

Strong photon antibunching and photon pair emission in a two-atom cavity QED system with van der Waals interaction

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We investigate the generation of strong antibunching photons/photon pairs in a two-atom cavity QED system leveraging interatomic van der Waals (vdW) interaction. We show that the vdW interaction shifts the energy of double excited state, enabling the suppression of two-photon excitation via destructive interference in a diamond configuration. This leads to antibunching photon emission with extremely high purity. Furthermore, by tuning the vdW interaction strength, the conditions for conventional and unconventional photon blockades can be satisfied simultaneously, which significantly enhances the generation of single photon. Conversely, at the antiblockade excitation frequency, quantum Monte Carlo simulations demonstrate the feasibility of generating antibunching photon pairs with a high purity and reasonable leaking rate by selecting appropriate driving field Rabi frequency and vdW interaction strength. These findings on enhanced photon antibunching and photon pair emission may lead to more efficient and brighter quantum light sources, which is a key mechanism for optical quantum devices.

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I. INTRODUCTION

Photon blockade (PB) is a quantum mechanical effect that not only leads to a nonclassical state of light with antibunching emission of photons [1–3], but also serves as a tool to unveil various quantum features at the single-photon level [4,5]. Conventional photon blockade (CPB) originates from the anharmonic splitting of eigenstates in cavity quantum electrodynamics (QED) with strong couplings between atoms and cavity [6,7]. It plays a crucial rule in quantum information [8–10], generating nonclassical lights [11–15] and building single-photon sources [16–18]. Contrary to the CPB, unconventional photon blockade (UCPB) has been proposed in the weak coupling regime [19–21]. It utilizes quantum destructive interference (DI) of transition pathways to eliminate the two-photon transitions [22–24]. As a result, extremely strong photon blockade behavior can be observed under the condition of UCPB. However, although the purity of antibunching photons generated by UCPB is very high, the emission rate of photons is much lower than that generated by CPB.

Recently, both CPB and UCPB have been extensively studied and experimentally demonstrated in various QED systems, including cavity QED systems [25–35], circuit-QED systems [36–39], optomechanical systems [40–47], and so on [48–52]. Similar method based on energy level anharmonicity (ELA) has been used to realize two-photon blockade, facilitating the resonant absorption of two photons and subsequently reducing the transition probability of the third photon [53–60]. This quantum scissor can thereby provide the emission of antibunching photon pairs [3,61]. Furthermore, multiphoton blockade can also be studied via multiphoton transitions [60]. While the third-order correlation function can readily be suppressed (indicating blockade of the third photon), achieving a sufficiently strong two-photon bunching, where the second-order correlation function is substantially greater than unity, remains elusive. This limitation hinders the efficient generation of strongly antibunching photon pairs, which are a key signature of two-photon blockade.

Since the key to achieving PB is to control energy levels and transition probabilities, the interaction between atoms provides a degree of freedom [62–67]. In atomic systems, the interatomic dipole-dipole interaction and the van der Waals (vdW) interaction are good candidates for manipulating atomic states. The former leads to energy shifts of single-atom excitation states, which can be used to realized strong single PB via the destructive interference [66]. However, the latter shifts the energy of the two-atom excitation state and leads to the well-known Rydberg blockade phenomenon, where only one atom can be excited to the Rydberg state [68–70]. In contrast to the Rydberg blockade, a well-detuned laser can

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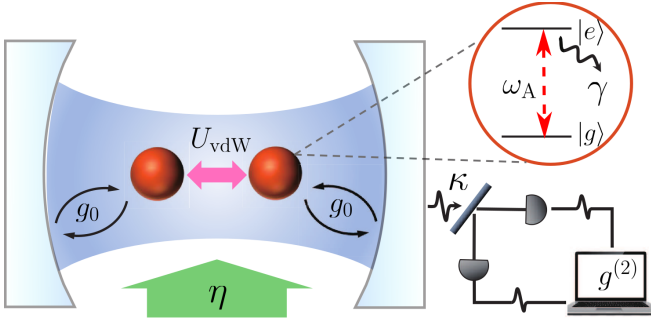


FIG. 1. Schematic diagram of two identical two-level atoms cavity QED system. Each atom is simplified as a two-level scheme with a ground state $|g\rangle$ and an excited state $|e\rangle$. The coupling between each atom and the cavity is labeled by g_0 , and the van de Waals interaction between two atoms is denoted by U_{vdW} . A coherent field with Rabi frequency η drives two atoms in phase. The field leaks from the right side of the cavity with the rate κ , and the excited state of each atom decays with the rate γ .

compensate the vdW-induced energy shift and two atoms can be simultaneously excited to the Rydberg states, which is known as the antiblockade phenomenon [71,72].

In this paper, we investigate how to realize strong photon antibunching and photon pair-emission via the van der Waals interaction in a two-atom cavity QED system. In the presence of the interatomic vdW interaction, we show that the double excited state is shifted. By choosing a specific driving field frequency, the two-photon excitation state will be forbidden due to the destructive interference effect with a diamond configuration. As a result, strong photon antibunching can be achieved based on the UCPB phenomenon. Adjusting the vdW interaction strength, conditions for the generation of CPB and UCPB can be satisfied simultaneously and the emission rate of single photon can be enhanced significantly. We also show, at the excitation frequency of antiblockade, photons are emitted in pairs with steady-state second-order correlation function $g^{(2)}(0) \gg 1$ and $g^{(3)}(0) \ll 1$, i.e., two-photon blockade. By choosing suitable driving field Rabi frequency and the vdW interaction strength, we carry out the quantum Monte Carlo (MC) simulation and demonstrate the possibility for the emission of antibunched photons and photon pairs with reasonable leaking rate.

II. MODEL

As shown in Fig. 1, we consider that two identical two-level atoms with the transition frequency ω_A are trapped in a single-mode cavity with the resonance frequency ω_{cav} . To realize strong single-photon and two-photon emissions, two atoms should be prepared close enough to realize the van de Waals interaction, denoted by U_{vdW} . Then, we coherently drive these two atoms in phase by an optical field with angular frequency ω_d . In the frame rotating with the frequency of the driving field, the Hamiltonian of atoms and cavity is $H_0 = \Delta_{\text{cav}} a^\dagger a + \Delta_A (\sigma_+^{(1)} \sigma_-^{(1)} + \sigma_+^{(2)} \sigma_-^{(2)})$ where $a^\dagger$ (a) is the photon creation (annihilation) operator and $\sigma_\pm^{(j)}$ is the atomic raising or lowering operator for the j th atom ($j = 1-2$). The atom and cavity detunings are defined as

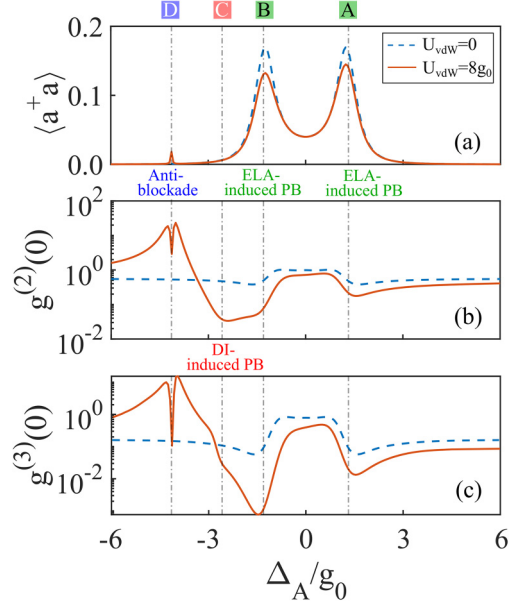


FIG. 2. (a) Cavity excitation spectrum $\langle a^\dagger a \rangle$, (b) the steady-state second-order correlation function $g^{(2)}(0)$, and (c) the third-order correlation function $g^{(3)}(0)$ vs the normalized atomic detuning Δ_A/g_0 . The blue dashed curves and the red solid curves in panels (a)–(c) represent the van der Waals interaction strength $U_{\text{vdW}} = 0$ and $U_{\text{vdW}} = 8g_0$, respectively. Here, labels A and B denote the frequency for the energy level anharmonicity-induced single-photon blockade, while label C denotes the frequency for the destructive interference-induced single-photon blockade. In addition, label D indicates the frequency for the antiblockade behavior, which leads to the two-photon emission with $g^{(2)}(0) > 1$ and $g^{(3)}(0) < 1$.

$\Delta_A = \omega_A - \omega_d$ and $\Delta_{\text{cav}} = \omega_{\text{cav}} - \omega_d$, respectively. In our system, the Hamiltonian for the interaction between atoms and cavity is described by $H_I = \sum_j g_j (a^\dagger \sigma_-^{(j)} + a \sigma_+^{(j)}) + U_{\text{vdW}} \sigma_+^{(1)} \sigma_-^{(1)} \sigma_+^{(2)} \sigma_-^{(2)}$, where the first term represents the atom-cavity interaction with g_j being the coupling strength for the j th atom. The second term indicates the van der Waals interaction between two atoms [73–75]. The Hamiltonian for the driving field is $H_d = \eta \sum_j (\sigma_+^{(j)} + \sigma_-^{(j)})$ with the Rabi frequency η .

In general, the dynamical behavior of such a system can be described by using the master equation, i.e.,

$$\frac{d\rho}{dt} = -i[H_{\text{sys}}, \rho] + \mathcal{L}_{\text{atom}}(\rho) + \mathcal{L}_{\text{cav}}(\rho), \quad (1)$$

where the Hamiltonian of the system is $H_{\text{sys}} = H_0 + H_I + H_d$. The last two terms in Eq. (1) represent the decays of atoms and the cavity field, which are defined as $\mathcal{L}_{\text{atom}}(\rho) = \gamma \sum_j (2\sigma_-^{(j)} \rho \sigma_+^{(j)} - \sigma_+^{(j)} \sigma_-^{(j)} \rho - \rho \sigma_+^{(j)} \sigma_-^{(j)})$ and $\mathcal{L}_{\text{cav}}(\rho) = \kappa \sum_j (2a \rho a^\dagger - a^\dagger a \rho - \rho a^\dagger a)$, respectively. The field decay rate is described by 2κ , while the excited state decays with the rate 2γ . Without losing the generality, we assume $\Delta_{\text{cav}} = \Delta_A$, i.e., $\omega_{\text{cav}} = \omega_A$, and $g_1 = g_2 = g_0$ in the following calculation. Then, we perform a steady-state numerical simulation based on the master equation [i.e., Eq. (1)] with the inverse power method. In Fig. 2, the cavity excitation spectrum $\langle a^\dagger a \rangle$ [panel (a)], the steady-state second-order correlation function $g^{(2)}(0) \equiv \langle a^\dagger a^\dagger a a \rangle / \langle a^\dagger a \rangle^2$

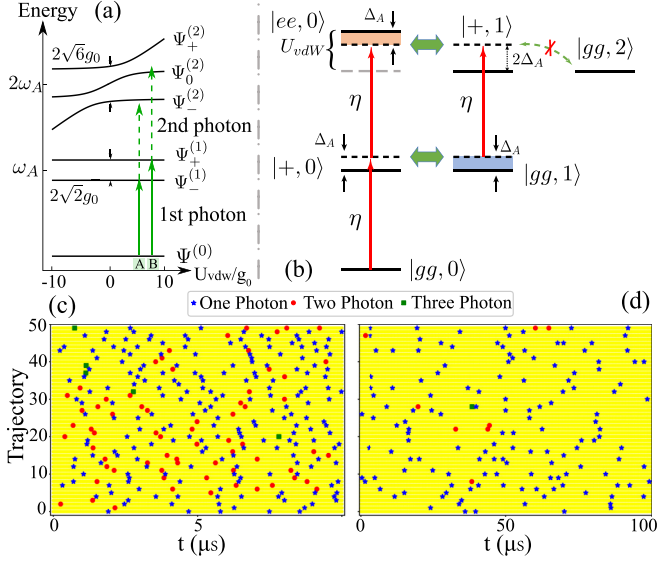


FIG. 3. (a) Dressed state picture of two-atom cavity QED system with the van der Waals interaction. The green arrows indicate the excitation pathways by absorbing a single photon, while the dashed ones denotes the blockade of the excitation by absorbing the second photon. This EIA-induced PB phenomenon leads to $g^{(2)}(0) < 1$ labeled by A and B in Fig. 2. (b) Collective state picture and the corresponding transition pathways of the system. When the detunings of states $|ee, 0\rangle$ and $|gg, 1\rangle$ have the same magnitude but opposite sign, the destructive interference with a diamond configuration is formed, which prevents the state $|gg, 2\rangle$ from being excited. Panels (c) and (d) show the MC simulations of photon emissions with the frequency labeled by A and C, respectively. They record photon clicks when the system undergoes a quantum jump.

[panel (b)], and the steady-state third-order correlation function $g^{(3)}(0) \equiv \langle a^\dagger a^\dagger a^\dagger aaa \rangle / \langle a^\dagger a \rangle^3$ [panel (c)] are depicted as a function of the normalized atomic detuning Δ_A/g_0 . Here, the van der Waals interaction strength is chosen as $U_{vdW} = 0$ (blue dashed curves) and $U_{vdW} = 8g_0$ (red solid curves) [76], respectively. Other system parameters are chosen as $g_0 = 5$ MHz, $\eta = 1$ MHz, $\kappa = 3$ MHz, and $\gamma = 5$ kHz, respectively [59,77]. To ensure numerical convergence, we truncate the Hilbert space of the cavity mode to a size of 30 in our numerical simulation.

In the absence of the van der Waals interaction, i.e., $U_{vdW} = 0$ (blue dashed curves in Fig. 2), there exist two peaks in the cavity excitation spectrum at the frequencies of $\Delta_A = \pm\sqrt{2}g_0$, where single-photon blockade behavior can be observed with $g^{(3)}(0) < g^{(2)}(0) < 1$ (labeled by A and B). The physical mechanism is well known as the energy level anharmonicity induced by the atom cavity coupling, demonstrated in Fig. 3(a). When the driving field frequency is tuned to be resonant with the $\Psi^{(0)} \leftrightarrow \Psi_{\pm}^{(1)}$ transition frequency, the $\Psi_{\pm}^{(1)} \leftrightarrow \Psi_{\pm}^{(2)}$ transition involving the absorption of the second cavity photon is prohibited due to the energy difference. In the presence of the van der Waals interaction, e.g., $U_{vdW} = 8g_0$ (red solid curves in Fig. 2), one can observe three peaks in the cavity excitation spectrum at the frequencies of $\Delta_A = \pm\sqrt{2}g_0$ and $\Delta_A = -U_{vdW}/2$, respectively. It is clear to see that the ELA-induced photon blockade behavior can be improved at

$\Delta_A = \pm\sqrt{2}g_0$. Likewise, the corresponding physical mechanism can be described by exploring the dressed states shown in Fig. 3(a). In this case, eigenvalues and eigenstates of the dressed states can be obtained by diagonalizing the Hamiltonian $H = \omega_A(a^\dagger a + \sigma_+^{(1)}\sigma_-^{(1)} + \sigma_+^{(2)}\sigma_-^{(2)}) + \sum_j g_j(a^\dagger\sigma_-^{(j)} + a\sigma_+^{(j)}) + U_{vdW}\sigma_+^{(1)}\sigma_-^{(1)}\sigma_+^{(2)}\sigma_-^{(2)}$ in one-photon and two-photon spaces, respectively. As shown in Fig. 3(a), increasing the van der Waals interaction, only dressed states in two-photon space will be affected. As a result, this atomic interaction induced energy shift in two-photon space enhances the energy anharmonicity, leading to the improvement of the single-photon blockade behavior, specially at the frequency of $\Delta_A = -\sqrt{2}g_0$ (see results at detuning B in Fig. 2). In addition, at the frequency of antiblockade regime, i.e., $\Delta_A = -U_{vdW}/2$, two atoms can be excited simultaneously and two-photon blockade behavior can be observed with $g^{(2)}(0) \gg 1$ and $g^{(3)}(0) < 1$ (see results at detuning D in Fig. 2). More features of two-photon emissions will be discussed in Sec. IV.

III. INTERFERENCE-INDUCED STRONG PHOTON ANTIBUNCHING

In Fig. 2, there exists a “magic” detuning labeled by C, representing the destructive interference-induced PB, where the second-order correlation function is much smaller than that at the frequencies for ELA-induced PB, i.e., $\Delta_A = \pm\sqrt{2}g_0$. To understand this interesting phenomenon, we use the collective operators to rewrite the Hamiltonian of the system, which yields

$$H' = \Delta_{\text{cav}}a^\dagger a + \Delta_A J_z + \sqrt{2}g_0(aJ_+ + a^\dagger J_-) + \sqrt{2}\eta(J_+ + J_-) + \frac{U_{vdW}}{2}(1 + J_z)J_z, \quad (2)$$

where atomic spin collective operators are defined as $J_z = \sigma_z^{(1)} + \sigma_z^{(2)}$, $J_{\pm} = (\sigma_{\pm}^{(1)} + \sigma_{\pm}^{(2)})/\sqrt{2}$. As discussed in the previous section, the last term (van der Waals interaction) in the Hamiltonian H' only affects the state $|ee, n\rangle$, leading to an energy shift of U_{vdW} . The collective states and their corresponding transition pathways in one-photon and two-photon spaces are demonstrated in Fig. 3(b).

Clearly, there exist two transition pathways for the excitation of the state $|gg, 2\rangle$, i.e., $|gg, 0\rangle \xrightarrow{\eta} |+, 0\rangle \xrightarrow{\eta} |ee, 0\rangle \xrightarrow{\sqrt{2}g_0} |+, 1\rangle \xrightarrow{2g_0} |gg, 2\rangle$ and $|gg, 0\rangle \xrightarrow{\eta} |+, 0\rangle \xrightarrow{\sqrt{2}g_0} |gg, 1\rangle \xrightarrow{2g_0} |+, 1\rangle \xrightarrow{2g_0} |gg, 2\rangle$. Therefore, the destructive interference with a diamond configuration will be formed if the detunings of states $|gg, 1\rangle$ and $|ee, 0\rangle$ have the same magnitude, but opposite signs, i.e., $U_{vdW} + 2\Delta_A = -\Delta_A$. Thus, the “magic” detuning can be derived as $\Delta_{\text{magic}} \equiv \Delta_A = -U_{vdW}/3$ [23]. Under this condition, the excitation of the state $|gg, 2\rangle$ can be prohibited due to the destructive interference effect and strong antibunching photons can be observed, i.e., the DI-induced PB phenomenon.

Next, we apply a quantum Monte Carlo approach to demonstrate this single PB behavior [78]. Here, we track individual system trajectories and record photon clicks whenever the system undergoes a quantum jump [79–81]. Figures 3(c) and 3(d) present a series of detection events with the detunings $\Delta_A = \sqrt{2}g_0$ (ELA) and $\Delta_A = -U_{vdW}/3$ (DI), respectively.

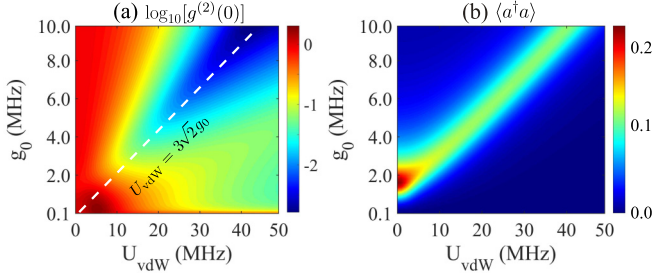


FIG. 4. Plots of the second-order correlation function $g^{(2)}(0)$ [panel (a)] and the mean photon number $\langle a^\dagger a \rangle$ [panel (b)] as a function of the van der Waals interaction strength U_{vdW} and the atom-cavity coupling strength g_0 . The dashed line in panel (a) corresponds to the condition of $U_{\text{vdW}} = 3\sqrt{2}g_0$, where the condition for ELA-induced PB coincides with the condition for DI-induced PB, leading to strong PB behavior with reasonable mean photon number.

The horizontal axis represents time, and each point denotes a single-photon, two-photon, or multiphoton emission event. As shown in Fig. 3(c), except for about 10% unexpected photon emission processes such as two-photon and multiphoton processes, most photons leaking from the cavity are antibunching due to ELA-induced PB at the detuning $\Delta_A = \sqrt{2}g_0$. However, in the regime of DI-induced PB, unexpected photon emissions (such as two-photon and multiphoton processes) are significantly suppressed due to the destructive interference effect, yielding an approximately 100% antibunching photon emission as shown in Fig. 3(d). Comparing these two figures, we also notice that total photon emission events in Fig. 3(d) is less than that in Fig. 3(c), since the destructive interference usually results in very small mean photon number, which prevents it from practical applications. To overcome this shortcoming, we find a condition satisfying both ELA- and DI-induced PBs, i.e., setting $\Delta_A = -\sqrt{2}g_0 = -U_{\text{vdW}}/3$. Therefore, one can obtain $U_{\text{vdW}} = 3\sqrt{2}g_0$, where strong single-photon blockade behavior can be observed with reasonable mean photon number. Figure 4 shows how the second-order correlation function $g^{(2)}(0)$ [panel (a)] and the corresponding mean photon number [panel (b)] change as a function of U_{vdW} and g_0 . Here, we set $\Delta_A = -U_{\text{vdW}}/3$ and other system parameters are the same as those used in Fig. 2. It is clear to see that, under the condition of $U_{\text{vdW}} = 3\sqrt{2}g_0$, the single-photon blockade behavior can be significantly improved and the corresponding second-order correlation function $g^{(2)}(0)$ decreases to the value less than 10^{-2} as the van der Waals interaction strength U_{vdW} increases. At the same time, the mean photon number remains unchanged with $\langle a^\dagger a \rangle \approx 0.1$. The corresponding leaking rate of the cavity photons reaches up to 100 kHz, which can be observed in experiments with current experimental techniques.

IV. ANTIBLOCKADE-INDUCED EMISSION OF PHOTON PAIRS

Now, let us focus on the two-photon emission at the frequency of antiblockade regime, i.e., $\Delta_A \approx -U_{\text{vdW}}/2$. Under this condition, as shown in Fig. 3(b), the state $|ee, 0\rangle$ can be resonantly excited via the two-photon process since the state $|ee, 0\rangle$ is shifted by an amount of U_{vdW} , which

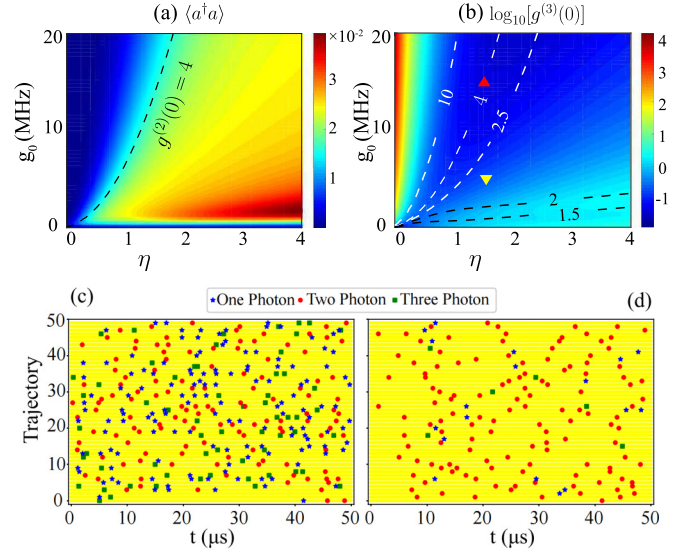


FIG. 5. Plots of the mean photon number $\langle a^\dagger a \rangle$ [panel (a)] and the third-order correlation function $g^{(3)}(0)$ [panel (b)] as a function of the atom cavity coupling strength g_0 and the driving field Rabi frequency η . The dashed curves denote the values of the second-order correlation function $g^{(2)}(0)$. Panels (c) and (d) show the MC simulations of photon emissions with the atom cavity coupling strength of panel (b) at $g_0 = 5$ MHz (labeled by a yellow inverted triangle) and $g_0 = 15$ MHz (labeled by a red triangle), respectively.

compensates the probe field detuning $2\Delta_A$. As a result, the transition pathway $|gg, 0\rangle \leftrightarrow |ee, 0\rangle$ is dominant so that photons are strongly bunched with $g^{(2)}(0) \gg 1$. Also notice that the excitation of the state $|+, 1\rangle$ is still off resonant, which prevents the excitation of the state in three-photon space (such as the state $|ee, 1\rangle$), leading to the third-order correlation function $g^{(3)}(0) < 1$. Therefore, two-photon blockade takes place and one can observe the emission of bunching photon pairs. To verify the above analysis, we calculate the mean photon number $\langle a^\dagger a \rangle$, the steady-state second-order correlation function $g^{(2)}(0)$, and the third-order correlation function $g^{(3)}(0)$, respectively. In Fig. 5(a), the mean photon number is depicted as a function of the coupling strength g_0 and the driving field Rabi frequency η . The corresponding steady-state third-order correlation function $g^{(3)}(0)$ is demonstrated in Fig. 5(b). In Figs. 5(a) and 5(b), the dashed curves denote values of the second-order correlation function $g^{(2)}(0)$. Here, the detuning is chosen as $\Delta_A = -U_{\text{vdW}}/2$ and other system parameters are the same as those used in Fig. 4. It is clear to see that strong two-photon blockade with $g^{(2)}(0) \gg 1$ and $g^{(3)}(0) < 0.1$ can be achieved by choosing suitable driving field η and atom cavity coupling strength g_0 .

To gain insights into the dynamical properties of photon emission, we apply a quantum MC approach to solve the master equation with driving field Rabi frequency $\eta = 1.5$. Figures 5(c) and 5(d) present a series of detection events for the atom cavity coupling strength of Fig. 5(b) at $g_0 = 5$ MHz (labeled by a yellow inverted triangle) and $g_0 = 15$ MHz (labeled by a red triangle), respectively. The horizontal axis represents time, and each point denotes a detection event. Although the number of total detection events in Fig. 5(d) is

far less than that in Fig. 5(c) (corresponding to a smaller mean photon number), single-photon and multiphoton emissions can be significantly suppressed under the strong coupling regime, i.e., $g_0 = 15$ MHz, leading to a purified emission of bunching photon pairs [see Fig. 5(d)].

V. CONCLUSION

Eventually, we briefly discuss the experimental feasibility of realizing such a two-atom cavity QED system. In experiments, such a system can be realized by combining outstanding achievements in the field of cavity QED [82,83] and Rydberg atom experiments [84,85]. For example, a far red-detuned laser system allows to create the microscopic optical tweezers used to trap the single ^{87}Rb atom. Then, one can use optical photons and Rydberg atoms are prepared via the two-photon transition. The vdW interaction strength between the two atoms can be adjusted by changing the distance of two atoms. Given the distance between two atoms is $R = 5$ μm and the principal quantum number $n = 62$, the corresponding vdW interaction strength $U_{\text{vdW}} \equiv C_6/R^6 \approx 47$ MHz with the coefficient $C_6 \approx 730$ GHz μm^6 [86].

In conclusion, we have shown how the van der Waals interaction in a two-atom cavity QED system offers a powerful knob for controlling photon statistics. By harnessing the vdW-induced shift in the two-atom excited state, we can achieve strong photon antibunching through destructive interference in a diamond configuration, a direct consequence of the unconventional photon blockade phenomenon. We also found that carefully adjusting the vdW interaction strength allows for an overlap between conventional and unconventional

photon blockade conditions, leading to significantly enhanced single-photon emission with extremely high purity and reasonable leaking rate. Interestingly, our investigation also revealed the emission of antibunching photon pairs at the antiblockade excitation frequency, characterized by the second-order correlation function $g^{(2)}(0) \gg 1$. Through quantum Monte Carlo simulations, we have confirmed the feasibility of generating antibunching photon pairs by optimizing the driving field Rabi frequency and vdW interaction strength, with a purity over 90% [the third-order correlation function $g^{(3)}(0) < 0.1$]. These findings introduce a method for generating nonclassical light, which opens the door to a variety of applications including quantum metrology and sensing, the photon transistor, and quantum information processing and communication.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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