Two-photon absorption and emission by Rydberg atoms in coupled cavities

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We study the dynamics of a system composed of two coupled cavities, each containing a single Rydberg atom. The interplay between Rydberg-Rydberg interaction and photon hopping enables the transition of the atoms from the collective ground state to the double Rydberg excitation state by individually interacting with the hybrid cavity modes and suppressing the up conversion process between them. The atomic transition is accompanied by the two-photon absorption and emission of the hybrid modes. Since the energy level structure of the atom-cavity system is photon number dependent, there is only a pair of states being in the two-photon resonance. Therefore, the system can act as a quantum nonlinear absorption filter through the nonclassical quantum process, converting coherent light field into a non-classical state. Meanwhile, the vacuum field in the cavity inspires the Rydberg atoms to simultaneously emit two photons into the hybrid mode, resulting in obvious emission enhancement of the mode.

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The generation of nonclassical states of light has been a central topic in quantum optics since the first demonstration of squeezed states of light [1]. Quantum field in nonclassical states reveals their nonclassical properties by exhibiting photon anti-bunching, sub-Poisson photon-number statistics, and clearly negative values of the Wigner function [2]. These states can be used for understanding of quantum fluctuations beyond the standard quantum noise limit and are essential sources in optical science and engineering [3]. In this context, the two-photon process, namely, the atoms transit from one energy level to another through an intermediate energy level that simultaneously involves two photons of the same frequency (the degenerate two-photon transition) or of different frequencies (the nondegenerate two-photon transition), has attracted great interest because it provides great opportunity for producing light with nonclassical properties [4]. Indeed, the two-photon absorption and emission are inherently nonclassical effects, which are expected to have potential applications in the realm of quantum techniques [5, 4].

Recently, high-finesse optical cavity has been used to couple Rydberg atoms with quantized cavity modes, which presents potential applications in studying photon nonlinearity and many-body physics [3, 10]. Neutral atoms excited by laser beams and the cavity field to high-lying Rydberg states can interact through strong and long-range dipole-dipole or van der Waals interaction [11]. The quantum anharmonicity of the energy level structure of the atom-cavity system enables the study of two-photon absorption and three-photon absorption from a probe beam [8]. The optical nonlinearity has been experimentally explored with strongly interacting Rydberg atoms in cavities [12], even at the level of individual quanta [13]. Moreover, Zhang et al. have shown that coupling of optical cavity to a lattice of Rydberg atoms can be described by the Dicke model, the competition between the atom-atom interaction and atom-light coupling can induce a novel superradiant solid phase [9]. On the other hand, rich quantum dynamics has been found in the coupling of the coupled cavities with neutral atoms [14, 15]. Its potential applications include realization of paradigmatic many-body models, such as the Bose-Hubbard and the anisotropic Heisenberg models [16].

Combine coupled cavities with interacting Rydberg atoms, a physical model in analogous to the quantum dot-cavity coupling system, where the cavity mode can be tuned to resonantly drive the two-photon transition between the ground and the biexciton states, while the exciton states are far-off resonance due to the biexciton binding energy [17], will be discussed here. In the paper, we study the two-photon absorption and emission process with two Rydberg atoms separately trapped in coupled cavities. There exists two hybrid optical modes due to the photon hopping between the two cavities. The collectively excited energy level of the Rydberg atoms is shifted up or down according to the sign of the Förster defects, which induces the two-photon resonant atomic transitions for either hybrid modes. In result, the blockade of simultaneous excitation to the Rydberg state fails due to the photon dynamics. The resonant transition frequency between the collective ground state and the double Rydberg excitation state is photon number dependent, leading to varied two-photon absorption rate for different states of the cavity modes. The system can be used for realization of quantum nonclassical processes and preparation of two-photon states. The results are discussed in the context of micro-cavities, however, the phenomenon may be found in other hybrid systems, such as Rydberg atoms interacting with superconducting microwave devices [18, 19].

Consider the system composed of two coupled cavities, each containing a Rydberg atom. The atoms have three relevant energy levels. As shown in Fig. 1, the transition from $5S_{1/2}$ atomic ground state denoted by $|g\rangle$ couples to a Rydberg excited level $|r\rangle$ through a two-photon pro-
and a frequency calibration between the left and right cavity leads to two hybrid delocalized modes $c_1$ and $c_2$, with the bare frequency separated by $2J$. (b) Effective model for heralded two-photon transition between atomic collective states $|G\rangle$ and $|R\rangle$ through the intermediate symmetric and antisymmetric entangled states $|S\rangle$ and $|A\rangle$. The $c_1$ mode is in two-photon resonance with the $|G\rangle \leftrightarrow |R\rangle$ transition for $V_{dd} = 2J$, while the $c_2$ mode is red detuned by $4J$. For $V_{dd} = -2J$, the situation is in reverse.

process via the $5P_{3/2}$ intermediate state $|e\rangle$ [8]. The bare energies for the corresponding energy levels are $\hbar \omega_g$, $\hbar \omega_c$ and $\hbar \omega_e$, respectively. The atomic transitions $|g\rangle \leftrightarrow |e\rangle$ and $|e\rangle \leftrightarrow |r\rangle$ are coupled to quantized cavity field of frequency $\omega$ and laser field of frequency $\omega_e$ with Rabi frequency $g$ and $\Omega$, respectively. The cavity field is detuned by $\delta$ to the blue of the $|g\rangle \leftrightarrow |e\rangle$ transition, and laser beam is detuned by $\delta$ to the red of the $|e\rangle \leftrightarrow |r\rangle$ transition. The energy shift $V_{dd}$ for the collective atomic state $|r\rangle_1|r\rangle_2$ stemming from the Rydberg-Rydberg interaction prevents the simultaneous excitation of the atoms to the Rydberg state $|r\rangle$. Photons can hop between the left and right cavities with the rate $J$, giving rise to a couple of hybrid optical modes with the frequencies $\omega \pm J$. The Hamiltonian for the coupled atom-cavity system in the rotating wave approximation (RWA) reads (assuming $\hbar = 1$)

$$H = H_c + H_a + H_{af},$$

with

$$H_c = \omega (a_1^\dagger a_1 + a_2^\dagger a_2) + J(a_1^\dagger a_2 + a_1 a_2^\dagger),$$

$$H_a = \sum_{k=1,2} \left( \omega_g |g\rangle_k \langle g| + \omega_c |e\rangle_k \langle e| + \omega_r |r\rangle_k \langle r| \right)$$

$$+ V_{dd} |r\rangle_1 |r\rangle_2 |22\rangle |1\rangle_1 |r\rangle,$$

and

$$H_{af} = \sum_{k=1,2} (g|e\rangle_k \langle g| a_k + \Omega \epsilon_{kc} |r\rangle_k \langle e|) + h.c.,$$

where $a_k(k = 1, 2)$ are annihilation operators for cavity fields 1 and 2, respectively. We have assumed that the coupling strengths of the two atoms interacting with the respective local cavity modes and laser beams are real and identical for simplicity. In the large detuning regime, i.e., $\delta \gg \Omega, g$, the intermediate state $|e\rangle$ will not be populated and can be adiabatically eliminated. Thus, we have an effective Hamiltonian, which, in the interaction picture, is given by

$$H_I = H_c' + H_a' + H_{af}',$$

with

$$H_c' = J(a_1^\dagger a_2 + a_1 a_2^\dagger),$$

$$H_a' = V_{dd} |r\rangle_1 |r\rangle_2 |22\rangle |1\rangle_1 |r\rangle,$$

and

$$H_{af}' = (\lambda \sum_{k=1,2} (|r\rangle_k |g\rangle_k + h.c.) + \lambda' \sum_{k=1,2} a_k^\dagger a_k |g\rangle_k \langle g|$$

$$+ \lambda'' \sum_{k=1,2} |r\rangle_k \langle r|,$$

where $\lambda = \Omega g/\delta$, $\lambda' = g^2/\delta$, and $\lambda'' = \Omega^2/\delta$.

For taking account of the Rydberg-Rydberg interaction between the atoms, it is convenient to rewrite the atom-cavity interaction in terms of the two-atom collective states $\{|G\rangle = |g\rangle_1 |g\rangle_2, |R\rangle = |r\rangle_1 |r\rangle_2, |S\rangle = (|g\rangle_1 |r\rangle_2 + |r\rangle_1 |g\rangle_2)/\sqrt{2}, |A\rangle = (|g\rangle_1 |r\rangle_2 - |r\rangle_1 |g\rangle_2)/\sqrt{2}\}$. (b) Effective model for heralded two-photon transition between atomic collective states $|G\rangle$ and $|R\rangle$ through the intermediate symmetric and antisymmetric entangled states $|S\rangle$ and $|A\rangle$. The $c_1$ mode is in two-photon resonance with the $|G\rangle \leftrightarrow |R\rangle$ transition for $V_{dd} = 2J$, while the $c_2$ mode is red detuned by $4J$. For $V_{dd} = -2J$, the situation is in reverse. For the taking account of the Rydberg-Rydberg interaction between the atoms, it is convenient to rewrite the atom-cavity interaction in terms of the two-atom collective states $\{|G\rangle = |g\rangle_1 |g\rangle_2, |R\rangle = |r\rangle_1 |r\rangle_2, |S\rangle = (|g\rangle_1 |r\rangle_2 + |r\rangle_1 |g\rangle_2)/\sqrt{2}, |A\rangle = (|g\rangle_1 |r\rangle_2 - |r\rangle_1 |g\rangle_2)/\sqrt{2}\}$. (b) Effective model for heralded two-photon transition between atomic collective states $|G\rangle$ and $|R\rangle$ through the intermediate symmetric and antisymmetric entangled states $|S\rangle$ and $|A\rangle$. The $c_1$ mode is in two-photon resonance with the $|G\rangle \leftrightarrow |R\rangle$ transition for $V_{dd} = 2J$, while the $c_2$ mode is red detuned by $4J$. For $V_{dd} = -2J$, the situation is in reverse.

$$H_I = H_c' + H_a' + H_{af}'$$

with

$$H_c' = J(c_1^\dagger c_1 - c_2^\dagger c_2),$$

$$H_a' = V_{dd} |R\rangle \langle R|,$$

and

$$H_{af}' = (\lambda c_1 (|S\rangle \langle G| + |R\rangle \langle S|) + \lambda c_2 (|A\rangle \langle G| - |R\rangle \langle A|)$$

$$+ \lambda' (c_1^\dagger c_2 + c_2^\dagger c_1) |S\rangle \langle A| + h.c.]$$

$$+ (\lambda'' (c_1^\dagger c_1 + c_2^\dagger c_2 + \lambda''') (|S\rangle \langle S| + |A\rangle \langle A|)$$

$$+ \lambda' (c_1^\dagger c_1 + c_2^\dagger c_2) |G\rangle \langle G| + 2\lambda'' |R\rangle \langle R|.}
with
\[
H_{tr} = (\lambda c_1 e^{-iJt} |S\rangle \langle G| + \lambda c_2 e^{iJt} |A\rangle \langle G| + h.c.) \\
+ (\lambda c_1 e^{i(V_{dd} - J)t} |R\rangle \langle S| - \lambda c_2 e^{iV_{dd} + Jt} |R\rangle \langle A| + h.c.) \\
+ (\lambda c_1^2 e^{2Jt} + c_1^2 c_2 e^{-2Jt}) |S\rangle \langle A| + h.c.,
\]
and
\[
H_{st} = (\lambda^2 c_1^2 + c_2^2 |S\rangle \langle |G| + 2\lambda'' |R\rangle \langle |R|).
\]

(i) Without considering the photon number dependent stark shifts \(H_{st}\). The hybrid mode \(c_1\) is blue-detuned by \(J\) from the transition \(|G\rangle \leftrightarrow |S\rangle\) and \(c_1\) is red-detuned by \(J\) from the transition \(|G\rangle \leftrightarrow |A\rangle\), see Fig. 1(b).

To decouple the transition channels \(|G\rangle \rightarrow |S\rangle \rightarrow |R\rangle\) and \(|G\rangle \rightarrow |A\rangle \rightarrow |R\rangle\), the optical frequency up conversion should be suppressed. This can be met if the frequency separation of the hybrid modes is much greater than the conversion rate (dispersive regime), i.e., \(2J \gg \sqrt{n_{c1} n_{c2}}\) \(\lambda/2\). Note that the sign of the Rydberg-Rydberg interaction strength is determined by the sign of the energy gap in the Förster process [20]. Now if \(V_{dd} = 2J\), the atomic transition \(|G\rangle \leftrightarrow |R\rangle\) mediated by symmetric entangled state \(|S\rangle\) is in resonance with twice the photon frequency of the hybrid mode \(c_1\), while the other channel \(|G\rangle \rightarrow |A\rangle \rightarrow |R\rangle\) is out of resonance and is detuned by \(4J\). Therefore, under the condition \(J \gg \sqrt{n_{c1} n_{c2}}\) \(\lambda\), we can finally obtain an effective Hamiltonian by using the time averaging approach to describe this two-photon transition process [21],

\[
H_{eff} = \xi(|G\rangle \langle R| c_1^2 + |R\rangle \langle G| c_2^2),
\]

where \(\xi = \lambda^2 / J\), and the stark shift terms \((\lambda^2 / J) (|c_1^2 c_1 - c_2^2 c_2| G\rangle \langle + R\rangle / |R\rangle c_1^1 c_2^1 c_2 / 3)\) have been neglected because they are much less than the photon number dependent energy \(H_{st}\). While if \(V_{dd} = -2J\), the transition channel \(|G\rangle \leftrightarrow |R\rangle\) mediated by the singlet state \(|A\rangle\) is in resonant with twice the frequency of the hybrid mode \(c_2\), and the channel \(|G\rangle \rightarrow |S\rangle \rightarrow |R\rangle\) related to \(c_1\) is out of resonance. The effective Hamiltonian is then otherwise given by

\[
H_{eff} = \xi(|G\rangle \langle R| c_2^2 + |R\rangle \langle G| c_1^2).
\]

Thus, the blockade of the double Rydberg excitations may be wrecked due to the photon hopping through two-photon absorption. (ii) Two-photon transition including \(H_{st}\). Taking stark shift \(H_{st}\) into consider, the two-photon resonant transition may break down if \(|G\rangle\) and \(|R\rangle\) are shifted by different amount depending on the photon number of hybrid modes. Set \(\Omega = g\), this implies the two-photon resonance condition \(V_{dd} = 2J + (\langle n_{c1} \rangle - 2)\lambda\) and \(V_{dd} = -2J + (\langle n_{c2} \rangle - 2)\lambda\) for \(c_1\) mode and \(c_2\) mode, respectively, when the atoms are initially in the ground state...
Without loss of generality, we focus on the two-photon dynamics discussed above can be analytically calculated for \( |\xi\rangle \) photons (Fig. 2(c) and 2(f)). Note that the effective couplings are shifted according to the photon number dependent Stark shifts described by \( H_{st} \). Other parameters as in Fig. 2.

The coherent quantum dynamics in this atom-cavity coupled system can be read from Fig. 2 in which we have shown the time-dependent population of the collective atomic states with the system initial state \(|G\rangle \otimes |1\rangle_{a1} |1\rangle_{a2} \). In terms of the delocalized hybrid modes, the initial state of the cavity fields can be rewritten as:

\[
|a_1 a_2 0 0\rangle = \frac{1}{\sqrt{2}} (|c_1^+\rangle + |c_2^+\rangle) (|c_1^-\rangle - |c_2^-\rangle) |0\rangle_{c1} |0\rangle_{c2} = (|2\rangle_{c1} |0\rangle_{c2} - |0\rangle_{c1} |2\rangle_{c2})/\sqrt{2}.
\]

In this case, the energy shifts for \(|G\rangle\) and \(|R\rangle\) are both 2\(\lambda\), which guarantee the two-photon resonance condition. In Fig. 2(a) and 2(d), the Rabi oscillation between \(|G\rangle\) and \(|R\rangle\) clearly demonstrates the photon absorption and emission processes, and the excitation of the symmetric and anti-symmetric entangled states are well suppressed. The probability for detecting \(|R\rangle\) can only reach 0.5 or so in each plot because the transition channels via the intermediate states \(|S\rangle\) and \(|A\rangle\) are selected by the sign of the energy shift \(V_{dd}\). In this process, the photon dynamics of the localized modes \(a_1\) and \(a_2\) display exactly the same behavior and remain symmetric. It means that the two localized photons are absorbed and emitted simultaneously all the time (see Fig. 2b) and 2e). In contrast, the coupling to the hybrid modes are symmetry breaking. Either the transition between \(|G\rangle \otimes |2\rangle_{c1} |0\rangle_{c2}\) and \(|R\rangle \otimes |0\rangle_{c1} |2\rangle_{c2}\) with \(V_{dd} = 2J\) or that between \(|G\rangle \otimes |0\rangle_{c1} |2\rangle_{c2}\) and \(|R\rangle \otimes |0\rangle_{c1} |2\rangle_{c2}\) with \(V_{dd} = -2J\) happens. That is accompanied by the atom absorption and emission of two delocalized photons (Fig. 2c and 2f). Note that the effective coupling strength should be revised by \(\xi' = \lambda^2/(J + \lambda/2)\) for \(V_{dd} = \pm 2J\) due to the energy shifts of \(|S\rangle\) and \(|A\rangle\), which leads to the slightly different time period of oscillation for \(|n_{c1}\rangle\) and \(|n_{c2}\rangle\).

The time evolution of the system dynamics discussed above can be analytically calculated by solving the Schrödinger equation \(i\hbar \dot{\psi}(t) = H_{eff} \psi(t)\). Without loss of generality, we focus on the two-photon transition with respect to the delocalized mode \(c_1\) described by Eq. (6), from which we can obtain the quantum state of the system at time \(t\),

\[
\psi(t) = \frac{1}{\sqrt{2}} (C_g(t) |G\rangle \otimes |2\rangle_{c1} |0\rangle_{c2} + C_r(t) |R\rangle \otimes |0\rangle_{c1} |2\rangle_{c2})
\]

\[
-\frac{1}{\sqrt{2}} (|G\rangle \otimes |0\rangle_{c1} |2\rangle_{c2}).
\]

In addition, by appropriately choosing the interaction time, the system will evolve from \(|G\rangle \otimes \sum |n_{c1}\rangle |0\rangle_{c2}\) to \(|R\rangle \otimes \sum |n_{c2}\rangle |c_1\rangle_{c2}\) onto \(|G\rangle \otimes \sum |n_{c1}\rangle |0\rangle_{c2}\) and \(|R\rangle \otimes \sum |n_{c2}\rangle |c_1\rangle_{c2}\). This is a two-photon NOON state for localized modes.

The physical model can be further understood by looking into the time averaged photon absorption \(n_{c1}(t) = \langle \xi(t)|c_1\rangle_{c1}(0)|c_1\rangle_{c1}(t)\) as a function of the Rydberg-Rydberg interaction strength for the initial state \(|G\rangle \otimes \sum |n_{c1}\rangle |0\rangle_{c2}\) and \(|G\rangle \otimes \sum |0\rangle_{c1} |n_{c2}\rangle_{c2}\) that are illustrated by \(H_{st}\). For \(V_{dd} \approx 2J\) or \(V_{dd} \approx -2J\), only when \(n_{c1} = 2\) and \(n_{c2} = 0\), or \(n_{c1} = 0\) and \(n_{c2} = 2\), the interplay between Rydberg-Rydberg interaction and photon tunneling will cause absorption of two photons from the hybrid modes. The resonant two-photon transition gives time-averaged photon absorption of one. For the initial state being \(|G\rangle \otimes |n_{c1}\rangle |0\rangle_{c2}\) and \(n_{c1} \neq 2\), this corresponds to the dispersive interaction regime because the \(|G\rangle \leftrightarrow |R\rangle\) transition is detuned by \((n_{c1} - 2)\lambda\), which is much larger than the effective \(|G\rangle \leftrightarrow |R\rangle\) coupling strength \(\sqrt{4\lambda^2}(n_{c1} - 1)\xi\). As shown in Fig. 3a, the photon absorption becomes weaker and weaker as the initial photon number of \(c_1\) mode increases. It is also interesting to study the time averaged photon emission \(n_{c1}(t)\) or \(-n_{c1}(0)\) with the system initial state \(|R\rangle \otimes |n_{c1}\rangle\) (see Fig. 3b)). Here, the atoms are both initially in the Rydberg excited state. The atomic transition \(|R\rangle \rightarrow |G\rangle\) regularly happens accompanied by simultaneously emitting two photons for the hybrid mode being in the vacuum state, giving rise to emission enhancement of the mode.
Fig. 5. Time dependent population of the collective atomic states and mean photon number for hybrid optical modes $c_1$ and $c_2$ for initially the atoms in the state $|G\rangle$ and the cavity modes in the coherent states $|\alpha\rangle_{a1}|\beta\rangle_{a2} = |(\alpha + \beta)/\sqrt{2}\rangle_{c1}|(\alpha - \beta)/\sqrt{2}\rangle_{c2}$, where $\alpha = \beta = 1/\sqrt{2}$. The interaction of the atoms with cavity fields is mainly dominated by the $|G\rangle \otimes |0\rangle_{c1} \rightarrow |R\rangle \otimes |0\rangle_{c1}$ transition. The oscillation amplitude is limited by the probability amplitude of $|2\rangle_c$ in the expansion of coherent state in the Fock space.

In Fig. 6(a), we consider the localized cavity fields that are initially in the coherent states $|\alpha\rangle_{a1}$ and $|\beta\rangle_{a2}$ respectively. The quantum state of the localized two-mode field can be rewritten as $|\alpha\rangle_{a1}|\beta\rangle_{a2} = |(\alpha + \beta)/\sqrt{2}\rangle_{c1}|(\alpha - \beta)/\sqrt{2}\rangle_{c2}$ in terms of the hybrid modes $c_1$ and $c_2$, which are the coherent states of mean photon number $\langle N_{c1} \rangle = |\alpha + \beta|^2/2$ and $\langle N_{c2} \rangle = |\alpha - \beta|^2/2$. For $\alpha = \beta$, the $c_2$ mode is in the vacuum state. Thus, if the atoms are both in the ground state $|g\rangle$ and the Rydberg-Rydberg interaction strength is $V_{dd} = 2J$, only the transition channel $|G\rangle \rightarrow |S\rangle \rightarrow |R\rangle$ will be open for the coupling of $|G\rangle$ with $|R\rangle$. We can then simply focus on the photon dynamics of the $c_1$ mode. To study the $c_2$ mode, we can alternatively set $\alpha = -\beta$. Without loss of generality, we will assume $\alpha = \beta$ in the following. The $c_1$ mode can be expanded in the Fock state representation as $|(\alpha + \beta)/\sqrt{2}\rangle_{c1} = e^{i\pi/2}\sum_{n_{c1}}((\alpha + \beta)/\sqrt{2})^n|n_{c1}\rangle_{c1}$. As discussed above, the effective Hamiltonian in Eq. 3 holds only when there are two photons in $c_1$ mode. Otherwise, the collective atomic states $|G\rangle$ and $|R\rangle$ will be shifted by different amount and the $|G\rangle \leftrightarrow |R\rangle$ transition is thus out of resonance. This can be used for demonstration of the quantum nonclassical process [24], where a coherent state will be transformed to a nonclassical state. The coherent dynamics of the Rydberg atoms interacting with coherent cavity fields is shown in Fig. 5. The oscillation is mainly due to the interplay of $|G\rangle \otimes |0\rangle_{c1}|2\rangle_{c2}$ with $|R\rangle \otimes |0\rangle_{c1}|0\rangle_{c2}$ weakly perturbed by $|G\rangle \otimes |n_{c1}\rangle_{c1}|0\rangle_{c2}$ ($n_{c1} \neq 2$). Therefore, the minimum of the atomic population $P_G$ and the mean photon number of $c_1$ mode $\langle n_{c1} \rangle$ are approximately given by $1 - P_{n_{c1}=2}$ and $(1 - 2P_{n_{c1}=2})$, respectively, with $P_{n_{c1}=2}$ the probability amplitude of $|2\rangle_c$ in the expansion of coherent state in the Fock space.

Using this system, we can realize a quantum nonlinear absorption filter and the nonclassical quantum optical process defined by Rahimi-Keshari et al. [22]. For the atoms interacting with coherent cavity fields with the initial state $|G\rangle \otimes |\sqrt{2}\alpha\rangle_{a1}|0\rangle_{c2}$, the measurement of the atoms in the state $|G\rangle$ at $t = \pi/2\sqrt{2}\xi$ will collapse the hybrid mode $c_1$ into a nonclassical quantum state finally by removing the component Fock state $|2\rangle$ from coherent $c_1$ mode. The Wigner function for such kind of nonclassical states is shown in Fig. 6(a), from which we clearly see the negative value close to the origin demonstrating its nonclassical nature. The component Fock state $|2\rangle_c$ has been absorbed by the atoms being excited to $|R\rangle$. On the other hand, while the atoms initially in the collective state $|R\rangle$ interact with the coherent fields, ideally, the measurement of the atoms in the collective state $|G\rangle$ will collapse the hybrid mode $c_1$ into the Fock state $|2\rangle_c$, which is weakly influenced by the components of the other Fock states. The Wigner function of the hybrid mode $c_1$ after atomic measurement is shown in Fig. 6(b), which predicates a Fock state $|2\rangle_c$ in spite of the tiny distortion. This can probably be used for realization of two-photon source, in particular, for the cavity fields initially being in the vacuum state. Although we assumed the cavity fields are initially in the coherent states, the characteristics of current model are applicable to tailor different quantum states of light, such like thermal fields in coupled cavities.

For experimental demonstration of the two-photon absorption and emission, the parameter $\xi$ of the effective Hamiltonian should be much larger than effective decoherence rates via the photons and the excited states. Set $\Omega = g$, $\delta = 10g$, and $J = g$, then we have $\lambda = \Omega g/\delta = 0.1g$, $\xi = \lambda^2/J = 0.01g$, and the time needed for preparing nonclassical states shown in Fig. 6 is $t = \pi/2\sqrt{2}\xi \approx 1.11 \times 10^2g^{-1}$. Since the intermediate state $|e\rangle$ is off-resonantly coupled with the ground state $|g\rangle$ and the Rydberg state $|r\rangle$, the effective decay rate for $|e\rangle$ is $\gamma_e = (g^2/\delta^2 + \Omega^2/\delta^2)\gamma$ for $\delta \gg \Omega, g, \gamma$, where $\gamma$ is the spontaneous emission rate.
Rydberg state with principal quantum number $n = 70$ has a spontaneous decay rate $\gamma_{r} = 2\pi \times 0.55 \text{kHz}$ that is much smaller than $\gamma$. The cavity decay rate should fulfill the condition $\kappa \ll \xi \sim 0.01J$, which implies the photons fast tunnel to next cavity before decay into free space. These decoherence sources will induce intrinsic errors for the implementation. On the other hand, the Rydberg-Rydberg interaction arises from the intrinsic Förster interaction, which can lead to the energy shift $\nu_{dd}$ approximating to $200 \text{MHz}$ with the interatomic distance around $7 \mu m$. The sign of the energy shift is determined by the sign of Förster defects correlated with the selected transition channels. The parameter regime above can be achieved with the micro-cavities, where atom-cavity interacting system with the cooperativity factor as high as $C = g^2/2\kappa_\gamma \sim 10^5$ is predicted to be available. The micro-cavities with the size tens of $\mu m$ can be coupled via the overlap of their evanescent fields. Set $\kappa \sim 10^{-3}g$ and $\gamma \sim 10^{-3}g$, the errors of the prepared nonclassical state due to decoherence approximates to $E \sim (\gamma_{c} + \gamma_{r} + \kappa)t \sim 0.12$. Without seeing a quantum jump from the coupled cavities, the dissipative dynamics of the system can be described by the non-Hermitian Hamiltonian $H_{NH} = H_{I} - i\kappa/2(a_{1}^{\dagger}a_{1} + a_{2}^{\dagger}a_{2})$, using which we find that the success probability of the current proposal decreases according to the exponential factor $e^{-n_{c}ct}$ with $n_{c}$ the mean photon number in coupled cavities, while the Wigner function for the prepared state is only slightly changed. To improve the fidelity, an atomic ensemble acting as a two-level “superatom” can be placed inside the cavities, instead of a single Rydberg atom.

In conclusion, we have studied the interaction between Rydberg atoms and the hybrid modes in coupled cavities. The dispersive-atom-cavity interaction results in nonlinear electronic-level shifts depending on the photon-number of the hybrid modes. The Rydberg atoms can simultaneously absorb (emit) two photons from (into) one of the hybrid modes relying on the sign of the Rydberg-Rydberg interaction induced energy shift. There is only one transition channel that is in two-photon resonance, which can be used for generation of nonclassical states of light. The physical realization of this scheme can be realized with coupled micro-cavities, however, the alternative candidates of experimental setup include ultrahigh-Q coupled nano-cavity based on photonic crystals and superconducting microwave devices. This scheme promises a new avenue for manipulation of quantum state of light and realization of nonclassical quantum optical process.

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