Hamiltonian in momentum space can be
written as $\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}$, where

$$\hat{H}_0 = \hbar \omega_c \hat{a}^\dagger \hat{a} + \sum_{k, \sigma} \varepsilon_{k\sigma} \hat{\psi}_{k\sigma}^\dagger \hat{\psi}_{k\sigma}$$

$$+ \sum_k g_0 \left( \hat{\psi}_{k\uparrow}^\dagger \hat{\psi}_{k\downarrow} \hat{a} + \text{H.c.} \right),$$

$$\hat{H}_{\text{int}} = \frac{U}{V} \sum_{k, k', q} \hat{\psi}_{q/2+k\uparrow}^\dagger \hat{\psi}_{q/2-k\uparrow} \hat{\psi}_{q/2-k\downarrow} \hat{\psi}_{q/2+k\downarrow},$$

where $\varepsilon_{k\sigma}(\lambda) = \delta k^2 + \lambda \hat{a}^\dagger \hat{a}$, with $\lambda \equiv |\mathbf{k}|/2$, and we define $\mathbf{k}_c$ to be along the $z$ axis of the cavity. Note that the atomic part $\varepsilon_k$ is formally equivalent to that of the one-dimensional spin-orbit coupling [31]. The contact attractive interaction $U$ between atoms (e.g., $^6\text{Li}$, $^{40}\text{K}$, etc.) is well captured by the low-energy $s$-wave scattering length $a_s$, which should be regularized as $\frac{m}{4\pi\hbar^2 a_s} = \frac{1}{\pi} + \frac{1}{\pi} \sum_k \frac{1}{\varepsilon_k}$, with $\epsilon_k = \hbar^2 k^2/2m$ and $V$ being the quantization volume [52]. For simplicity, we take $m = \hbar = 1$.

In the case of a single atom, we can easily diagonalize $\hat{H}_0$ to find its spectrum [53]. Here the total excitation number $N_c \equiv \hat{a}^\dagger \hat{a} + \sum_k \hat{\psi}_{k\uparrow}^\dagger \hat{\psi}_{k\uparrow}$ is conserved. For given $N_c = N$, the spectrum contains two branches, and the dispersion of the lower branch is given by

$$E_-(\mathbf{k}) = \left( N - 1 \right) \omega_c + \frac{\omega_c + \mathbf{k}^2 + \lambda^2}{2} - \sqrt{\left( k^2 \pm \delta/2 \right)^2 + 4 Ng_c^2},$$

where $\delta = \omega_a - \omega_c$ denotes the atom-cavity detuning. For $\delta = 0$, i.e., when the cavity photons are resonant with the bare atomic transition, which will be the focus of this work, there exists a critical ALC strength $g_c = \lambda^2/\sqrt{N}$. When $g_0 < g_c$, the ground state is doubly degenerate and occurs at $k_z = \pm q_{\text{min}} = \pm \sqrt{\lambda^2 - Ng_c^2}/\lambda$. When $g_0 \geq g_c$, $q_{\text{min}} = 0$ and the two degenerate ground states merge into a single one. $q_{\text{min}}$ as a function of $g_0$ is plotted in Fig. 1(b) as the dotted line.

It is noteworthy that the above single-atom polariton spectrum under constant excitation number is similar to the single-atom spectrum of the SOC system induced by the classical Raman field [31]. However, when we consider the two-atom problem, the atom-cavity system would differ significantly from the classical field system. As it is well known, in the absence of atomic interactions, the energy of two atoms coupled to a classical field would be simply given by the sum of the individual single-atom energies. By contrast, when the two atoms are coupled to a cavity field, the latter can induce an effective inter-atomic interaction. As a result, the two atoms can no longer be regarded as independent from each other. In fact, they will be entangled with the cavity photons and form a nontrivial unbound two-atom polariton continuum. We now turn to study the properties of such an unbound polariton in detail.

### III. UNBOUND TWO-ATOM POLARITON CONTINUUM

In the absence of the $s$-wave interaction, $U = 0$. The momentum of the two atoms is individually conserved. Take the momentum of the atoms to be $\mathbf{k}_1 = \frac{\mathbf{q}}{2} + \mathbf{k}$ and $\mathbf{k}_2 = \frac{-\mathbf{q}}{2} - \mathbf{k}$ ($\mathbf{k}$ and $\mathbf{q}$ are the relative and the COM momenta, respectively) and a fixed excitation $N$ (we assume $N > 1$); the wave function of the unbound two-atom polariton (UP) can be written as

$$\Phi_{\mathbf{k}, \mathbf{q}} = \psi_{\mathbf{k}, \mathbf{q}}^\dagger \psi_{\mathbf{k}, \mathbf{q}}^\dagger \psi_{\mathbf{k}_1 \uparrow} \psi_{\mathbf{k}_1 \downarrow} | N - 1 \rangle + \psi_{\mathbf{k}, \mathbf{q}}^\dagger \psi_{\mathbf{k}_1 \uparrow} \psi_{\mathbf{k}_1 \downarrow} \psi_{\mathbf{k}_2 \uparrow} \psi_{\mathbf{k}_2 \downarrow} | N - 2 \rangle + \psi_{\mathbf{k}, \mathbf{q}}^\dagger \psi_{\mathbf{k}_1 \uparrow} \psi_{\mathbf{k}_1 \downarrow} \psi_{\mathbf{k}_2 \uparrow} \psi_{\mathbf{k}_2 \downarrow} | N \rangle,$$

where $|n\rangle = \frac{1}{\sqrt{m}}(|\hat{a}^\dagger|^n |0\rangle$ denotes the $n$-photon Fock state. From the Schrödinger equation $\hat{H}_{\text{ST}} \Phi_{\mathbf{k}, \mathbf{q}} = E_{\mathbf{k}, \mathbf{q}} \Phi_{\mathbf{k}, \mathbf{q}}$, one can derive the equation for the eigenergym $E_{\mathbf{k}, \mathbf{q}}$, which is given by

$$E_{\mathbf{k}, \mathbf{q}} = \frac{\sqrt{N(N-1)g_c^2} + \sqrt{N(N-1)g_c^2 + 4Ng_c^2}}{\sqrt{\delta - k_c q_z/2} + \sqrt{\delta + k_c q_z/2}} + \frac{(k_c q_z^2)^2}{E_{\mathbf{k}, \mathbf{q}} - \delta + k_c q_z/2},$$

where $\delta = \omega_2 - \omega_1$.

The first and second terms at the right-hand side of Eq. (4) describe the energy corrections from the processes of first annihilating and then creating a photon, and vice versa, while
the third term originates from the singlet component induced by the finite momentum transfer from photons. Due to the ALC, $E_{\text{th}}$ consists of four branches forming the continuum. The ground-state energy of the UP is given by the minimum of the lowest branch, which is also the threshold of the continuum spectrum, and hence we denote it as $E_{\text{th}}$. In Fig. 1(a), we plot $E_{\text{UP}} = E_{\text{th}} - (N - 1)\omega_c$ as a function of the ALC strength $g_0$, where $(N - 1)\omega_c$ is the ground-state energy in the limit of $g_0 = 0$ for excitation number $N$ and $\delta = 0$. One can see that $E_{\text{UP}}$ is a decreasing function of $g_0$, which scales as $g_0$ for large $g_0$, and as $\sqrt{N}$ for large excitation number $N$.

By solving the Schrödinger equation, we can also obtain the wave function. The wave function represented by Eq. (2) can be decomposed into triplet and singlet components. In particular, the singlet component is given by $\psi_{k,q} = \frac{1}{\sqrt{2}}(\phi_{k-q}^{+} - \phi_{k+q}^{+})$ and can be shown to be proportional to $k_z$, the $z$ component of the relative momentum between the two atoms (see Appendix A). Numerically solving Eq. (4) to obtain the minimal energy, we find that the ground-state UP always has $k_z = 0$, and hence the ground-state UP is purely spin triplet.

Next, we address the question concerning the COM momentum of the ground-state UP, denoted as $q_{\text{UP}}$. Our calculation shows that $q_{\text{UP}} = q_{\text{UP}} e_z$ is always nonzero. In Fig. 1(b), we plot $q_{\text{UP}}$ as a function of $g_0$. In the limit of small $g_0$, we have

$$ q_{\text{UP}} \simeq k_z \sqrt{1 - (4N - 2)(g_0/E_c)^2}. $$

When $g_0 = 0$, $q_{\text{UP}} = k_z$, which is twice the ground-state momentum of a single atom $q_{\text{min}}$. However, for finite $g_0$, we have $q_{\text{UP}} \neq 2q_{\text{min}}$. In particular, when $g_0 \geq g_c = \lambda^2/\sqrt{N}$, $q_{\text{min}}$ vanishes, whereas $q_{\text{UP}}$ remains finite and approaches $k_z/(4N - 2)$ in the limit of large $g_0$. This is in qualitative difference when the SOC is induced by classical laser beams, in which case the COM momentum of a two-atom system should just be $2q_{\text{min}}$ regardless of the atom-photon coupling strength, and hence vanishes in the limit of large $g_0$. In our system, this classical limit is reached when the excitation number $N$ becomes large.

For small $N$, however, photon-number fluctuation is significant and the quantum result deviates away from the classical limit.

Finally, we consider the entanglement between the atoms and the cavity field, which can be quantified by the von Neumann entanglement entropy defined as $S = -tr\rho_A \ln \rho_A$, where $\rho_A$ is the reduced density matrix for either atoms or photons (see Appendix C). Figure 1(c) depicts the entanglement entropy $S_{\text{UP}}$ for the ground-state UP as a function of $g_0$. $S_{\text{UP}}$ increases as $g_0$ and saturates at a value of $\ln 2 + \ln(2N - 1)/2 - N \ln N / (4N - 2) - (N - 1)\ln(N - 1)/(4N - 2)$ in the limit of large $g_0$.

IV. BOUND TWO-ATOM POLARITONS

Now we turn to consider the attractive atomic interactions, which favors the singlet pairing between the two atoms. We are interested in the regime with $\alpha_s > 0$ [54] and hence the two atoms, together with cavity photons, can form a bound polariton (BP). First we note that the $s$-wave interaction term $H_{\text{int}}$ in Eq. (1) has no effect on the triplet component of the UP wave function. As a result, the ground-state UP remains as an eigenstate of the full Hamiltonian $\hat{H}$. In general, however, the interatomic interactions can mix states with different relative momentum $k$, and only conserve the COM momentum $q$. Therefore, a general wave function of BP with COM momentum $q$ for a given excitation number $N$ can be written as

$$ \Psi_q = \sum_k \phi_{k,q}. $$

where the summation over the relative momentum $\sum_k$ is limited to $k_z \geq 0$, and $\phi_{k,q}$ takes the same form as the UP wave function given in Eq. (2). Putting this into the Schrödinger equation $\hat{H}_q \Psi_q = E_q \Psi_q$, we can find that the eigenenergy $E_q$ is given by

$$ E_q = E_{k,q}^{\text{up}} + \epsilon_2^{\text{up}} + \epsilon_2^{\text{k}} - \frac{k_z^2}{4} + (N - 1)\omega_c, $$

where $E_{k,q}^{\text{up}}$ satisfies (see Appendix B)

$$ \sum_k \left[ \left( E_{k,q} - \frac{k_z^2}{4} \right) \left( \frac{2N\epsilon_2}{E_{k,q}^{\text{up}} + k_z^2} \right)^{-1} + \frac{1}{2\epsilon_2} \right] = \frac{V}{4\pi \alpha_s}. $$

earlier). Next, we turn to the transition between the BP and the UP as $g_0$ is tuned.

V. BOUND-TO-UNBOUND POLARITON TRANSITION

A BP is only energetically stable, and hence truly bound, when its energy is below the continuum threshold. Therefore, it is instructive to calculate the quantity $\epsilon_b = E_{\text{UP}} - E_{\text{th}}$, which is plotted in Fig. 3(a) as a function of $g_0$ for a given scattering length $(k_0 \alpha_s)^{-1} = 1$. The BP is stable if $\epsilon_b < 0$. As one can see from the figure, stable BP exists for small $g_0$. As $g_0$ increases, $\epsilon_b$ increases and eventually reaches zero and the BP is no longer stable. From this calculation, we can thus generate a phase diagram in the parameter space spanned by $g_0$ and the inverse
functions of the normalized ALC strength is of first order. This can be clearly seen from the true ground state of the system.

The transition between the BP and the UP across the critical ALC strength is of first order. This can be clearly seen from the discontinuous jump exhibited by the ground-state photon number for \( n_{ph} \) and the singlet population in the ground state changes from BP to the UP, and the photon population jumps upward. On the other hand, the UP is dominated by the singlet component due to the strong s-wave interaction, and the singlet population suddenly vanishes across the critical point as the UP is purely triplet. In addition to the photon and the singlet population, the entanglement entropy also has a discontinuous jump across the transition point, which can be inferred from Figs. 1(c) and 2(c).

In the case where the SOC is induced by classical Raman laser beams and in the presence of strong attractive s interaction, we can also find a bound two-atom dimer state which has also a sudden jump across the transition point, which can be inferred from Figs. 1(c) and 2(c).

In the case where the SOC is induced by classical Raman laser beams and in the presence of strong attractive s interaction, we can also find a bound two-atom dimer state which becomes unbound when the Raman coupling strength exceeds a critical value. This bound-to-unbound transition is, however, continuous [55]. The first-order transition in our current study is due to the quantum nature of the cavity field. The s-wave interaction favors singlet pairing associated with a relatively small photon population, whereas, as can be seen from the wave function (2), the largest photon component is always associated with a spin-triplet state with both atoms occupying the \( | \downarrow \rangle \) state. Since a large photon number enhances effective atom-photon coupling and decreases the total energy, there exists a competition between the s-wave interaction and the ALC, which results in the first-order transition observed here.

VI. DISCUSSIONS

We first discuss some experimentally related issues. Recently, a small but definite number of fermions [56] and homogeneous box potentials, both for the boson [57] and fermion [58], have been created. Noticing that it has been routine to couple a Bose-Einstein condensate with a high-finesse optical cavity [1,2,5] and that a tunable Jaynes-Cummings-type coupling between atomic internal states and an optical cavity has also been realized [11], and considering that it does not seem to be fundamentally difficult to couple a Fermi gas with an optical cavity, it should be possible to realize the model studied in this paper for two atoms in the near future. More recently, two neutral atoms coupled to a cavity field collectively has been reported [59]. Beyond two atoms, the situation becomes much more complicated and, besides the bounded two-atom state, other exotic states may exist in the absence of ALC, e.g., the Efimov-type trimer state [60], the polaron [61], and so on. One may expect that these states would also be changed dramatically by the ALC. Nevertheless, the detailed analysis...
of such states is beyond the scope of this work, and we leave it for future study.

So far, we focus on the equilibrium behaviors and have neglected the impacts of dissipation. For a realistic atom-cavity system, the most important decay channel is the leakage of cavity photons with decay rate $\kappa$. In the good cavity limit, $\kappa$ is much smaller than $g_0$ and $U$, the system can be viewed in a quasiequilibrium state within timescale $1/\kappa$ and the above results can be applied. Recently, the polariton-type spectrum has been observed for a BEC in an optical cavity [1], while in the bad cavity limit, $\kappa$ is comparable with (or larger than) other energy scales. To maintain a long-run cavity field, one needs to implement a driven laser field, which does not conserve the polariton number $N_p$. In that case, the cavity field relaxes to a stationary coherent state in a timescale that is typically faster than the dynamics of the atoms [62]. An appropriate treatment is to replace the photon operators in Eq. (2) with a coherent field, which may lead to a steady-state solution [5].

VII. CONCLUSION

In conclusion, we have shown that a system of two spin-$1/2$ fermions coupled to a single-mode cavity field displays a rich variety of physics, due to the interplay between the interatomic interaction and the atom-cavity coupling. The atoms and the cavity field form a two-atom polariton. As the atom-cavity coupling strength is tuned, a first-order transition between a bound and an unbound polariton state is observed. Accompanying this transition, the photon population, atomic spin-singlet fraction, atom-photon entanglement entropy, and the momentum of the polariton all exhibit discontinuous jumps. These jumps render the transition readily detectable in experiments. Our work paves a way for future studies of quantum few-body and many-body physics in atom-cavity systems.

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APPENDIX A: UNBOUND TWO-ATOM POLARITON CONTINUUM

First consider two noninteracting atoms with momenta $\mathbf{k}_{1,2} = \frac{\mp}{2} \pm \mathbf{k}$ (where $\mathbf{k}$ are the relative and the COM momenta, respectively); the polariton wave function $\Phi$ for a given excitation number $N$ can be written as

$$\Phi_{\mathbf{k}, q} = \psi_{\mathbf{k}, q}^{+} + \psi_{\mathbf{k}, q}^{\dagger},$$

where $|n\rangle = \frac{1}{\sqrt{m}}(|\hat{a}^{\dagger}|^{n}|0\rangle$ denotes the $n$-photon Fock state. In this case, the Schrödinger equation $\hat{H}_0|\Phi_{\mathbf{k}, q}\rangle = E_{\mathbf{k}, q}|\Phi_{\mathbf{k}, q}\rangle$ can be cast into a matrix form $(\hat{M} - E_{\mathbf{k}, q})\Phi = 0$ with

$$\hat{M} = \begin{pmatrix} \epsilon_{\mathbf{k}, \uparrow} + \epsilon_{\mathbf{k}, \downarrow} + \delta & g_0\sqrt{N - 1} & g_0\sqrt{N - 1} & 0 \\ g_0\sqrt{N - 1} & \epsilon_{\mathbf{k}, \uparrow} + \epsilon_{\mathbf{k}, \downarrow} & 0 & g_0\sqrt{N} \\ g_0\sqrt{N - 1} & 0 & \epsilon_{\mathbf{k}, \uparrow} + \epsilon_{\mathbf{k}, \downarrow} & g_0\sqrt{N} \\ 0 & g_0\sqrt{N} & g_0\sqrt{N} & \epsilon_{\mathbf{k}, \uparrow} + \epsilon_{\mathbf{k}, \downarrow} - \delta \end{pmatrix}.$$ (A2)

Then, the eigenvalue $E_{\mathbf{k}, q}$ of $\hat{M}$ satisfies

$$E_{\mathbf{k}, q} = \frac{2(N - 1)g_0^2}{\tilde{\epsilon}_{\mathbf{k}, q} - \delta - k_c q_z/2m} + \frac{2N g_0^2}{\tilde{\epsilon}_{\mathbf{k}, q} + \delta + k_c q_z/2m} + \frac{(k_c^2)^2}{\tilde{\epsilon}_{\mathbf{k}, q}},$$ (A3)

where $\tilde{\epsilon}_{\mathbf{k}, q} = E_{\mathbf{k}, q} - \epsilon_{\mathbf{k}, \uparrow} + \epsilon_{\mathbf{k}, \downarrow} - \frac{k_c^2}{2m} - (N - 1)\hbar\omega_0$ with $\epsilon_k = \frac{k^2}{2m}$. As indicated in the context, the lowest $E_{\mathbf{k}, q}$ defines the threshold value $E_0$ of the unbound polariton continuum. Correspondingly, the wave function of each $E_{\mathbf{k}, q}$ is given by

$$\psi_{\mathbf{k}, q}^{\pm} = \frac{1}{\sqrt{C}} \psi_{\mathbf{k}, q}^{\pm},$$

where $\psi_{\mathbf{k}, q}^{\pm} = \frac{1}{\sqrt{2}}(\psi_{\mathbf{k}, q}^{\uparrow \downarrow} - \psi_{\mathbf{k}, q}^{\downarrow \uparrow})$ and $\psi_{\mathbf{k}, q}^{\uparrow \downarrow} = \frac{1}{\sqrt{2}}(\psi_{\mathbf{k}, q}^{\uparrow} + \psi_{\mathbf{k}, q}^{\downarrow})$, and $C$ is the normalization constant such that

$$|\psi_{\mathbf{k}, q}^{\uparrow \downarrow}|^2 + |\psi_{\mathbf{k}, q}^{\downarrow \uparrow}|^2 + |\psi_{\mathbf{k}, q}^{\uparrow}|^2 + |\psi_{\mathbf{k}, q}^{\downarrow}|^2 = 1.$$
the interaction term $\hat{H}_{\text{int}}$ is included. It can be straightforwardly shown that $\hat{H}|\Phi_{k,q}\rangle_G = (\hat{H}_0 + \hat{H}_{\text{int}})|\Phi_{k,q}\rangle_G = \hat{H}_0|\Phi_{k,q}\rangle_G = E_{k,q}|\Phi_{k,q}\rangle_G$.

To determine the COM momentum $q_{\text{UP}}$ of the ground state at $\delta = 0$, we first set $k_z = 0$ in Eq. (A3), which results in a cubic equation of $E_{0,q_i}$: $E_{0,q_i}(E_{0,q_i} - \frac{k_{zc}^2}{m})(E_{0,q_i} - \frac{k_{zc}^2}{m}) = (4N - 2)\delta_0^2 E_{0,q_i} - g_0^2 \frac{k_{zc}^2}{m}$. Solving $E_{0,q_i}$ from above $E_{0,q_i}$, and further minimizing it with respect to $q_z$, one can obtain $q_{\text{UP}}$. There are two limits (discussed in the main text): (1) for small $g_0$, $E_{0,q_i}/E_c \simeq \frac{1}{2}\frac{(q_z/k_z)^2}{(q_z/k_z)^2} + (4N - 2)(g_0/E_c)^2 - \frac{g_0^2}{4N - 2}(q_z/k_z)^2 - \frac{1}{4N - 2}(g_0/E_c^2) + O(g_0^{-1})$, $q_{\text{UP}} \simeq \frac{k_z}{\sqrt{4N - 2}}$.

**APPENDIX B: THE BOUND POLARITON**

For given excitation number $N$, the wave function of a bound polariton (BP) can be generally constructed as

$$\Psi_q = \sum_k \Phi_{k,q} = \sum_k \{\psi_{k,q}^{\uparrow\uparrow}|\Phi_{k,q}\rangle|N - 1\rangle + \psi_{k,q}^{\uparrow\downarrow}|\Phi_{k,q}\rangle|N - 2\rangle + \varphi_{k,q}^{\uparrow\downarrow}|\Phi_{k,q}\rangle|N\rangle\},$$

where $q$ is the COM momentum and the summation over the relative momentum $\sum_k$ is limited to $k_z > 0$. Substituting $\Psi_q$ into the Schrödinger equation $\hat{H}|\Psi_q\rangle = E_q|\Psi_q\rangle$ and writing it explicitly, we obtain

$$\{E_q - (\epsilon_{\frac{3}{2} + \frac{k_z}{2}} + \epsilon_{\frac{1}{2} - \frac{k_z}{2}}) - (N - 1)\omega_c\} \psi_{k,q}^{\uparrow\uparrow} = \frac{U}{V} \sum_k (\psi_{k,q}^{\uparrow\downarrow} - \varphi_{k,q}^{\uparrow\downarrow}) + g_0 \sqrt{N} \psi_{k,q}^{\uparrow\uparrow} + g_0 \sqrt{N - 1} \varphi_{k,q}^{\uparrow\downarrow},$$

$$\{E_q - (\epsilon_{\frac{3}{2} + \frac{k_z}{2}} + \epsilon_{\frac{1}{2} - \frac{k_z}{2}}) - (N - 1)\omega_c\} \psi_{k,q}^{\uparrow\downarrow} = \frac{U}{V} \sum_k (\psi_{k,q}^{\uparrow\uparrow} - \varphi_{k,q}^{\uparrow\downarrow}) + g_0 \sqrt{N} \psi_{k,q}^{\uparrow\downarrow} + g_0 \sqrt{N - 1} \varphi_{k,q}^{\uparrow\downarrow},$$

$$\{E_q - 2(\epsilon_{\frac{3}{2} + \frac{k_z}{2}} + \epsilon_{\frac{1}{2} - \frac{k_z}{2}}) - (N - 1)\omega_c\} \varphi_{k,q}^{\uparrow\downarrow} = g_0 \sqrt{N - 1}(\psi_{k,q}^{\uparrow\downarrow} + \varphi_{k,q}^{\uparrow\downarrow}),$$

Introducing $E'_{k,q} = E_q - \epsilon_{\frac{3}{2} + \frac{k_z}{2}} - \epsilon_{\frac{1}{2} - \frac{k_z}{2}} - \frac{\epsilon_0}{\omega_c} - (N - 1)\hbar \omega_c$, and defining $\psi_q = \frac{1}{\sqrt{2}}(\psi_{k,q}^{\uparrow\downarrow} - \varphi_{k,q}^{\uparrow\downarrow})$ and $\varphi_q = \frac{1}{\sqrt{2}}(\psi_{k,q}^{\uparrow\downarrow} + \varphi_{k,q}^{\uparrow\downarrow})$, Eqs. (9)–(12) turn to

$$E'_{k,q} \psi_q^{k,q} = \frac{k_z k_0}{m} \psi_q^{k,q} = \sqrt{2}g_0 \sqrt{N} \psi_{k,q}^{\uparrow\downarrow} + \sqrt{2}g_0 \sqrt{N - 1} \varphi_{k,q}^{\uparrow\downarrow},$$

$$E'_{k,q} \varphi_q^{k,q} = \frac{k_z k_0}{m} \varphi_q^{k,q} = \frac{2U}{V} \sum_k \varphi_q^{k,q},$$

$$\{E'_{k,q} - \delta - \frac{k_z q_z}{2m}\} \psi_{k,q}^{\uparrow\downarrow} = \sqrt{2}g_0 \sqrt{N - 1} \psi_{k,q}^{\uparrow\downarrow},$$

$$\{E'_{k,q} + \delta + \frac{k_z q_z}{2m}\} \varphi_{k,q}^{\uparrow\downarrow} = \sqrt{2}g_0 \sqrt{N} \psi_{k,q}^{\uparrow\downarrow}.$$

From the above equations, a nontrivial bound-state solution $E_q$ (derived from the coefficient of singlet $\varphi_q$) can be found, which satisfies the following self-consistent equation (as in the context):

$$\frac{m}{4\pi \hbar^2 \alpha_c} = \frac{1}{V} \sum_k \left(\frac{E'_{k,q} - \frac{k_z^2 k_0^2}{m^2}}{E'_{k,q} - \frac{2Ng_0^2}{\epsilon_{k,q} + \delta + k_z q_z/2m} - \frac{2(N - 1)g_0^2}{\epsilon_{k,q} - \delta - k_z q_z/2m}} \right)^{-1} + \frac{1}{2\epsilon_0}.$$

The ground BP state is determined by further minimizing $E_q$ with respect to the COM momentum $q$.

Once the energy $E_q$ is solved from Eq. (B10), one can obtain the wave function $\Psi_q$ straightforwardly, with the coefficients given by

$$\psi_q^{k,q} = \frac{1}{\sqrt{C_x}} \left[ E'_{k,q} - \frac{k_z^2 k_0^2}{m^2} \right]^{-1} \left[ E'_{k,q} - \frac{2Ng_0^2}{\epsilon_{k,q} + \delta + k_z q_z/2m} - \frac{2(N - 1)g_0^2}{\epsilon_{k,q} - \delta - k_z q_z/2m} \right]^{-1} \psi_q^{k,q},$$

$$\varphi_q^{k,q} = \frac{k_z k_0}{m} \left( E'_{k,q} - \frac{2Ng_0^2}{\epsilon_{k,q} + \delta + k_z q_z/2m} - \frac{2(N - 1)g_0^2}{\epsilon_{k,q} - \delta - k_z q_z/2m} \right) \psi_q^{k,q}.$$
\[ \varphi_{k,q}^{\uparrow} = \sqrt{\frac{2g_0}{C'_k}} \varphi_{k,q} \]

where constant \( C' \) is the normalization constant such that \( \sum_k (|\varphi_{k,q}^{\uparrow}|^2 + |\varphi_{k,q}^{\downarrow}|^2 + \varphi_{k,q}^{\uparrow\downarrow}) = 1 \). Apparently, the coefficients of the above triplet components are asymmetric and depend on the excitation number \( N \) and the COM momentum \( q \).

The photon population in the BP state is then given by \( n_{\text{ph}} = (N - 1) + \sum_k (|\varphi_{k,q}^{\uparrow}|^2 - |\varphi_{k,q}^{\downarrow}|^2). \) And the spin-singlet population plotted in Fig. 3(d) in the main text is defined as \( |\varphi_s|^2 = \sum_k |\varphi_{k,q}|^2. \)

**APPENDIX C: ENTANGLEMENT ENTROPY**

The von Neumann entanglement entropy for a bipartite system \( H_{AB} (= H_A \otimes H_B) \) is defined as

\[ S_A \equiv -tr_B \rho_A \ln \rho_A. \]

Here, \( \rho_A = tr_B \rho \) is the reduced density matrix of subsystem \( A \) by tracing all degrees of freedom in subsystem \( B \). Notice that for a pure state, \( S_A = S_B \). In our case, the total system is comprised of the atom and photon part, and exhibits nontrivial entanglement properties between them. For the bound polariton, \( \rho_{\text{BP}} = \langle \Phi_{k,q} | \Phi_{k,q} \rangle \), and

\[ S_{\text{BP}} = - \sum_k \left( |\varphi_{k,q}^{\uparrow}|^2 + |\varphi_{k,q}^{\downarrow}|^2 \right) \ln \left( |\varphi_{k,q}^{\uparrow}|^2 + |\varphi_{k,q}^{\downarrow}|^2 \right) + \left( \sum_k |\varphi_{k,q}^{\uparrow\downarrow}|^2 \ln \left( |\varphi_{k,q}^{\uparrow\downarrow}|^2 \right) \right) \right). \]

For the unbound polariton continuum, \( \rho_{\text{UP}} = |\Phi_{k,q} \rangle \langle \Phi_{k,q}| \), and

\[ S_{\text{UP}} = - \sum_k \left( |\varphi_{k,q}^{\uparrow}|^2 + |\varphi_{k,q}^{\downarrow}|^2 \right) \ln \left( |\varphi_{k,q}^{\uparrow}|^2 + |\varphi_{k,q}^{\downarrow}|^2 \right) + \left( \sum_k |\varphi_{k,q}^{\uparrow\downarrow}|^2 \ln \left( |\varphi_{k,q}^{\uparrow\downarrow}|^2 \right) \right) \right). \]