Scaling enhancement of photon blockade in output fields

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Photon blockade enhancement is an exciting and promising subject that has been well studied for photons in cavities. However, whether photon blockade can be enhanced in the output fields remains largely unexplored. We show that photon blockade can be greatly enhanced in the mixing output field of a nonlinear cavity and an auxiliary (linear) cavity, where no direct coupling between the nonlinear and auxiliary cavities is needed. We uncover a biquadratic scaling relation between the second-order correlation of the photons in the output field and intracavity nonlinear interaction strength, in contrast to a quadratic scaling relation for the photons in a nonlinear cavity. We identify that this scaling enhancement of photon blockade in the output field is induced by the destructive interference between two of the paths for two photons passing through the two cavities. We then extend the theory to the experimentally feasible Jaynes-Cummings model consisting of a two-level system strongly coupled to one of the two uncoupled cavities, and also predict a biquadratic scaling law in the mixing output field. Our proposed scheme is general and can be extended to enhance blockade in other bosonic systems.

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I. INTRODUCTION

A single-photon resource [1–3] with simultaneously high degrees of efficiency, single-photon purity, and photon indistinguishability is a crucial device in the implementation of quantum communication [4], quantum computing [5], and quantum metrology [6]. Photon blockade, preventing the resonant injection/emission of more than one photon [7], provides an efficient way for single-photon generation with high purity. The field of photon blockade is extended to the high-order [8–12], multimode [13], and multi-dimensional [14] correlations, and its application is expanded from generating single photons to demonstrating photonic quantum logic gate [15,16] and fractional quantum Hall state [17].

Since the prediction of photon blockade [7], diverse efforts have been made to observe and enhance the effect. Strong coupling between light and matter at the single-photon level enabled the observation of photon blockade in experiments, including single atoms coupled to an optical resonator [18–20], a quantum dot coupled to a photonic crystal resonator [21,22], and a superconducting qubit coupled to a transmission line resonator [23,24]. Different from the photon blockade based on strong nonlinearity (conventional photon blockade), Liew and Savona showed that photon blockade can also be achieved with weak nonlinearity through quantum interference, known as unconventional photon blockade (UPB) [25,26]. Subsequently, UPB has been predicted in different setups [27–33], and has been observed for both optical [34] and microwave [35] photons.

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While the UPB with weak nonlinearity is interesting, the fact that there is a very small amount of photon in the cavity makes it inconvenient for applications [36]. Besides, photon blockade is also predicted by nonlinear driving [37,38] and nonlinear loss [39–42]. Moreover, photon blockade enhancement is proposed based on multimode-resonant interaction [43,44], non-Hermitian coupling [45–48], dynamical excitation [49,50], and coupled-resonator chain [51–53].

We note that the previous studies mainly focus on the photon blockade in the cavities, but photon statistics in the output fields becomes even more complex, such as we can observe photon antibunching for the reflected light but bunching for transmitted light [20,54,55]. Photon statistics for the mixing of two output channels has been investigated in Ref. [56], and it shows that the photon antibunching in the mixing output field is not suppressed but rather just displaced in a different region of the system's parameter space. Besides, photon antibunching as well as bunching effect are observed in the mixed field of a narrow band two-photon source and a coherent field [57], and tunable photon statistics have been proposed in the admixing of a coherent state with a squeezed state [58,59]. Nevertheless, whether photon blockade can be enhanced in the mixing fields output from two cavities has not been studied thoroughly.

In this paper, we combine conventional and unconventional photon blockade and show that photon blockade can be greatly enhanced in the mixing fields output from a nonlinear cavity and an auxiliary linear cavity. Different from the previous studies on UPB in weakly nonlinear photonic molecules [25–33], here the photon blockade enhancement in the output fields is achieved without direct coupling between the two cavities, which brings three advantages. Firstly,

there is no time oscillation in the temporal second-order correlation function. Secondly, the single-photon output efficiency is relatively greater. Thirdly, there is no strict relationship between the nonlinear strength and the coupling strength of the two cavities to observe optimal photon blockade.

We analytically identify that there is a biquadratic scaling relation between the second-order correlation of the photons in the output field and the intracavity nonlinear interaction strength, in contrast to a quadratic scaling law for the photons in a nonlinear cavity. Our scheme is general and can be extended to other platforms. As an example, we consider an experimentally feasible Jaynes-Cummings (JC) model for two (uncoupled) cavities with a two-level system (TLS) coupled to one of them, and demonstrate a biquadratic scaling relation between the second-order correlation of the photons in the output field and TLS-cavity interaction strength.

II. γ⁽³⁾ MODEL

Without loss of generality, we first consider the photon blockade in the mixing fields output from a cavity containing $\chi^{(3)}$ nonlinear medium and an auxiliary (linear) cavity [Fig. 1(a)]. The total Hamiltonian of the system in the frame rotating at the probe laser frequency ω_p can be written as $(\hbar = 1)$

$$H = \Delta_1 a_1^{\dagger} a_1 + U a_1^{\dagger} a_1^{\dagger} a_1 a_1 + i \varepsilon (a_1^{\dagger} - a_1) + \Delta_2 a_2^{\dagger} a_2 + i \varepsilon (a_2^{\dagger} - a_2),$$
 (1)

where a_i and a_i^{\dagger} are the annihilation and creation operators of the *i*th cavity with frequency ω_i (i=1,2), $\Delta_i=\omega_i-\omega_p$ is the laser detuning from the cavity resonance, $\delta=\omega_2-\omega_1$ is the detuning between the two cavities, U is the nonlinear interaction strength, and ε is the pumping strength on each cavity. According to the input-output relation [60], the mixing fields a_{out} and A_{out} output from the two cavities can be described by $a_{\text{out}}=(\sqrt{\kappa_1}a_1+e^{i\phi}\sqrt{\kappa_2}a_2)/\sqrt{2}-a_{\text{vac}}$ and $A_{\text{out}}=(\sqrt{\kappa_1}a_1-e^{i\phi}\sqrt{\kappa_2}a_2)/\sqrt{2}-a'_{\text{vac}}$, where κ_i is the one-sided decay rate of the *i*th cavity, ϕ is the relative phase between the two output fields (tunable by using the phase shifter), and a_{vac} (a'_{vac}) is the input vacuum field from the right-hand side of the cavities. Here, we focus on the output field a_{out} , and the results of A_{out} can be obtained just by replacing ϕ by $\phi+\pi$ (see Appendix A).

Photon statistics of the output field in the steady state can be described by the second-order correlation function

$$g_{\text{out}}^{(2)}(\tau) = \frac{\langle a_{\text{out}}^{\dagger} a_{\text{out}}^{\dagger}(\tau) a_{\text{out}}(\tau) a_{\text{out}} \rangle}{\langle a_{\text{out}}^{\dagger} a_{\text{out}} \rangle^{2}}$$

$$= \sum_{j,k,l,m=1}^{2} e^{in\phi} \sqrt{\kappa_{j} \kappa_{k} \kappa_{l} \kappa_{m}} \frac{\langle a_{j}^{\dagger} a_{k}^{\dagger}(\tau) a_{l}(\tau) a_{m} \rangle}{N_{\text{out}}^{2}}, \quad (2)$$

where n=l+m-j-k, $N_{\rm out}=\kappa_1\langle a_1^\dagger a_1\rangle+\kappa_2\langle a_2^\dagger a_2\rangle+2\sqrt{\kappa_1\kappa_2}{\rm Re}(e^{i\phi}\langle a_1^\dagger a_2\rangle)$, and τ is the time delay. Different from the second-order correlation function in the cavities $g_i^{(2)}(0)=\langle a_i^\dagger a_i^\dagger a_i a_i\rangle/\langle a_i^\dagger a_i\rangle^2$ $(i=1,2),~g_{\rm out}^{(2)}(0)$ also depends on the cross correlation between the two cavities (i.e.,

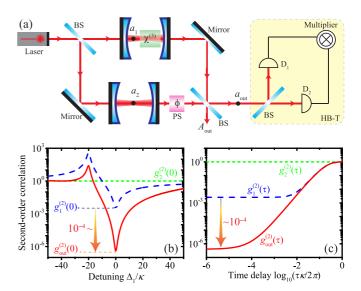


FIG. 1. (a) A Mach–Zehnder interferometer with two cavities (a_1 and a_2) in the paths. A laser is divided into two beams by a 50/50 beam splitter (BS), and they are injected into a cavity containing $\chi^{(3)}$ nonlinear medium and an auxiliary (linear) cavity, respectively. The output fields from these two cavities mix by another BS. A phase shifter (PS) is placed in one path to induce tunable phase difference ϕ between the two paths. The second-order correlation of the output field is measured by a Hanbury-Brown-Twiss (HB-T) set-up. Second-order correlations of the photons in the two cavities $[g_1^{(2)}(\tau)$ and $g_2^{(2)}(\tau)]$ and the mixing output field $[g_{\text{out}}^{(2)}(\tau)]$ are plotted (b) as functions of the detuning Δ_1/κ for the time delay $\tau=0$, and (c) as functions of $\log_{10}(\tau\kappa/2\pi)$ for $\Delta_1=0$. The parameters are $\phi=\pi$, $U=20\kappa$, and $\delta=2U$.

 $\langle a_2^{\dagger} a_2 a_1^{\dagger} a_1 \rangle$, $\langle a_1^{\dagger} a_1^{\dagger} a_2 a_2 \rangle$, $\langle a_1^{\dagger} a_1^{\dagger} a_1 a_2 \rangle$, and $\langle a_2^{\dagger} a_1^{\dagger} a_2 a_2 \rangle$), and there are phase factors $e^{in\phi}$ in front of the terms, which can be negative and induce the enhancement of photon blockade in the output field, without changing the photon statistics in the cavities.

The dynamics of the system are governed by the master equation [61] $d\rho/dt = -i[H, \rho] + \sum_{i=1,2} \kappa_i (2a_i \rho a_i^{\dagger} - a_i^{\dagger} a_i \rho - \rho a_i^{\dagger} a_i)$, where ρ is the density matrix of the system. For simplicity, here, we set $\kappa_1 = \kappa_2 = \kappa$ (the discussions on $\kappa_1 \neq \kappa_2$ are given in the Appendix B), and rescale other parameters by κ , such as $\varepsilon = \kappa/10$ for weak pumping.

III. PHOTON BLOCKADE ENHANCEMENT

To demonstrate the photon blockade enhancement more clearly, the second-order correlation functions for photons in the two cavities $g_i^{(2)}(0)$ and in the output field $g_{\text{out}}^{(2)}(0)$ are shown in Fig. 1(b). As expected, the photon statistics in the two uncoupled cavities are independent of each other: strong photon blockade $g_1^{(2)}(0) \approx 2.54 \times 10^{-3}$ in the cavity a_1 for strong enharmonicity ($U = 20\kappa$), and no photon blockade $g_2^{(2)}(0) = 1$ in the cavity a_2 without nonlinearity. Surprisingly, a much stronger photon blockade is obtained in the mixing output field a_{out} for $[g_{\text{out}}^{(2)}(0)/g_1^{(2)}(0)] \sim 10^{-4}$ at resonant frequency ($\Delta_1 = 0$). Moreover, as there is no direct coupling

between the two cavities (a_1 and a_2), there is no oscillation in $g_{\text{out}}^{(2)}(\tau)$ [Fig. 1(c)], in contrast to the rapid oscillations in photon correlations as a result of amplitude oscillation between different cavities [26].

Figure 2(a) is a color plot of $\log_{10}[g_{\text{out}}^{(2)}(0)]$ as a function of the phase ϕ/π and detuning δ/κ , for $\Delta_1=0$ and $U=20\kappa$. The minimum of $\log_{10}[g_{\text{out}}^{(2)}(0)]$ is reached for $\phi\approx\pi$ and $\delta\approx 2U$ (or $\phi\approx0$ and $\delta\approx-2U$). Two cuts taken from the color plot for $\delta=2U$ and $\phi=\pi$ are shown in Figs. 2(b) and 2(c), respectively. The photon blockade is enhanced, i.e., $g_{\text{out}}^{(2)}(0)< g_1^{(2)}(0)$, in the regime of $0.66\pi<\phi<1.33\pi$, and the minimal value of $g_{\text{out}}^{(2)}(0)$ is about 5.4 orders smaller than $g_1^{(2)}(0)$ at $\phi\approx0.996\pi$ [Fig. 2(b)]. Moreover, $g_{\text{out}}^{(2)}(0)$ also strongly depends on the detuning δ/κ between the two cavities, and it is about 3.8 orders smaller than $g_1^{(2)}(0)$ at $\delta\approx2U$ [Fig. 2(c)]. These results suggested that quantum interference might be responsible for the great enhancement of photon blockade in the output field.

In addition, the mean photon numbers in the cavities $(n_i = \langle a_i^{\dagger} a_i \rangle)$ are plotted as functions of the detuning δ/κ in Fig. 2(d). Under the optimal condition $\delta \approx 2U$, the singlephoton generation in the auxiliary cavity a_2 is suppressed seriously $(n_2/n_1 \approx 10^{-3.2})$ for large detuning $\delta \gg \kappa$. Hence, almost all of the single photons in the mixing output field are emitted from the cavity a_1 , and they are about one half of the photons emitted from cavity a_1 . Nevertheless, the auxiliary cavity a_2 provides another path for two photons passing through the whole system, which is the key ingredient for the enhancement of photon blockade in the output field as discussed bellow. By the way, the mean photon number in the auxiliary cavity a_2 is almost the same as the one in cavity a_1 $(n_1 \approx n_2)$ under the resonant condition $\delta = 0$, and they cancel each other at $\phi = \pi$ for destructive interference, which induces a strong bunching effect $[g_{\text{out}}^{(2)}(0) \gg 1]$ [30,58] in the output field [Fig. 2(c)].

IV. BIQUADRATIC SCALING

In order to understand the origin of the giant enhancement of photon blockade in the output field, we derive the expressions of the second-order correlations $[g_{\text{out}}^{(2)}(0)]$ and $g_1^{(2)}(0)]$ analytically (see Appendix B). Here, including the effect of optical decay, an effective Hamiltonian $H_{\text{eff}} = H - i\kappa(a_1^{\dagger}a_1 + a_2^{\dagger}a_2)$ is introduced according to the quantum-trajectory method [62]. Under weak driving condition ($\varepsilon \ll \kappa$), the wave function on a Fock-state basis can be truncated to the two-photon manifold as $|\psi\rangle = \sum_{n_2=0}^{2-n_1} \sum_{n_1=0}^2 C_{n_1 n_2} |n_1, n_2\rangle$. Here, $|n_1, n_2\rangle$ represents the Fock state of n_1 photons in cavity a_1 and a_2 photons in cavity a_2 , with the probability amplitude $C_{n_1 n_2}$. The expression of $C_{n_1 n_2}$ can be obtained by solving the Shrödinger equation $d|\psi\rangle/dt = -iH_{\text{eff}}|\psi\rangle$ in the steady states.

Under the conditions for photon blockade enhancement ($\phi = \pi$, $\Delta_1 = 0$, and $\delta = 2U \gg \kappa$), the second-order correlation function can be written as

$$g_{\text{out}}^{(2)}(0) \approx \frac{2\kappa^2}{N_{\text{out}}^2} \{ |C_{20} - \sqrt{2}C_{11}|^2 + |C_{02}|^2 - 2\text{Re}[(\sqrt{2}C_{11}^* - C_{20}^*)C_{02}] \},$$
(3)

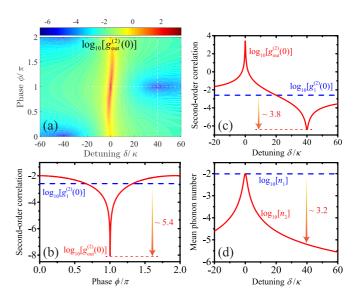


FIG. 2. (a) The second-order correlation $\log_{10}[g_{\text{out}}^{(2)}(0)]$ for different phase ϕ/π and detuning δ/κ . The second-order correlations $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ vs (b) phase ϕ/π with $\delta=2U$ and (c) detuning δ/κ with $\phi=\pi$. (d) The mean photon number $\log_{10}(n_1)$ and $\log_{10}(n_2)$ vs detuning δ/κ with $\phi=\pi$. The other parameters are $\Delta_1=0$, $U=20\kappa$, and $\varepsilon=0.1\kappa$.

where $N_{\rm out} \approx \kappa [|C_{10}|^2 + |C_{01}|^2 - 2\text{Re}(C_{01}C_{10}^*)]$. The probability amplitudes for two-photon states in the steady state are approximately given by

$$C_{20} \approx \frac{-i}{U - i\kappa} \frac{\varepsilon^2}{\sqrt{2}\kappa}, \quad C_{02} \approx -\frac{\sqrt{2}}{2U - i\kappa} \frac{\varepsilon^2}{4U},$$

$$C_{11} \approx \frac{-i}{U - i\kappa} \left(\frac{\varepsilon^2}{2\kappa} - \frac{i\varepsilon^2}{4U}\right) \tag{4}$$

with $|C_{20}| \approx \sqrt{2|C_{11}|} \gg |C_{02}|$. The first and last terms inside the curly brace of Eq. (3) are canceled out by the destructive interference between C_{20} and C_{11} . Then the second-order correlation function in the output field is approximately given by

$$g_{\text{out}}^{(2)}(0) \approx \left[\kappa/(2U)\right]^4.$$
 (5)

The second-order correlation of the photons in the output field depends on the strength of the nonlinear interaction with a biquadratic scaling law, which is different from the second-order correlation of photons in the cavity a_1 , i.e., $g_1^{(2)}(0) \approx (\kappa/U)^2$, with a quadratic scaling law.

Both $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$, obtained by solving the master equation numerically, are plotted as functions of $\log_{10}(U/\kappa)$ in Fig. 3. In the strong nonlinear regime $U/\kappa \gg 1$, the slope of $\log_{10}[g_{\text{out}}^{(2)}(0)]$ versus $\log_{10}(U/\kappa)$ is -4, which is much larger than the slope of -2 for $\log_{10}[g_1^{(2)}(0)]$ versus $\log_{10}(U/\kappa)$. The numerical results agree well with the analytical expressions in the strong nonlinear regime (black-dashed lines in Fig. 3). Thus, the scheme we proposed cannot only greatly enhance photon blockade by several orders, but also change the scaling exponent of the second-order correlation on the nonlinear interaction strength from -2 to -4.

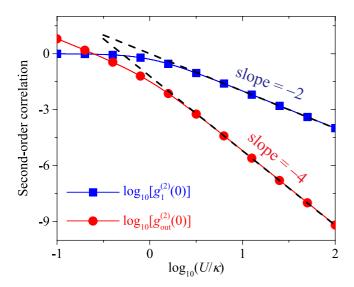


FIG. 3. The second-order correlations $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ vs the nonlinear interaction strength $\log_{10}(U/\kappa)$ with $\Delta_1=0, \phi=\pi$, and $\delta=2U$.

V. JC MODEL

The scheme for photon blockade enhancement in output field is general. It can be extended to other optical platforms with anharmonic energy levels, such as the JC model [63]. The strong coupling between a single cavity and a TLS has been realized decades ago [64–67], and photon blockade was demonstrated in a large number of experiments based on JC model [18–24]. Here, we demonstrate a scaling enhancement of photon blockade in the mixing field output from two (uncoupled) cavities with a TLS strongly coupled to one of them.

The scheme can be extended to the JC model just by replacing the $\chi^{(3)}$ nonlinear medium [Fig. 1(a)] by a TLS [Fig. 4(a)], and the system is described by

$$H_{JC} = \Delta_1 a_1^{\dagger} a_1 + \Delta_a \sigma_+ \sigma_- + g(a_1^{\dagger} \sigma_- + \sigma_+ a_1) + \Delta_2 a_2^{\dagger} a_2 + i \varepsilon (a_1^{\dagger} + a_2^{\dagger} - \text{H.c.}),$$
 (6)

where σ_+ and σ_- are the raising and lowering operators of the TLS with transition frequency ω_a , $\Delta_a = \omega_a - \omega_p$ is the laser detuning from the TLS, and g is the TLS-cavity coupling strength. We assume that the TLS is resonant with the cavity ($\Delta_a = \Delta_1 = \Delta$), and the decay rate of the TLS is $\kappa_a = 2\kappa$.

In order to confirm the applicability of our scheme in JC model, we perform a fully numerical simulation of the second-order correlation in the output field based on the master equation. $\log_{10}[g_{\text{out}}^{(2)}(0)]$ is plotted as a function of ϕ/π and Δ_2/κ in Fig. 4(b), for $g=20\kappa$ and $\Delta=-g$. The minimum of $\log_{10}[g_{\text{out}}^{(2)}(0)]$ appears around $\phi=\pi$ and $\Delta_2\approx-13.3\kappa\approx-2g/3$. From the cuts of the color scale plot shown in Figs. 4(c) and 4(d), $g_{\text{out}}^{(2)}(0)$ is about 4.3 orders smaller than $g_1^{(2)}(0)$ at $\phi=0.99\pi$ with $\Delta_2=-2g/3$ [Fig. 4(c)], and about three orders smaller than $g_1^{(2)}(0)$ at $\Delta_2\approx-13.38\kappa$ with $\phi=\pi$ [Fig. 4(d)].

In order to understand the enhancement of photon blockade in the output field, we derive the expression of $g_{\rm out}^{(2)}(0)$ by using the effective

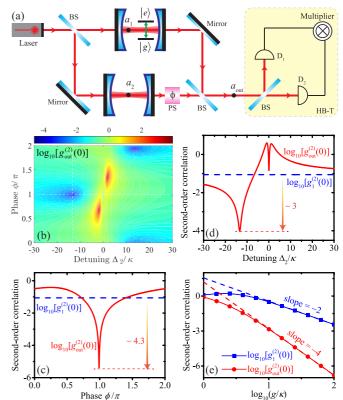


FIG. 4. Photon blockade enhancement in the mixing output field of two cavities with a TLS in one of them. (a) Sketch of the proposed scheme with a TLS (excited state $|e\rangle$ and ground state $|g\rangle$) in cavity a_1 . (b) The second-order correlation $\log_{10}[g_{\text{out}}^{(2)}(0)]$ for different phase ϕ/π and detuning Δ_2/κ . The second-order correlations $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ vs (c) phase ϕ/π with $\Delta_2=-2g/3$ and (d) detuning Δ_2/κ with $\phi=\pi$. (e) The second-order correlations $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ versus the interaction strength $\log_{10}(g/\kappa)$ with $\phi=\pi$, $\Delta_1=-g$, and $\Delta_2=-2g/3$. The other parameters are $g=20\kappa$ and $\kappa_a=2\kappa$.

Hamiltonian $H_{\text{JC,eff}} = H_{\text{JC}} - i\kappa (a_1^\dagger a_1 + a_2^\dagger a_2 + \sigma_+ \sigma_-)$ and wave function $|\varphi\rangle = \sum_{n_2=0}^{2-n_1} \sum_{n_1=0}^2 C_{gn_1n_2} |g, n_1, n_2\rangle + \sum_{n_2=0}^{1-n_1} \sum_{n_1=0}^1 C_{en_1n_2} |e, n_1, n_2\rangle$ (see Appendix C). Here, $|g, n_1, n_2\rangle$ ($|e, n_1, n_2\rangle$) denotes the Fock state of n_1 photons in cavity a_1, n_2 photons in cavity a_2 , and the TLS in the ground (excited) state, with the probability amplitude $C_{gn_1n_2}$ ($C_{en_1n_2}$). The optimal condition $\Delta_2 = -2g/3$ for photon blockade in the output field is obtained by setting $C_{g02} \approx \sqrt{2}C_{g11}$, and they are canceled out by the destructive interference in the output field at phase difference $\phi = \pi$. Thus the second-order correlation function in the output field becomes (see more details in Appendix C)

$$g_{\text{out}}^{(2)}(0) \approx 16(\kappa/g)^4.$$
 (7)

We also have the second-order correlation in cavity a_1 as $g_1^{(2)}(0) \approx 36(\kappa/g)^2$ for single-mode JC model, and they [dash lines in Fig. 4(e)] agree well with numerical results in the strong coupling regime. Similar to the case for the cavity containing $\chi^{(3)}$ nonlinear medium, the scheme with JC model can also enhance photon blockade in the output field by several

orders, and change the scaling exponent of the second-order correlation on the strength of the TLS-cavity interaction from -2 to -4.

VI. CONCLUSIONS

In conclusion, we have proposed a scheme to achieve scaling enhancement of photon blockade in the mixing field output from a nonlinear cavity (in the strong nonlinear regime) and an auxiliary (linear) cavity. We identify that the probability for two photons in the output field can be significantly inhibited by the quantum interference between two of the paths for two photons passing through the whole system, leading to a biquadratic scaling relation between the second-order correlation of the photons in the output field and intracavity nonlinear interaction strength, in contrast to a quadratic scaling relation for the photons in a nonlinear cavity. The scheme for photon blockade enhancement is general, for it not only achievable in the cavity containing $\chi^{(3)}$ nonlinearity [68] and TLS [69,70], but also applicable in cavities with other nonlinear interactions, such as $\chi^{(2)}$ nonlinearity [71,72] and optomechanical interactions [73-79] (see Appendices D and E). Furthermore, our scheme can be directly extended to enhance phonon blockade [80–83], magnon blockade [84–88], and polariton blockade [89-93]. It is worth mentioning that, as the second-order correlations become very small, there are some other noises in the experiments, such as the noises in the lasers and photodetectors, that may weaken the photon blockade effect, and such effect should be considered case by case.

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APPENDIX A: SECOND-ORDER CORRELATION OF A_{out}

The second-order correlation function of the output field A_{out} is defined by

$$g_{\text{OUT}}^{(2)}(\tau) \equiv \frac{\langle A_{\text{out}}^{\dagger} A_{\text{out}}^{\dagger}(\tau) A_{\text{out}}(\tau) A_{\text{out}} \rangle}{\langle A_{\text{out}}^{\dagger} A_{\text{out}} \rangle^{2}}$$

$$= \sum_{j,k,l,m=1}^{2} e^{in\phi'} \sqrt{\kappa_{j} \kappa_{k} \kappa_{l} \kappa_{m}} \frac{\langle a_{j}^{\dagger} a_{k}^{\dagger}(\tau) a_{l}(\tau) a_{m} \rangle}{N_{\text{OUT}}^{2}},$$
(A1)

where n=l+m-j-k, $\phi'=\phi+\pi$, and $N_{\rm OUT}=\kappa_1\langle a_1^\dagger a_1\rangle+\kappa_2\langle a_2^\dagger a_2\rangle+2\sqrt{\kappa_1\kappa_2}{\rm Re}(e^{i(\phi+\pi)}\langle a_1^\dagger a_2\rangle)$. The second-order correlation $g_{\rm OUT}^{(2)}(0)$ can be obtained from $g_{\rm out}^{(2)}(0)$ just with ϕ replaced by ϕ' .

The second-order correlation $\log_{10}[g_{\mathrm{OUT}}^{(2)}(0)]$ for different phase ϕ/π and detuning δ/κ are shown in Fig. 5(a). Different from the second-order correlation $\log_{10}[g_{\mathrm{out}}^{(2)}(0)]$ [Fig. 2(a)], the minimal values of $\log_{10}[g_{\mathrm{OUT}}^{(2)}(0)]$ are obtained for $\{\delta\approx$

 $-2U,\ \phi\approx\pi\}$ or $\{\delta\approx 2U,\ \phi\approx 0\}$. In order to show the relation between $\log_{10}[g_{\rm out}^{(2)}(0)]$ and $\log_{10}[g_{\rm OUT}^{(2)}(0)]$ clearly, we plot $\log_{10}[g_{\rm out}^{(2)}(0)],\ \log_{10}[g_{\rm OUT}^{(2)}(0)],\$ and $\log_{10}[g_{1}^{(2)}(0)]$ versus phase ϕ/π in Fig. 5(b). In comparing with $\log_{10}[g_{1}^{(2)}(0)],\$ log $_{10}[g_{\rm out}^{(2)}(0)],\$ and $\log_{10}[g_{\rm OUT}^{(2)}(0)]$ are enhanced or suppressed periodically with the phase ϕ/π , and there is a phase difference of π between them, which are consistent with Eqs. (2) and (A1). To avoid unnecessary duplication, we focus on the output field $a_{\rm out}$ in the main text.

APPENDIX B: $\chi^{(3)}$ NONLINEARITY

In this Appendix, we will derive the analytical expressions of the second-order correlation function for photons in the mixing output field of two (uncoupled) cavities with $\chi^{(3)}$ nonlinearity in one of them. In order to obtain the analytical expression of the second-order correlation function of the photons in the output field, we use the wave function

$$|\psi\rangle = C_{00}|0,0\rangle + C_{10}|1,0\rangle + C_{01}|0,1\rangle + C_{20}|2,0\rangle + C_{11}|1,1\rangle + C_{02}|0,2\rangle + \cdots,$$
 (B1)

where $|n_1, n_2\rangle$ represents the Fock state with n_1 photons in cavity a_1 and n_2 photons in cavity a_2 , with the probability amplitude $C_{n_1n_2}$. According to the quantum-trajectory method [62], we introduce an effective Hamiltonian

$$H_{\text{eff}} = H - i\kappa_{1}a_{1}^{\dagger}a_{1} - i\kappa_{2}a_{2}^{\dagger}a_{2}$$

$$= (\Delta_{1} - i\kappa_{1})a_{1}^{\dagger}a_{1} + Ua_{1}^{\dagger}a_{1}^{\dagger}a_{1}a_{1} + i\varepsilon_{1}(a_{1}^{\dagger} - a_{1})$$

$$+ (\Delta_{2} - i\kappa_{2})a_{2}^{\dagger}a_{2} + i\varepsilon_{2}(a_{2}^{\dagger} - a_{2}), \tag{B2}$$

to include the decay effect of cavities. By substituting the wave function and effective Hamiltonian into the Schrödinger equation $id|\psi\rangle/dt=H_{\rm eff}|\psi\rangle$, we get the dynamic equations for the probability amplitude $C_{n_1n_2}$ as

$$i\frac{d}{dt}C_{10} = (\Delta_{1} - i\kappa_{1})C_{10} + i\varepsilon_{1}C_{00} - i\varepsilon_{2}C_{11} - i\sqrt{2}\varepsilon_{1}C_{20},$$

$$i\frac{d}{dt}C_{01} = (\Delta_{2} - i\kappa_{2})C_{01} + i\varepsilon_{2}C_{00} - i\varepsilon_{1}C_{11} - i\sqrt{2}\varepsilon_{2}C_{02},$$

$$i\frac{d}{dt}C_{20} = (2\Delta_{1} + 2U - i2\kappa_{1})C_{20} + i\sqrt{2}\varepsilon_{1}C_{10},$$

$$i\frac{d}{dt}C_{02} = (2\Delta_{2} - i2\kappa_{2})C_{02} + i\sqrt{2}\varepsilon_{2}C_{01},$$

$$i\frac{d}{dt}C_{11} = (\Delta_{1} + \Delta_{2} - i\kappa_{1} - i\kappa_{2})C_{11} + i\varepsilon_{2}C_{10} + i\varepsilon_{1}C_{01},$$

$$\cdots$$
(B3)

In the steady state, e.g., $dC_{n_1n_2}/dt = 0$, we have

$$-i\varepsilon_{1}C_{00} = (\Delta_{1} - i\kappa_{1})C_{10} - i\varepsilon_{2}C_{11} - i\sqrt{2}\varepsilon_{1}C_{20},$$

$$-i\varepsilon_{2}C_{00} = (\Delta_{2} - i\kappa_{2})C_{01} - i\varepsilon_{1}C_{11} - i\sqrt{2}\varepsilon_{2}C_{02},$$

$$0 = (2\Delta_{1} + 2U - i2\kappa_{1})C_{20} + i\sqrt{2}\varepsilon_{1}C_{10},$$

$$0 = (2\Delta_{2} - i2\kappa_{2})C_{02} + i\sqrt{2}\varepsilon_{2}C_{01},$$

$$0 = (\Delta_{1} + \Delta_{2} - i\kappa_{1} - i\kappa_{2})C_{11} + i\varepsilon_{2}C_{10} + i\varepsilon_{1}C_{01}.$$

$$\cdots$$
(B4)

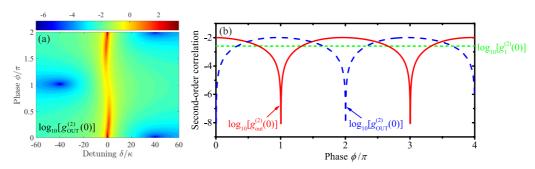


FIG. 5. (a) The second-order correlation $\log_{10}[g_{OUT}^{(2)}(0)]$ for different phase ϕ/π and detuning δ/κ . (b) The second-order correlations $\log_{10}[g_{Out}^{(2)}(0)]$, $\log_{10}[g_{OUT}^{(2)}(0)]$, and $\log_{10}[g_{10}^{(2)}(0)]$ vs phase ϕ/π with $\delta=2U$. The other parameters are $\Delta_1=0$, $U=20\kappa$, and $\varepsilon=0.1\kappa$.

Under weak driving conditions $\{\varepsilon_1, \varepsilon_2\} \ll \{\kappa_1, \kappa_2\}$, we have $|C_{00}| \approx 1 \gg \{|C_{10}|, |C_{01}|\} \gg \{|C_{20}|, |C_{02}|, |C_{11}|\}$, then the probability amplitudes of one-photon states $(C_{10}$ and $C_{01})$ are obtained as

$$C_{10} = \frac{-i\varepsilon_1}{\Delta_1 - i\kappa_1},\tag{B5}$$

$$C_{01} = \frac{-i\varepsilon_2}{\Delta_2 - i\kappa_2},\tag{B6}$$

and the probability amplitudes of two-photon states (C_{20} , C_{11} , and C_{02}) are obtained as

$$C_{20} = \frac{-i\sqrt{2}\varepsilon_1}{2\Delta_1 + 2U - i2\kappa_1}C_{10},$$
 (B7)

$$C_{11} = \frac{-i\varepsilon_2 C_{10} - i\varepsilon_1 C_{01}}{\Delta_1 + \Delta_2 - i(\kappa_1 + \kappa_2)},$$
 (B8)

$$C_{02} = \frac{-i\sqrt{2}\varepsilon_2}{2\Delta_2 - i2\kappa_2} C_{01}.$$
 (B9)

Under weak driving conditions, the first-order correlation functions can be given by the steady state probability amplitudes $C_{n_1n_2}$ as

$$\langle a_1^{\dagger} a_1 \rangle \approx |C_{10}|^2, \quad \langle a_2^{\dagger} a_2 \rangle \approx |C_{01}|^2,$$

 $\langle a_1^{\dagger} a_2 \rangle \approx C_{01} C_{10}^*,$ (B10)

and the second-order correlation functions can be given by

$$\begin{split} \langle a_{1}^{\dagger} a_{1}^{\dagger} a_{1} a_{1} \rangle &\approx 2 |C_{20}|^{2}, \quad \langle a_{1}^{\dagger} a_{1}^{\dagger} a_{1} a_{2} \rangle \approx \sqrt{2} C_{11} C_{20}^{*}, \\ \langle a_{2}^{\dagger} a_{2} a_{1}^{\dagger} a_{1} \rangle &\approx |C_{11}|^{2}, \quad \langle a_{1}^{\dagger} a_{1}^{\dagger} a_{2} a_{2} \rangle \approx 2 C_{02} C_{20}^{*}, \\ \langle a_{2}^{\dagger} a_{1}^{\dagger} a_{2} a_{2} \rangle &\approx \sqrt{2} C_{02} C_{11}^{*}, \quad \langle a_{2}^{\dagger} a_{2}^{\dagger} a_{2} a_{2} \rangle \approx 2 |C_{02}|^{2}, \quad (B11)^{2}. \end{split}$$

Thus, the second-order correlation function in the output field can be given approximately as

$$g_{\text{out}}^{(2)}(0) \approx \frac{2}{N_{R,\text{out}}^2} \{ |\kappa_1 C_{20} + e^{i\phi} \sqrt{2\kappa_1 \kappa_2} C_{11}|^2 + |\kappa_2 C_{02}|^2 + 2\text{Re}[(\kappa_1 C_{20} + e^{i\phi} \sqrt{2\kappa_1 \kappa_2} C_{11})e^{-i2\phi} \kappa_2 C_{02}^*] \},$$
(B12)

where $N_{\rm out} \approx |\sqrt{\kappa_1}C_{10} + e^{i\phi}\sqrt{\kappa_2}C_{01}|^2$.

First of all, let us consider the conditions that $\varepsilon_1 = \varepsilon_2 = \varepsilon$ and $\kappa_1 = \kappa_2 = \kappa$. According to the numerical results shown in Fig. 2 in the main text, photon blockade in the output field is enhanced greatly under the conditions $\{\delta = 2U, \ \phi = \pi\}$ or

 $\{\delta=-2U,\ \phi=0\}$, with the other parameters $\{\Delta_1=0\ \text{and}\ U\gg\kappa\}$. In order to understand these phenomena analytically, we rewrite the second-order correlation as

$$g_{\text{out}}^{(2)}(0) \approx \frac{2\kappa^2}{N_{\text{out}}^2} \{ |C_{20} + e^{i\phi}\sqrt{2}C_{11}|^2 + |C_{02}|^2 + 2\text{Re}[(C_{20} + e^{i\phi}\sqrt{2}C_{11})(e^{i2\phi}C_{02})^*] \}$$
(B13)

where

$$C_{10} = \frac{\varepsilon}{\kappa}, \quad C_{01} \approx -i\frac{\varepsilon}{2U},$$
 (B14)

and

$$C_{20} pprox \frac{-i\sqrt{2}\varepsilon}{2U - i2\kappa} \frac{\varepsilon}{\kappa},$$
 (B15)

$$C_{11} pprox \left(\frac{-\varepsilon^2}{\delta - i2\kappa}\right) \left(\frac{i}{\kappa} + \frac{1}{2U}\right),$$
 (B16)

$$C_{02} \approx -\frac{\sqrt{2}\varepsilon}{4U - i2\kappa} \frac{\varepsilon}{2U}.$$
 (B17)

In the strong nonlinear regime, i.e., $|\delta| = 2U \gg \kappa$, we have

$$|C_{10}| \gg |C_{01}|, |C_{20}| \approx |\sqrt{2}C_{11}| \gg |C_{02}|.$$
 (B18)

Under the conditions $\{\delta=2U,\ \phi=\pi\}$ or $\{\delta=-2U,\ \phi=0\}$, C_{20} and $\sqrt{2}C_{11}$ cancel each other out by destructive interference, with

$$|C_{20} + e^{i\phi}\sqrt{2}C_{11}| \ll |C_{20}| \approx |\sqrt{2}C_{11}|.$$
 (B19)

In this case, the second-order correlation function for the photons in the output field is given by

$$g_{\text{out}}^{(2)}(0) \approx \frac{1}{16} \left(\frac{\kappa}{U}\right)^4,$$
 (B20)

which is much smaller than the second-order correlation function for the photons in the cavity a_1

$$g_1^{(2)}(0) = \frac{\langle a_1^{\dagger} a_1^{\dagger} a_1 a_1 \rangle}{\langle a_1^{\dagger} a_1 \rangle^2} \approx \frac{2|C_{20}|^2}{|C_{10}|^4} \approx \left(\frac{\kappa}{U}\right)^2$$
(B21)

in the strong nonlinear regime.

In addition, we discuss how to achieve scaling enhancement of photon blockade with $\kappa_1 \neq \kappa_2$. According to the definition of ε_i , i.e., $\varepsilon_i = \sqrt{\kappa_i P_{\rm in}/\hbar \omega_p}$, where $P_{\rm in}$ is the driving power of the two cavities, we have $\varepsilon_1/\varepsilon_2 = \sqrt{\kappa_1/\kappa_2}$. According to Eq. (B12), in order to achieve scaling enhancement of photon blockade, the coefficients (C_{20} , C_{11} , and C_{02}) should

satisfy the conditions $\{|\kappa_2 C_{02}|, |\kappa_1 C_{20} + e^{i\phi}\sqrt{2\kappa_1\kappa_2}C_{11}|\} \ll |\kappa_1 C_{20}| \approx |\sqrt{2\kappa_1\kappa_2}C_{11}|$. Based on the expressions of the coefficients $(C_{20}, C_{11}, \text{ and } C_{02})$ [Eqs. (B7)-(B9)], the conditions are satisfied with

$$\Delta_1 = 0, \quad \delta \approx 2U\kappa_2/\kappa_1, \quad \phi \approx \pi,$$
 (B22)

or

$$\Delta_1 = 0, \quad \delta \approx -2U\kappa_2/\kappa_1, \quad \phi \approx 0,$$
 (B23)

in the strong nonlinear regime $(U \gg \{\kappa_1, \kappa_2\})$ and the ratio $\kappa_2/\kappa_1 \gg (\kappa_1/2U)^2$. In this case, the second-order correlation function for the photons in the output field is given by

$$g_{\text{out}}^{(2)}(0) = \frac{1}{16} \left(\frac{\kappa_1}{U}\right)^4,$$
 (B24)

which also depends on the strength of the nonlinear interaction U with a biquadratic scaling law.

As an example, we show the second-order correlation $\log_{10}[g_{\text{out}}^{(2)}(0)]$ in Fig. 6 for $\kappa_2 = \kappa_1/10$. We can see that the numerical results shown in Fig. 6 are very similar to the Figs. 2 and 3, except that the optimal detunings are $\delta \approx \pm 2U\kappa_2/\kappa_1$. In brief, we can achieve scaling enhancement of photon blockade with $\kappa_1 = \kappa_2$ or $\kappa_1 \neq \kappa_2$. In the main text, we set $\kappa_1 = \kappa_2 = \kappa$, without loss of generality.

APPENDIX C: TLS-CAVITY INTERACTION

In this Appendix, we will derive the second-order correlation function of the output field for the system consisting of two (uncoupled) cavities with a TLS in one of them. The system can be described by a JC model, including decay effects, as

$$H_{\text{JC,eff}} = H_{\text{JC}} - i\kappa_1 a_1^{\dagger} a_1 - i\kappa_2 a_2^{\dagger} a_2 - i\frac{\kappa_a}{2}\sigma_+ \sigma_-$$

$$= (\Delta_1 - i\kappa_1)a_1^{\dagger} a_1 + (\Delta_a - i\frac{\kappa_a}{2})\sigma_+ \sigma_-$$

$$+ g(a_1^{\dagger} \sigma_- + \sigma_+ a_1) + (\Delta_2 - i\kappa_2)a_2^{\dagger} a_2$$

$$+i\varepsilon_1(a_1^{\dagger}-a_1)+i\varepsilon_2(a_2^{\dagger}-a_2),$$
 (C1)

and the wave function

$$\begin{aligned} |\varphi\rangle &= C_{g00}|g,0,0\rangle + C_{g10}|g,1,0\rangle + C_{g01}|g,0,1\rangle \\ &+ C_{g20}|g,2,0\rangle + C_{g11}|g,1,1\rangle + C_{g02}|g,0,2\rangle \\ &+ C_{e00}|e,0,0\rangle + C_{e10}|e,1,0\rangle + C_{e01}|e,0,1\rangle, \end{aligned}$$
(C2)

with Fock-state basis truncated to the two-photon manifold, under the weak driving conditions ($\varepsilon \ll \{\kappa_1, \kappa_2, \kappa_a\}$). Here, $|g, n_1, n_2\rangle$ ($|e, n_1, n_2\rangle$) denotes the Fock state of n_1 photons in cavity a_1 , n_2 photons in cavity a_2 , and the TLS in the ground (excited) state, with the probability amplitude $C_{gn_1n_2}$ ($C_{en_1n_2}$). By substituting the wave function $|\varphi\rangle$ and effective Hamiltonian $H_{JC,eff}$ into the Schrödinger equation, $d|\varphi\rangle/dt = -iH_{JC,eff}|\varphi\rangle$, we get the dynamic equations for the probability amplitudes $C_{gn_1n_2}$ ($C_{en_1n_2}$), and the probability amplitudes $C_{gn_1n_2}$ ($C_{en_1n_2}$) can be obtained analytically in the steady state $dC_{gn_1n_2}/dt = dC_{en_1n_2}/dt = 0$. Under weak driving conditions, we have $|C_{g00}| \approx 1 \gg \{|C_{g10}|, |C_{g01}|, |C_{e00}|\} \gg \{|C_{g20}|, |C_{g11}|, |C_{g02}|, |C_{e10}|, |C_{e01}|\}$, then the probability amplitudes of one-particle excitation states (C_{g10} , C_{g01} , and C_{e00}) are obtained as

$$C_{g10} = \frac{-i\varepsilon \left(\Delta_a - i\frac{\kappa_a}{2}\right)}{(\Delta_1 - i\kappa_1)\left(\Delta_a - i\frac{\kappa_a}{2}\right) - g^2},\tag{C3}$$

$$C_{e00} = \frac{i\varepsilon g}{(\Delta_1 - i\kappa_1)(\Delta_a - i\frac{\kappa_a}{2}) - g^2},$$
 (C4)

$$C_{g01} = \frac{-i\varepsilon}{(\Delta_2 - i\kappa_2)},\tag{C5}$$

and the probability amplitudes of two-photon states (C_{g20} , C_{g11} , and C_{g02}) are obtained as

$$C_{g20} = -i\varepsilon\sqrt{2} \frac{(\Delta_1 + \Delta_a - i\kappa_2 - i\kappa_a/2)C_{g10} - gC_{e00}}{(2\Delta_1 - i2\kappa_1)(\Delta_1 + \Delta_a - i\kappa_2 - i\kappa_a/2) - 2g^2},$$
(C6)

$$C_{g11} = \frac{-i\varepsilon[(\Delta_a + \Delta_2 - i\kappa_2 - i\kappa_a/2)C_{g10} - gC_{e00}] - i\varepsilon C_{g01}(\Delta_a + \Delta_2 - i\kappa_2 - i\kappa_a/2)}{(\Delta_1 + \Delta_2 - i\kappa_1 - i\kappa_2)(\Delta_a + \Delta_2 - i\kappa_2 - i\kappa_a/2) - g^2},$$
(C7)

$$C_{g02} = -\frac{i\sqrt{2}\varepsilon}{(2\Delta_2 - i2\kappa_2)}C_{g01}.$$
 (C8)

The second-order correlation function for photons in the output field can be given approximately by the probability amplitudes $C_{gn_1n_2}$ ($C_{en_1n_2}$) as

$$g_{\text{out}}^{(2)}(0) \approx \frac{1}{N_{\text{out}}^2} \left[2\kappa_1^2 |C_{g20}|^2 + 2\kappa_2^2 |C_{g02}|^2 + 4\kappa_1 \kappa_2 |C_{g11}|^2 + 4\kappa_1 \kappa_2 \text{Re}(e^{i2\phi} C_{g02} C_{g20}^*) + 4\sqrt{2}\kappa_1 \sqrt{\kappa_1 \kappa_2} \text{Re}(e^{i\phi} C_{g11} C_{g20}^*) + 4\sqrt{2}\kappa_2 \sqrt{\kappa_1 \kappa_2} \text{Re}(e^{i\phi} C_{g02} C_{g11}^*) \right]$$
(C9)

with
$$N_{\text{out}} \approx \kappa_1 |C_{g10}|^2 + \kappa_2 |C_{g01}|^2 + 2\sqrt{\kappa_1 \kappa_2} \text{Re}(e^{i\phi} C_{g01} C_{g10}^*).$$

In order to obtain the parameter conditions for photon blockade enhancement in the output field, based on the quantum interference between the coefficients C_{g20} and C_{g11} , the second-order correlation function can be rewritten as

$$\begin{split} g_{\text{out}}^{(2)}(0) &\approx \frac{2\kappa^2}{N_{\text{out}}^2} \{ |C_{g20} + e^{i\phi} \sqrt{2} C_{g11}|^2 + |C_{g02}|^2 \\ &+ 2 \text{Re}[e^{i2\phi} (C_{g20} + e^{i\phi} \sqrt{2} C_{g11})^* C_{g02}] \} \end{split}$$
 (C10)

under the condition $\kappa_1 = \kappa_2 = \kappa_a/2 = \kappa$. In order to cancel the terms related to C_{g20} and C_{g11} , we need

$$\frac{|C_{g20}|}{|C_{g11}|} = \sqrt{2},\tag{C11}$$

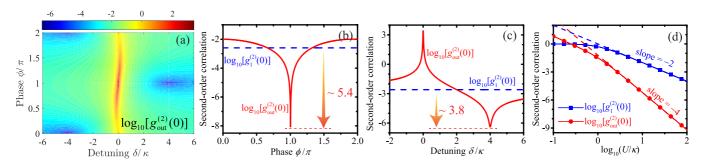


FIG. 6. (a) The second-order correlation $\log_{10}[g^{(2)}_{out}(0)]$ for different phase ϕ/π and detuning δ/κ with $\kappa_2 = \kappa_1/10$ and $U = 20\kappa$. The second-order correlations $\log_{10}[g^{(2)}_{out}(0)]$ and $\log_{10}[g^{(2)}_{10}(0)]$ vs (b) phase ϕ/π with $\delta = 2U\kappa_2/\kappa_1$ and $U = 20\kappa$, (c) detuning δ/κ with $\phi = \pi$ and $U = 20\kappa$, and (d) the $\chi^{(3)}$ nonlinear interaction strength $\log_{10}(U/\kappa)$ with $\delta = 2U\kappa_2/\kappa_1$ and $\phi = \pi$. The other parameters are $\Delta_1 = 0$, $\kappa_1 = \kappa$, $\kappa_2 = \kappa/10$, $\varepsilon_1 = 0.1\kappa$, and $\varepsilon_2 = \varepsilon_1\sqrt{\kappa_2/\kappa_1}$.

which is satisfied with

$$\Delta_2 = \pm \frac{2}{3}g,\tag{C12}$$

under the optimal conditions ($\Delta_1 = \Delta_a = \pm g$ and $g \gg \kappa$) for photon blockade in cavity a_1 . In the case of $\{\Delta_2 = -2g/3, \ \phi \approx \pi\}$ (or $\{\Delta_2 = 2g/3, \ \phi \approx 0\}$), the second-order correlation function for photons in the output field is simplified as

$$g_{\text{out}}^{(2)}(0) \approx 16 \left(\frac{\kappa}{g}\right)^4,$$
 (C13)

which is much smaller than the second-order correlation function for photons in cavity a_1 as

$$g_1^{(2)}(0) = \frac{\langle a_1^{\dagger} a_1^{\dagger} a_1 a_1 \rangle}{\langle a_1^{\dagger} a_1 \rangle^2} \approx \frac{2 |C_{g20}|^2}{|C_{g10}|^4} \approx 36 \left(\frac{\kappa}{g}\right)^2,$$
 (C14)

in the strong coupling regime $g \gg \kappa$.

APPENDIX D: SECOND-ORDER NONLINEAR INTERACTION

The second-order nonlinear interaction in non-centrosymmetric materials is another typical nonlinear interactions that has attracted great attentions for it is usually orders of magnitude higher than the third-order interaction [71,72]. In this Appendix, we will consider the $\chi^{(2)}$ interaction in one of the cavities, then the system Hamiltonian becomes

$$H_{2\text{nd}} = \Delta_1 a_1^{\dagger} a_1 + \Delta_b b^{\dagger} b + g \left(a_1^{\dagger 2} b + b^{\dagger} a_1^2 \right) + \Delta_2 a_2^{\dagger} a_2 + i \varepsilon_1 \left(a_1^{\dagger} - a_1 \right) + i \varepsilon_2 \left(a_2^{\dagger} - a_2 \right).$$
 (D1)

Here, $\Delta_b = \omega_b - 2\omega_p$ is the detuning of the second-harmonic mode (b with frequency ω_b), and g is the corresponding second-order nonlinear interaction strength.

In the presence of optical decay, the system can be written as an effective Hamiltonian

$$H_{\text{2nd,eff}} = H_{\text{2nd}} - i\kappa_{1}a_{1}^{\dagger}a_{1} - i\kappa_{2}a_{2}^{\dagger}a_{2} - i\kappa_{b}b^{\dagger}b,$$

$$= (\Delta_{1} - i\kappa_{1})a_{1}^{\dagger}a_{1} + (\Delta_{b} - i\kappa_{b})b^{\dagger}b$$

$$+ g(a_{1}^{\dagger^{2}}b + b^{\dagger}a_{1}^{2}) + (\Delta_{2} - i\kappa_{2})a_{2}^{\dagger}a_{2}$$

$$+ i\varepsilon_{1}(a_{1}^{\dagger} - a_{1}) + i\varepsilon_{2}(a_{2}^{\dagger} - a_{2}), \tag{D2}$$

where κ_b is the one-sided decay rate of the second-harmonic mode b. For a very weak probe field, the state of the system can be truncated to the first several Fock states as

$$|\psi'\rangle = C_{000}|0,0,0\rangle + C_{010}|0,1,0\rangle + C_{001}|0,0,1\rangle + C_{100}|1,0,0\rangle + C_{020}|0,2,0\rangle + C_{011}|0,1,1\rangle + C_{002}|0,0,2\rangle.$$
(D3)

Here, $|n_b, n_1, n_2\rangle$ represents the Fock state of n_b photons in cavity b, n_1 photons in cavity a_1 , and n_2 photons in cavity a_2 , with the probability amplitude $C_{n_bn_1n_2}$. Substituting the wave function $|\psi'\rangle$ and effective Hamiltonian $H_{2\text{nd,eff}}$ into the Schrödinger's equation $i\partial |\psi'\rangle/\partial t = H_{2\text{nd,eff}}|\psi'\rangle$, we get the dynamic equations for the probability amplitudes $C_{n_bn_1n_2}$. For simplicity, we assume that $\kappa_b = 2\kappa_1 = 2\kappa_2 = 2\kappa$, $\Delta_b = \Delta_1 = 0$, $\varepsilon_1 = \varepsilon_2 = \varepsilon$. Under weak driving conditions, i.e., $\varepsilon \ll \kappa$, we have $C_{000} \approx 1 \gg \{|C_{010}|, |C_{001}|\} \gg \{|C_{100}|, |C_{020}|, |C_{011}|, |C_{002}|\}$, and the probability amplitudes in the steady state are obtained analytically as

$$C_{010} \approx \frac{\varepsilon}{\kappa},$$
 (D4)

$$C_{001} \approx \frac{-i\varepsilon}{(\Delta_2 - i\kappa)},$$
 (D5)

for one-photon states, and

$$C_{020} \approx \frac{\sqrt{2}\varepsilon^2}{2\kappa^2 + g^2},\tag{D6}$$

$$C_{011} \approx \left[-\frac{i\varepsilon^2}{\kappa} - \frac{\varepsilon^2}{(\Delta_2 - i\kappa)} \right] \frac{1}{(\Delta_2 - i2\kappa)},$$
 (D7)

$$C_{002} \approx -\frac{\sqrt{2}\varepsilon^2}{2(\Delta_2 - i\kappa)(\Delta_2 - i\kappa)},$$
 (D8)

for two-photon states.

Based on the probability amplitudes, the second-order correlation function for photons in the output field can be rewritten as

$$g_{\text{out}}^{(2)}(0) \approx \frac{2\kappa^2}{N_{\text{out}}^2} \{ |C_{020} + e^{i\phi} \sqrt{2} C_{011}|^2 + |C_{002}|^2 + 2\text{Re}[e^{i2\phi} (C_{020} + e^{i\phi} \sqrt{2} C_{011})^* C_{002}] \}.$$
 (D9)

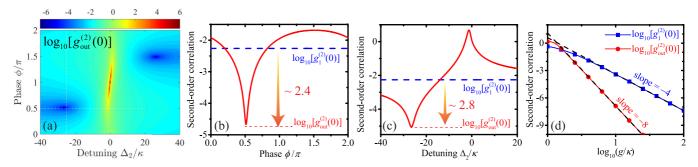


FIG. 7. (a) The second-order correlation $\log_{10}[g_{\text{out}}^{(2)}(0)]$ for different phase ϕ/π and detuning Δ_2/κ with $g=5\kappa$. The second-order correlations $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ vs (b) phase ϕ/π with $\Delta_2=-g^2/\kappa$ and $g=5\kappa$, (c) detuning Δ_2/κ with $\phi=\pi/2$ and $g=5\kappa$, and (d) the second-order nonlinear interaction strength $\log_{10}(g/\kappa)$ with $\Delta_2=-g^2/\kappa$ and $\phi=\pi/2$. The other parameters are $\Delta_1=\Delta_b=0$, $\kappa_b=2\kappa$, and $\varepsilon=0.001\kappa$.

In order to realize destructive quantum interference between C_{020} and C_{011} , i.e., $C_{010} + e^{i\phi}\sqrt{2}C_{001} \approx 0$, we have

$$\Delta_2 = \pm \frac{g^2}{\kappa}, \quad \phi = \mp \pi/2 \tag{D10}$$

for $\{|\Delta_2|, g\} \gg \kappa$. Under the above conditions, the second-order correlation function for photons in the output field is simplified as

$$g_{\text{out}}^{(2)}(0) \approx 13 \left(\frac{\kappa}{g}\right)^8,$$
 (D11)

which is much smaller than the second-order correlation function for the photons in the cavity a_1 ,

$$g_1^{(2)}(0) \approx 4 \left(\frac{\kappa}{g}\right)^4 \tag{D12}$$

in the strong coupling regime $(g > \kappa)$.

Figure 7(a) is a color plot of $\log_{10}[g_{\text{out}}^{(2)}(0)]$ as a function of the phase ϕ/π and detuning Δ_2/κ , for $\Delta_1 = \Delta_b = 0$ and $g = 5\kappa$. The minimum of $\log_{10}[g_{\text{out}}^{(2)}(0)]$ is reached for $\phi \approx \pi/2$ and $\Delta_2 \approx -g^2/\kappa$ (or $\phi \approx 3\pi/2$ and $\Delta_2 \approx g^2/\kappa$). Two cuts taken from the color plot for $\Delta_2 \approx -g^2/\kappa$ and $\phi = \pi/2$ are shown in Figs. 7(b) and 7(c), respectively. The photon blockade is enhanced significantly as $g_{\text{out}}^{(2)}(0)$ is about 2.4 orders smaller than $g_1^{(2)}(0)$ at $\phi \approx \pi/2$ [Fig. 7(b)] and about 2.8 orders smaller at $\Delta_2 \approx -g^2/\kappa$ [Fig. 7(c)]. Both $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ are plotted as functions of $\log_{10}(g/\kappa)$ in Fig. 7(d). Different from the scaling behaviors for $\chi^{(3)}$ nonlinearity and TLS-cavity interaction, in the strong second-order nonlinear regime $g/\kappa \gg 1$, the slope of $\log_{10}[g_{\text{out}}^{(2)}(0)]$ versus $\log_{10}(g/\kappa)$ is -8, which is much larger than the slope of -4 for $\log_{10}[g_1^{(2)}(0)]$ versus $\log_{10}(g/\kappa)$. The numerical results agree well with the analytical expressions in the strong nonlinear regime [black dashed lines in Fig. 7(d)]. Thus, the scheme we proposed can change the scaling exponent of the secondorder correlation on the second-order nonlinear interaction strength from -4 to -8.

APPENDIX E: OPTOMECHANICAL INTERACTION

In this Appendix, we consider a mechanical mode c with frequency ω_m in the cavity a_1 and they are coupled through optomechanical interaction [73–79], then the whole system is

described by the Hamiltonian

$$H_{\text{om}} = (\Delta_1 - i\kappa_1)a_1^{\dagger}a_1 + (\Delta_2 - i\kappa_2)a_2^{\dagger}a_2$$
$$+ (\omega_m - i\gamma)c^{\dagger}c + ga_1^{\dagger}a_1(c + c^{\dagger})$$
$$+ i\varepsilon_1(a_1^{\dagger} - a_1) + i\varepsilon_2(a_2^{\dagger} - a_2), \tag{E1}$$

where γ is the mechanical decay rate and g is the single-photon optomechanical interaction strength. In order to understand the optimal conditions for the strong photon blockade, it is convenient to transform the Hamiltonian into a displaced oscillator representation $H_{\text{om,eff}} = SH_{\text{om}}S^{\dagger}$, by the unitary transformation

$$S = \exp\left[\frac{g}{\omega_m} a_1^{\dagger} a_1 (c^{\dagger} - c)\right], \tag{E2}$$

as

$$H_{\text{om,eff}} \approx (\Delta_1' - i\kappa_1)a_1^{\dagger}a_1 + (\Delta_2 - i\kappa_2)a_2^{\dagger}a_2$$
$$+ (\omega_m - i\gamma)c^{\dagger}c - U_{\text{om}}a_1^{\dagger}a_1^{\dagger}a_1a_1$$
$$+ i\varepsilon_1(a_1^{\dagger} - a_1) + i\varepsilon_2(a_2^{\dagger} - a_2). \tag{E3}$$

Here, $\Delta_1' \equiv \Delta_1 - U_{\rm om}$, $U_{\rm om} \equiv g^2/\omega_m$, and $i\varepsilon_1\{a_1^\dagger \exp[\frac{g}{\omega_m}(c^\dagger-c)] - {\rm H.c.}\} \approx i\varepsilon_1(a_1^\dagger-{\rm H.c.})$ for $g < \omega_m$. We also assume $\kappa_1 = \kappa_2 = \kappa \gg \gamma$, $\varepsilon_1 = \varepsilon_2 = \varepsilon$, $\varepsilon \ll \kappa$, in the following.

The effective Hamiltonian (E3) for the system with optomechanical interaction is almost the same as the one (B2) for the system with $\chi^{(3)}$ nonlinearity, except the phonon mode. According to the results obtained in Appendix B, we can obtain the second-order correlation

$$g_{\text{out}}^{(2)}(0) \approx \frac{1}{16} \left(\frac{\kappa}{U_{\text{om}}}\right)^4 = \frac{1}{16} \left(\frac{\kappa \omega_m}{g^2}\right)^4$$
 (E4)

for photons in the output field, and

$$g_1^{(2)}(0) \approx \left(\frac{\kappa}{U_{\text{om}}}\right)^2 = \left(\frac{\kappa \omega_m}{g^2}\right)^2$$
 (E5)

for photons in the cavity a_1 , with the optimal conditions

$$\Delta_1 = U_{\text{om}}, \quad \Delta_2 = -2U_{\text{om}}, \quad \phi \approx \pi,$$
(E6)

or

$$\Delta_1 = U_{\text{om}}, \quad \Delta_2 = 2U_{\text{om}}, \quad \phi \approx 0.$$
 (E7)

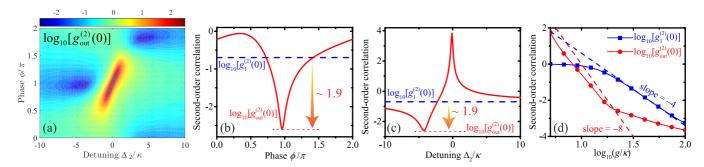


FIG. 8. (a) The second-order correlation $\log_{10}[g_{\text{out}}^{(2)}(0)]$ for different phase ϕ/π and detuning Δ_2/κ with $g=20\kappa$. The second-order correlations $\log_{10}[g_{\text{out}}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ vs (b) phase ϕ/π with $\Delta_2=-4.3\kappa$ and $g=20\kappa$, (c) detuning Δ_2/κ with $\phi=0.96\pi$ and $g=20\kappa$, and (d) the optomechanical interaction strength $\log_{10}(g/\kappa)$ with $\Delta_2=-2U_{\text{om}}$ and $\phi=0.96\pi$. The other parameters are $\Delta_1=U_{\text{om}}$, $\omega_m=200\kappa$, $\gamma=0.01\kappa$, and $\varepsilon=0.01\kappa$.

We show $\log_{10}[g_{\rm out}^{(2)}(0)]$ numerically in Fig. 8. We can see that the photon blockade is enhanced significantly as $g_{\rm out}^{(2)}(0)$ is about 1.9 orders smaller than $g_1^{(2)}(0)$ in Figs. 7(b) and 7(c). Both $\log_{10}[g_{\rm out}^{(2)}(0)]$ and $\log_{10}[g_1^{(2)}(0)]$ are plotted as functions of $\log_{10}(g/\kappa)$ in Fig. 8(d). Similar to the scaling behaviors for second-order nonlinear interaction, in the strong optomechanical interaction regime $g/\kappa \gg 1$, the slope of $\log_{10}[g_{\rm out}^{(2)}(0)]$

versus $\log_{10}(g/\kappa)$ is -8, which is much larger than the slope of -4 for $\log_{10}[g_1^{(2)}(0)]$ versus $\log_{10}(g/\kappa)$. But the numerical result is a sharp departure from the analytical expressions for the output field [red dashed lines in Fig. 8(d)]. That is because there are many phonon states in the optomechanical system, and the resonant transition between the states with different phonons may suppress photon blockade [76,78].

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