Collective multiphoton blockade in cavity quantum electrodynamics

C. J. Zhu,[1,2] Y. P. Yang,[1,*] and G. S. Agarwal[2,3,†]
[1]*MOE Key Laboratory of Advanced Micro-Structured Materials, School of Physics Science and Engineering, Tongji University, Shanghai, 200092, China*
[2]*Institute for Quantum Science and Engineering, and Department of Biological and Agricultural Engineering Texas A&M University, College Station, Texas 77843, USA*
[3]*Department of Physics, Oklahoma State University, Stillwater, Oklahoma 74078, USA*

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We present a study of collective multiphoton blockade in coherently driven atoms in a single-mode cavity. Considering two atoms strongly coupled to an optical cavity, we show that the two-photon blockade with two-photon antibunching, and the three-photon blockade with three-photon antibunching can be observed simultaneously. The three-photon blockade probes both dressed states in the two-photon and three-photon spaces. The two-photon and three-photon blockades strongly depend on the location of the two atoms in the strong-coupling regime. The asymmetry in the atom-cavity coupling constants opens pathways for multiphoton blockade which is also shown to be sensitive to the atomic decay and pumping strengths. The work presented here predicts many quantum statistical features due to the collective behavior of atoms and can be useful to generate nonclassical photon pairs.

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I. INTRODUCTION

The well-known dressed state spectrum of an atom coupled strongly to a high-quality cavity has many features which have been the hallmarks of cavity quantum electrodynamics [1–3]. The first doublet in dressed states which leads to the vacuum Rabi splitting has been the subject of innumerable investigations [3–9]. An important property of the dressed state spectrum is its anharmonicity, especially for low-lying states. Thus the next doublet is to be studied to show evidence of an anharmonic spectrum. This can be done by studying two-photon transitions in the regime of strong atom-cavity coupling [10,11]. Birnbaum *et al.* reported the photon blockade effect in the absorption of a second photon when the photon was tuned to one of the states of the lowest doublet [12]. As a result, an incident photon stream with Poissonian distribution can be converted into a sub-Poissonian, antibunching photon stream. To date, many works have shown that the two-photon blockade phenomenon is indeed feasible for many configurations [13], such as artificial atoms on a chip [14,15] and superconducting circuits [16,17]. In the strong-coupling regime, other high-lying doublets in the dressed state spectrum predict many new features of quantum nonlinear optics, which can still be explored by studying multiphoton processes [10] or by studying the dynamical behavior in strongly pumped systems [3,18,19]. Photon blockade in a cavity containing a high-Kerr medium has been discussed [20–22].

Recent experiments on two atoms trapped at well-characterized positions have unraveled many new aspects of the collective behavior in a high-quality cavity [23,24]. Interesting results include the saturation of resonance fluorescence for constructive interference, bunching photon emission for destructive interference, and suppression of superradiant behavior due to strong backreaction of the cavity [24]. If the atoms feel asymmetric coupling to the cavity field while these are driven symmetrically then one can obtain very large second-order photon-photon correlation $g^{(2)}$, which has been related to the predominance of two-photon processes [23]. Since cavities allow the possibility of a wide range of parameters, the two-atom system can, in a different parameter domain, exhibit hyper-radiance which is enhanced radiation beyond superradiance [25]. These features can be understood by using the possible transitions among the dressed states which depend on the two coupling constants of the cavity mode with the atoms. When the coupling constants are different, then new channels open up leading to new physical effects. For example, if the two coupling constants differ in phase by π, then the two-atom system permits a two-photon resonant process which is also one-photon resonant. Thus it is worthwhile to examine the nature not only of second-order photon-photon correlation but also of third-order photon-photon correlation $g^{(3)}$. The anharmonicity of the dressed state spectrum enables us to study many features of the photon blockade for a system of two atoms.

In this work we present some interesting results for the multiphoton blockade in a collective system. Specifically, we present new results when the external field is tuned to either one-photon resonance or two-photon resonance. We note that a recent experiment with a single atom in a high-quality cavity reports higher-order photon blockade [26], i.e., when the absorption of a third photon is forbidden if the two-photon absorption is resonant. However, we find many interesting features of multiphoton blockade associated with the collective behavior of two atoms that a single atom does not possess. We show that in the case of in-phase radiation, sub-Poissonian statistics of third-order photon-photon correlation can be observed as a signature of three-photon blockade. We also show that the quantum statistical properties changes significantly if the atoms feels different coupling strength to the cavity. For example, in the case of a $\pi/2$ phase shift, two-photon blockade can be significantly improved, while three-photon blockade with two-photon bunching can be realized in the case of out-of-phase radiation.

*yang_yaping@tongji.edu.cn
†girish.agarwal@tamu.edu

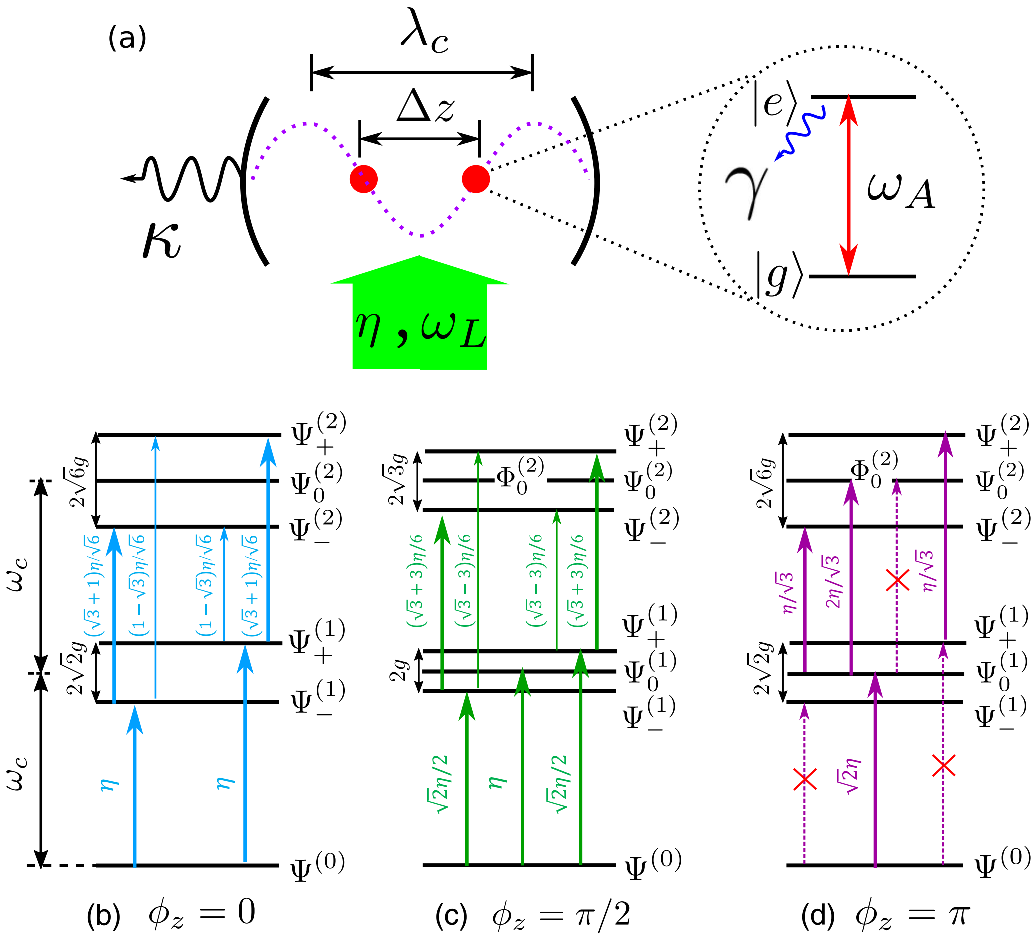

FIG. 1. (a) Schematic diagram of a single-mode cavity (wavelength λ_c) coupled with two two-level atoms driven by a pump field of frequency ω_L. The Rabi frequency of the pump field is labeled η. The spontaneous emission rate of the excited state for both atoms is 2γ and the cavity decay rate is 2κ. Panels (b)–(d) represent the dressed state structure for the same coupling strength but with $\phi_z = 0$, $\pi/2$, and π, respectively. Here, only few two-photon transitions are shown in panel (c).

II. MODEL AND DRESSED STATE PICTURE OF MULTIATOM BLOCKADE

To realize multiphoton blockade, we now consider a scheme consisting of two two-level atoms in a single-mode cavity [see Fig. 1(a)].

The dynamical behavior of the system shown in Fig. 1(a) can also be treated according to the Jaynes–Cummings model and by using the master equation

$$\frac{d}{dt}\rho = -i[H_0 + H_I + H_L, \rho] + \mathcal{L}_\gamma \rho + \mathcal{L}_\kappa \rho, \qquad (1)$$

where ρ is the density matrix operator of this atom-cavity system. In a frame rotating with the frequency of the external field, the Hamiltonian of the atoms and the cavity field can be expressed as $H_0 = \hbar\Delta_A(S_+^1 S_-^1 + S_+^2 S_-^2) + \hbar\Delta_c(a^\dagger a)$ where $\Delta_A = \omega_A - \omega_L$ and $\Delta_c = \omega_c - \omega_L$ are the detunings with respect to the pump field frequency ω_L. Here, ω_A is the atomic transition frequency and $\omega_c = 2\pi/\lambda_c$ is the cavity frequency with λ_c being the wavelength of the cavity mode. S_+^i (S_-^i) is the atomic raising (lowering) operator of the ith ($i = 1,2$) atom, and $a^\dagger$ (a) is the photon creation (annihilation) operator. $H_I = \hbar\sum_{i=1,2} g_i(a^\dagger S_-^i + aS_+^i)$ is the interaction Hamiltonian between atoms and cavity, and the position-dependent atom-cavity coupling strength is given by $g_i = g\cos(2\pi z_i/\lambda_c)$ with z_i being the position of the ith atom. $H_L = \hbar\eta\sum_{i=1,2}(S_-^i + S_+^i)$ represents the interaction Hamiltonian between atoms and the coherent pumping field with Rabi frequency η. The Liouvillian $\mathcal{L}_\kappa\rho = \kappa(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a)$ is associated with the cavity decay at rate κ, whereas $\mathcal{L}_\gamma\rho = \gamma\sum_{i=1,2}(2S_-^i\rho S_+^i - S_+^i S_-^i\rho - \rho S_+^i S_-^i)$ arises from the spontaneous decay of the excited state of each atom at rate γ. As in Ref. [25], it is convenient to show the physical mechanism of the system by using the collective states $|gg\rangle$, $|\pm\rangle = (|eg\rangle \pm |ge\rangle)/\sqrt{2}$ and $|ee\rangle$ as basis to rewrite the Hamiltonian. At this end, the Hamiltonian can be expressed in terms of the collective operators $D_\pm^\dagger = (S_+^1 \pm S_+^2)/\sqrt{2}$, yielding $H_L = \sqrt{2}\hbar\eta(D_+^\dagger + D_+)$ and $H_I = H_+ + H_-$ with $H_\pm = \hbar g_\pm(aD_\pm^\dagger + a^\dagger D_\pm)/\sqrt{2}$ and $g_\pm = g(1 \pm \cos\phi_z)$. Here, $\phi_z = 2\pi\Delta z/\lambda_c$ is the phase shift between the radiation of two atoms with $\Delta z = z_2 - z_1$ being the distance between two atoms.

First, we consider the simplest condition of in-phase radiation of two atoms by taking $\phi_z = 0$ and $g_1 = g_2 = g$ (i.e., the two atoms have the same coupling strength). In this case the state $|-\rangle$ is uncoupled from the interaction with cavity.

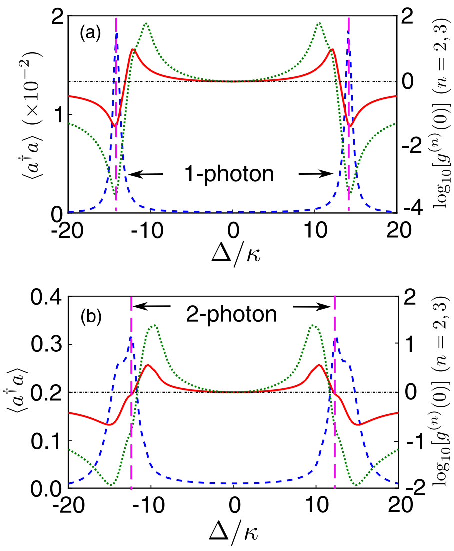

FIG. 2. The plots of field correlation function $g^{(2)}(0)$ (red solid curve), $g^{(3)}(0)$ (green dotted curve), and the mean photon number $\langle a^\dagger a\rangle$ (blue dashed curve) as a function of the normalized detuning Δ/κ in the case of $\phi_z = 0$. The black dash-dotted line indicates $g^{(2)}(0) = g^{(3)}(0) = 1$ for Poissonian statistics, and the pink dashed lines indicate the frequencies of the one-photon excitation in panel (a) and the two-photon excitation in panel (b). Here, we choose $\eta/\kappa = 0.1$ and 1 for panels (a) and (b), respectively. The coupling constant $g/\kappa = 10$ and the decay rate $\gamma/\kappa = 1$ are taken in both panels.

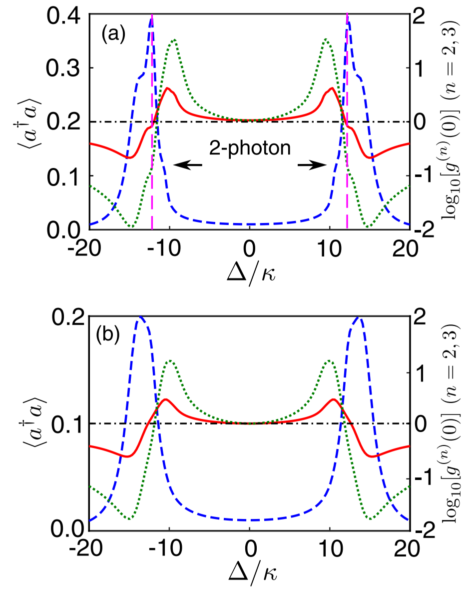

FIG. 3. Plots of the average photon number (blue dashed curve), the field correlation function $g^{(2)}(0)$ (red solid curves), and $g^{(3)}(0)$ (green dotted curves) versus the normalized detuning Δ/κ with $\phi_z = 0$, $g/\kappa = 10$, and $\eta/\kappa = 1$. The black dash-dotted line indicates $g^{(2)}(0) = g^{(3)}(0) = 1$ for Poissonian statistics and the pink dashed line indicates the frequency for two-photon excitation. Here, we take $\gamma = \kappa/3$ for panel (a) and $\gamma = 3\kappa$ for panel (b).

In the absence of the coherent pump field, it is easy to obtain a set of eigenstates $\Psi_\pm^{(1)} = \frac{\sqrt{2}}{2}|+,0\rangle \pm \frac{\sqrt{2}}{2}|gg,1\rangle$ in one-photon space with eigenvalues $\lambda_\pm^{(1)} = \hbar\omega_c \pm \sqrt{2}g\hbar$. Similarly, in two-photon space, we can also obtain a set of eigenstates $\Psi_0^{(2)} = -\frac{\sqrt{3}}{3}|gg,2\rangle + \frac{\sqrt{6}}{3}|ee,0\rangle$ with eigenvalue $\lambda_0^{(2)} = 2\hbar\omega_c$, and $\Psi_\pm^{(2)} = \frac{\sqrt{3}}{3}|gg,2\rangle + \frac{\sqrt{6}}{6}|ee,0\rangle \pm \frac{\sqrt{2}}{2}|+,1\rangle$ with eigenvalues $\lambda_\pm^{(2)} = 2\hbar\omega_c \pm \sqrt{6}g\hbar$ [shown in Fig. 1(b)]. The inclusion of the pumping field would dress these eigenstates. If the pump is weak then we can treat it as causing one- or two-photon transitions. The study of such transitions provides useful information.

III. NUMERICAL RESULTS FOR MULTIPHOTON BLOCKADE

Numerically solving Eq. (1) and assuming $\omega_c = \omega_A$ (i.e., $\Delta_A = \Delta_c \equiv \Delta$), we plot the mean photon number (blue dashed curve), the field correlation function $g^{(2)}(0) = \langle a^\dagger a^\dagger aa\rangle/(\langle a^\dagger a\rangle)^2$ (red solid curve), and $g^{(3)}(0) = \langle a^\dagger a^\dagger a^\dagger aaa\rangle/(\langle a^\dagger a\rangle)^3$ (green dotted curve) versus the normalized detuning Δ/κ in Fig. 2. When the atoms are driven by a weak pump field (e.g., $\eta/\kappa = 0.1$), we show in Fig. 2(a) that an excitation doublet can be observed at $\Delta = \pm\sqrt{2}g$ corresponding to the one-photon excitation (indicated by the pink dashed lines), leading to the transitions $\Psi^{(0)} = |gg,0\rangle \rightarrow \Psi_\pm^{(1)}$ shown in Fig. 1(b). Because the pump field is detuned for the transitions $\Psi_\pm^{(1)} \rightarrow \Psi_\pm^{(2)}$, the two-photon transition is off-resonance and thus weak leading to photon blockade. Therefore, the corresponding two-photon correlation function $g^{(2)}(0) \approx 0.05 < 1$ is the evidence of the two-photon blockade. At these frequencies we can also find that three-photon correlation function $g^{(3)}(0)$ is extremely small. If one increases the pump field, we find that the one-photon and two-photon excitations ($\Psi^{(0)} \rightarrow \Psi_\pm^{(1)} \rightarrow \Psi_\pm^{(2)}$) are both important, resulting in four peaks in the excitation spectrum at $\Delta = \pm\sqrt{2}g$ and $\Delta = \pm\sqrt{6}g/2$, respectively [see Fig. 2(b)]. Here, we take $\eta/\kappa = 1$ and the pink dashed lines indicate the frequencies for the two-photon excitations. It is clear to see that the field correlation functions at $\Delta = \pm\sqrt{2}g$ satisfy $g^{(2)}(0) \approx 0.3 < 1$ due to the two-photon blockade. The $g^{(3)}(0)$ remains much smaller than unity because the three-photon transition is highly detuned. More interestingly, we find that at $\Delta = \pm\sqrt{6}g/2$ the three-photon correlation function $g^{(3)}(0) = 0.19$ accompanied with simultaneous two-photon correlation $g^{(2)}(0)$ ($\approx$1.04) of nearly unity. Thus at the two-photon excitation condition the third photon is not absorbed, leading to *three-photon blockade*. Here, we must point out that, in the case of in-phase radiations of two atoms, the frequency regime to realize the three-photon blockade [i.e., $g^{(2)}(0) > 1$ and $g^{(3)}(0) < 1$] is very narrow, so that it is not a good candidate for experiments.

The numerical calculations show that the blockade is sensitive to the atomic decay rate γ as compared with κ. This is illustrated in Fig. 3. For a small atomic decay rate, e.g., $\gamma = \kappa/3$, the two-photon transition can be excited much more easily than in the case of $\gamma = \kappa$, so that the mean photon number at $\Delta = \pm\sqrt{6}g/2$ becomes much larger than that at $\Delta = \pm\sqrt{2}g$ and the three-photon blockade can also be observed with $g^{(2)}(0) \approx 0.8$ [see Fig. 3(a)] [27]. However, in the case of large decay rate, i.e., $\gamma = 3\kappa$, the two-photon transition can hardly be excited since the atom rapidly decays

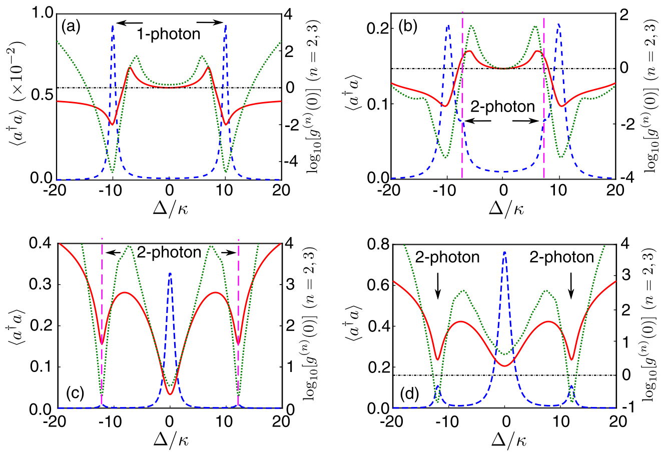

FIG. 4. Plots of the average photon number (blue dashed curve), the field correlation function $g^{(2)}(0)$ (red solid curves), and $g^{(3)}(0)$ (green dotted curves) versus the normalized detuning Δ/κ. The phase shift is chosen as (a), (b) $\phi_z = \pi/2$ and (c), (d) π, respectively. The pump field is (a) $\eta/\kappa = 0.1$, (b), (c) 1.0, and (d) 2.0.

to the ground state after one-photon transition takes place. Therefore, the mean photon number decreases slightly and the peaks associated with the one-photon and two-photon excitations merge together [shown in Fig. 3(b)]. As a result, the width of the peak becomes large. It is worth pointing out that at $\Delta = \pm\sqrt{6}g/2$ we can obtain $g^{(2)}(0) \approx 1.25$, $g^{(3)}(0) \approx 0.4$, and we can find $g^{(2)}(0) \approx 0.32$, $g^{(3)}(0) \approx 0.02$ at $\Delta = \pm\sqrt{2}g$, indicating that we can realize two-photon and three-photon blockade simultaneously.

In the following, we study the case of two atoms for $\phi_z \neq 0$ so that $g_1 \neq g_2$. In this case additional pathways open up as the asymmetric state $|-\rangle = (|eg\rangle - |ge\rangle)/\sqrt{2}$ also participates in the transitions. To understand the physical mechanism, one can use a set of collective states to describe the atomic transitions. Defining $\alpha = (1+\cos\phi_z)/(1+\cos^2\phi_z)^{1/2}$ and $\beta = (-1+\cos\phi_z)/(1+\cos^2\phi_z)^{1/2}$, we can find three eigenstates in one-photon space, i.e., $\Psi_0^{(1)} = \frac{\sqrt{2}}{2}\alpha|-,0\rangle + \frac{\sqrt{2}}{2}\beta|+,0\rangle$ with eigenvalue $\lambda_0^{(1)} = \hbar\omega_c$ and $\Psi_\pm^{(1)} = \mp\frac{\sqrt{2}}{2}|gg,1\rangle - \frac{1}{2}\beta|-,0\rangle + \frac{1}{2}\alpha|+,0\rangle$ with eigenvalues $\lambda_\pm^{(1)} = \hbar\omega_c \pm \hbar g_0(1+\cos^2\phi_z)^{1/2}$, respectively. In two-photon space, we can obtain four eigenstates, labeled as $\Psi_0^{(2)} = -\frac{\sqrt{3}}{3}|gg,2\rangle + \frac{\sqrt{6}}{3}|ee,0\rangle$ and $\Phi_0^{(2)} = \frac{\sqrt{2}}{2}\alpha|-,1\rangle + \frac{\sqrt{2}}{2}\beta|+,1\rangle$ with the same eigenvalue $\lambda_0^{(2)} = 2\hbar\omega_c$ and $\Psi_\pm^{(2)} = \frac{\sqrt{3}}{3}|gg,2\rangle \mp \frac{1}{2}\beta|-,1\rangle \pm \frac{1}{2}\alpha|+,1\rangle + \frac{\sqrt{6}}{6}|ee,0\rangle$ with eigenvalues $\lambda_\pm^{(2)} = 2\hbar\omega_c \pm \hbar g[3(1+\cos^2\phi_z)]^{1/2}$, respectively. As shown in Figs. 1(b)–1(d), one can easily obtain the transition strength by calculating the dipole matrix element of operator $\sqrt{2}\eta(D_+^\dagger + D_+)$. These dressed states along with some of the important transitions are shown in Figs. 1(c) and 1(d).

In Fig. 4, we plot the mean photon number $\langle a^\dagger a\rangle$ and field correlation functions $g^{(2)}(0)$ and $g^{(3)}(0)$ versus the normalized detuning Δ/κ with different pump field and phase shift. We show that the properties of the quantum statistics of the system changes significantly with the collective behavior of two atoms. First, we consider the case of $\pi/2$ phase shift with pump field $\eta = 0.1\kappa$ [Fig. 4(a)] and $\eta = \kappa$ [Fig. 4(b)], we find that the one-photon excitations are dominant although the eigenstate become much more complicated, as shown in Fig. 1(c). Now the one-photon excitations occur at $\Delta = \pm g$. Comparing the results of in-phase radiation (see Fig. 2), it is obvious that the two-photon blockade phenomenon is even more pronounced. We find that $g^{(2)}(0) \approx 0.01$ in Fig. 4(a) and $g^{(2)}(0) \approx 0.03$ in Fig. 4(b), which is nearly ten times less than that of the in-phase condition for a strong pump field. This characteristic can be explained by calculating the dipole matrix elements associated with the one-photon and two-photon excitations. In the $\pi/2$ case, the two-photon transition strength is much smaller than that in the in-phase condition, so that the second-order photon-photon correlation drops significantly. For the out-phase case (i.e., $\phi_z = \pi$), the two-photon excitation becomes important because $\Psi^{(0)} \to \Psi_\pm^{(1)}$ transitions are not allowed [see Fig. 1(d)]. As a result, we can observe a single peak at the $\delta = 0$ and simultaneously both field correlation functions $g^{(2)}(0)$ and $g^{(3)}(0)$ are larger than unity because the two-photon and the three-photon pathways are fully resonant [see Figs. 4(c) and 4(d)]. If we increase the strength of the pump to $\eta = 2\kappa$ [Fig. 4(d)], then the two-photon excitations $\Psi^{(0)} \to \Psi_\pm^{(2)}$ become strong enough to be observed. In the excitation spectrum shown in Fig. 4(c) there exist three peaks associated with the one-photon excitations ($\Delta = 0$) and two-photon excitations ($\Delta = \pm\sqrt{6}g/2$). At $\Delta = 0$, we find $g^{(2)}(0) \approx 2.15$ and $g^{(3)}(0) \approx 3.45$ since the two-photon and three-photon pathways are fully resonant. At $\Delta = \pm\sqrt{6}g/2$, we find $g^{(2)}(0) \approx 36.55$ and $g^{(3)}(0) \approx 1.89$ so that the three-photon blockade is not well developed for very weak pump. Increasing the pump field, the three-photon blockade occurs with two-photon bunching, as shown in Fig. 4(d). The corresponding field correlation functions are given by $g^{(2)}(0) \approx 3.54$ and $g^{(3)}(0) \approx 0.18$. The very large values of $g^{(2)}(0)$ and $g^{(3)}(0)$ occur in the region where photon numbers are very small. These large values are not very meaningful because the measurements of quantum statistics in regions of very small photon numbers are difficult. We have thus seen that the collective two-atom system can lead to a wide variety of multiphoton blockade.

We conclude this paper by comparing results with the single-atom case (see Fig. 5). In Fig. 5(a), we take $\eta/2\pi = 1.6$ MHz and other system parameters are given by $\kappa/2\pi = 2.0$ MHz, $\gamma/2\pi = 3.0$ MHz, and $g/2\pi = 20$ MHz according to Ref. [26]. Obviously, the two-photon blockade phenomenon can be observed at $\Delta = \pm g$ since the two-photon excitations are nonresonant, which has been widely studied in the literature [12,14,15]. The small peaks at $\Delta = \pm\sqrt{2}g/2$ correspond to the two-photon excitation process if the pump field frequency satisfies $\omega_L = \omega_c \pm \sqrt{2}g/2$. We find that the photon-photon correlation satisfy $g^{(2)}(0) \approx 1.8$ and $g^{(3)}(0) \approx 0.8$ at $\Delta/2\pi = \pm 15$ MHz (near two-photon excitation). Thus the allowed two-photon transition leads to $g^{(2)}(0) > 1$; however, the three-photon blockade occurs which has been recently reported [26]. In Fig. 5(b), we show the numerical results of a two-atom system with π phase shift. The system parameters are the same as those used in Fig. 5(a), but we take $\eta/2\pi = 6$ MHz. We find that the photon-photon correlation satisfies $g^{(2)}(0) \approx 2.0$ and $g^{(3)}(0) \approx 0.2$ at $\Delta = \pm\sqrt{6}g/2$ (two-photon excitation).

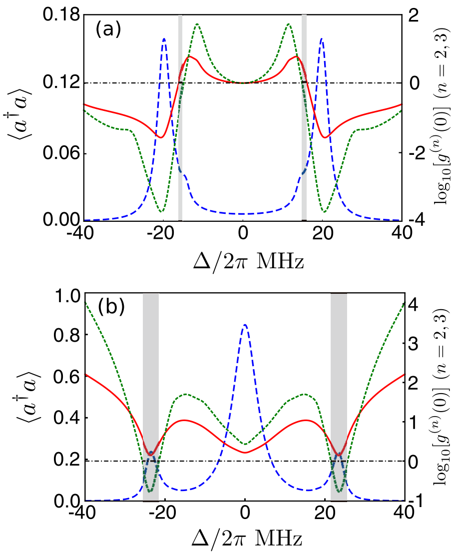

FIG. 5. Plots of field correlation function $g^{(2)}(0)$ (red solid curve), $g^{(3)}(0)$ (green dotted curve), and mean photon number (blue dashed curve) as a function of normalized detuning Δ/κ for (a) one-atom system and (b) two atoms system with π phase shift. The black dash-dotted line indicates $g^{(2)}(0) = g^{(3)}(0) = 1$ for Poissonian statistics and the gray area indicates the frequency regime to realize three-photon blockade. Here, we choose (a) $\eta/2\pi = 1.6$ MHz and (b) 6 MHz. Other system parameters are given by $\kappa/2\pi = 2.0$ MHz, $\gamma/2\pi = 3.0$ MHz, and $g/2\pi = 20$ MHz according to Ref. [26].

Comparing these two plots, we find some advantages for the system with two out-of-phase atoms: (i) The frequency regime to realize three-photon blockade in a two-atom system is much wider than that in a one-atom system. (ii) The three-photon blockade phenomenon can be significantly improved in the two-atom system since the one-photon excitations are not allowed in the out-of-phase case.

IV. CONCLUSIONS

In conclusion, we have shown that a collective system of two two-level atoms in a high-quality cavity can display a much richer variety of photon blockade phenomena. This arises from the dressed state structure of the system of interacting cavity field and atoms. The dressed state structure and allowed transitions depend on the two coupling constants of the atoms to the cavity field. We found that, in the case of in-phase radiations of two atoms, the three-photon blockade can be observed with two-photon bunching if the excitation frequency is tuned to the two-photon transition. The collective behavior depends on the relative phase, i.e., the location of atoms can change significantly the properties of quantum statistics of the system. In the case of a $\pi/2$ phase shift the two-photon blockade can become quite prominent even under conditions of a weak pump field, if the coupling constants g_1 and g_2 are out of phase. In addition, the two-photon blockade phenomenon disappears, but the three-photon blockade phenomenon is significantly improved. Thus the photon blockade effects in two-atom systems can be quite different from the case of single-atom systems. The two-atom system displays very pronounced three-photon blockade which should be observable in experiments of the type reported recently. The three-photon blockade should be useful for generating nonclassical photon pairs.

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