Photon pair source via two coupling single quantum emitters*

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We study the two coupling two-level single molecules driven by an external field as a photon pair source. The probability of emitting two photons, P_2, is employed to describe the photon pair source quality in a short time, and the correlation coefficient R_{AB} is employed to describe the photon pair source quality in a long time limit. The results demonstrate that the coupling single quantum emitters can be considered as a stable photon pair source.

Keywords: coupling quantum emitters, photon source, photon statistics

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1. Introduction

The deterministic single photon source and photon pair source are important for quantum information, quantum computing, and quantum communications.[1–5] In quantum communications, most of the communication protocols are based on entangled photon pairs.[6–9] Generally, there are two ways to generate entangled photon pairs: one is to use parametric down conversion in nonlinear optical media, and the emission photons satisfy the Poissonian statistics;[10] the other way is to use the radiative recombination of the bi-exciton state in single quantum dots.[10] The last method cannot avoid the effect of the single photon process.

We suggest that two single quantum emitters are coupled as a stable photon pairs source. Because the coupled two single quantum emitters show the same energy structure as that of the single quantum dots, and the coupling between the single quantum emitters demonstrates the two-photon resonance peak at $\omega_{\rm L}\sin(\omega_{\rm eg}^{A}+\omega_{\rm eg}^{B})/2$, with $\omega_{\rm eg}^{A}$, and $\omega_{\rm eg}^{B}$ being the transition frequencies of the single quantum emitters A and B, respectively. At the two photon resonance peak position, the coupled quantum emitters emit two photons (more than one photon) at a time. The interaction strength between the single quantum emitters can be conveniently modified by changing the separated distance between the two quantum emitters.

In this paper, we consider the two coupled single quantum emitters as a photon pair source. The coupling of the two single quantum emitters provides a way to excite the two single quantum emitters to their excited states (the bi-exciton states) by a single external field. The external field excites the emitter A into its excited state, then the emitters A and B exchange their status, and the external field then excites the emitter A into its excited state again, which combines into a bi-exciton state. When the external field satisfies $\omega_{\rm L}\sim(\omega_{\rm eg}^{A}+\omega_{\rm eg}^{B})/2$, the external field can excite the two emitters into their excited states. On this condition, the coupled two single quantum emitters can be considered as a photon pair source. The probability of emitting two photons P_2 and the correlation coefficient R_{AB} are employed to describe the photon pair source quality. $R_{AB}\sim 1$ corresponds to the fact that the emitters A and B emit photons at the same time, while $R_{AB}\sim 0$ corresponds to the fact that the emitters A and B randomly emit photons, while Mandel's Q parameters are used to describe the self correlations of the photons emitted from the molecule A, molecule B, and the coupling system.

The rest of this paper is organized as follows. In Section 2 the model of the coupled two single quantum emitters is introduced, and the generating function approach is briefly reviewed. In Section 3, we demonstrate the main numerical results, and a short discussion is also presented. Some conclusions are drawn in Section 4.

2. Theory

We consider two separated single quantum emitters A and B, which are coupled through the dipole–dipole interaction. The emitters A and B can be described as a two-level system. The energy states of the emitters A and B are denoted by $|g_A\rangle$ and $|g_B\rangle$ (ground states), respectively, $|e_A\rangle$ and $|e_B\rangle$ (excited states) respectively. The transition frequencies of emitters A and B are $\omega_{\rm eg}^{A}$ and $\omega_{\rm eg}^{B}$, respectively. The coupled emitters can be described in the direct product space of the eigenstates space of emitters A and B. Then the Hamiltonian can be written as[11,12]

$$\mathcal{H}=(\mathcal{H}_0^A+\mathcal{H}_{\rm L}^A)\otimes\mathcal{I}_B+\mathcal{I}_A\otimes(\mathcal{H}_0^B+\mathcal{H}_{\rm L}^B)+\mathcal{H}_{\rm dd}, \qquad (1)$$

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where $\mathcal{H}_0^A$ and $\mathcal{H}_0^B$ are the "bare" Hamiltonians of emitters A and B, $\mathcal{H}_{\rm L}^A$ and $\mathcal{H}_{\rm L}^B$ are the Hamiltonians of emitters A and B interacting with an external light field, $\mathcal{I}_A$ and $\mathcal{I}_B$ are the unity operators in the Hilbert space of emitters A and B, and $\mathcal{H}_{dd}$ is the Hamiltonian of the dipole–dipole interaction between the emitters.

The "bare" Hamiltonians, in the rotating wave approximation (RWA), can be written as

$$\mathcal{H}_0^n = -\frac{\Delta_n}{2}|e_n\rangle\langle e_n| + \frac{\Delta_n}{2}|g_n\rangle\langle g_n|, (n = A, B), \tag{2}$$

where $\Delta_n = \omega_{\rm L} - \omega_{\rm eg}^n$ are the detuning frequencies, and $\omega_{\rm L}$ is the angular frequency of the external field.

The Hamiltonians of the interaction between emitters and external field in the RWA can be written as

$$\mathcal{H}_{\rm L}^n = \hbar\Omega_n(\boldsymbol{r}_n)(|e_n\rangle\langle g_n| + \text{h.c.}), \tag{3}$$

where h.c. represents the Hermitian conjugate, $\Omega_n(\boldsymbol{r}_n) = -\boldsymbol{\mu}_n \cdot \mathcal{E}_0(\boldsymbol{r}_n)/(2\hbar)$ are the Rabi frequencies, $\boldsymbol{r}_n$ is the position of emitter n, $\boldsymbol{\mu}_n$ is the transition dipole moments of the emitter n $(n = A, B)$, and $\mathcal{E}_0(\boldsymbol{r}_n)$ is the amplitude of the external field at the position $\boldsymbol{r}_n$.

The coupling Hamiltonian of the two emitters, the dipole–dipole interaction, can be written as[11,13,14]

$$\mathcal{H}_{dd} = \hbar\mathcal{V}(|e_A g_B\rangle\langle g_A e_B| + \text{h.c.}), \tag{4}$$

with

$$\begin{aligned}\mathcal{V} = \frac{3\sqrt{\Gamma_A\Gamma_B}}{2}\Bigg\{ &-[(\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{\mu}}_B) - (\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{r}}_{AB})(\hat{\boldsymbol{\mu}}_B\cdot\hat{\boldsymbol{r}}_{AB})]\frac{\cos(z)}{z} \\ &+[(\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{\mu}}_B) - 3(\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{r}}_{AB})(\hat{\boldsymbol{\mu}}_B\cdot\hat{\boldsymbol{r}}_{AB})] \\ &\times\left[\frac{\sin(z)}{z^2} + \frac{\cos(z)}{z^3}\right]\Bigg\},\end{aligned} \tag{5}$$

where $\hat{\boldsymbol{\mu}}_A$ and $\hat{\boldsymbol{\mu}}_B$ are the unit vectors of the dipole moments of molecules A and B, and $\hat{\boldsymbol{r}}_{AB}$ is the unit vector of the displacement of emitters A and B. The $z = k_0 r_{AB}$, $k_0 = (\omega_{\rm eg}^A + \omega_{\rm eg}^B)/(2c)$, $r_{AB} = |\boldsymbol{r}_{AB}| \equiv |\boldsymbol{r}_A - \boldsymbol{r}_B|$ is the distance between the emitters A and B, and c is the speed of light in a vacuum.

The evolution of the system can be described by the Liouville-von Neumann equation[15,16]

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho(t) = -\frac{\mathrm{i}}{\hbar}[\mathcal{H}, \rho(t)] + \mathcal{L}\rho(t), \tag{6}$$

where $\mathcal{L}\rho(t)$ describes the disspative part of evolution, which can be expressed as[13]

$$\begin{aligned}\mathcal{L}\rho(t) = &-\frac{1}{2}\Gamma_A(|e_A\rangle\langle e_A|\rho + \rho|e_A\rangle\langle e_A| \\ &-2|g_A\rangle\langle e_A|\rho|e_A\rangle\langle g_A) \\ &-\frac{1}{2}\Gamma_{AB}(|e_A g_B\rangle\langle g_A e_B|\rho + \rho|e_A g_B\rangle\langle g_A e_B| \\ &-2|g_B\rangle\langle e_B|\rho|e_A\rangle\langle g_A|) \\ &-\frac{1}{2}\Gamma_{AB}(|g_A e_B\rangle\langle e_A g_B|\rho + \rho|g_A e_B\rangle\langle e_A g_B| \\ &-2|g_A\rangle\langle e_A|\rho|e_B\rangle\langle g_B|) \\ &-\frac{1}{2}\Gamma_B(|e_B\rangle\langle e_B|\rho + \rho|e_B\rangle\langle e_B| \\ &-2|g_B\rangle\langle e_B|\rho|e_B\rangle\langle g_B|),\end{aligned} \tag{7}$$

where Γ_A and Γ_B are the spontaneous emission rates of emitters A and B, Γ_{AB} is the cross damping rate, which can be written as[11,13,14]

$$\begin{aligned}\Gamma_{AB} = \frac{3}{2}\sqrt{\Gamma_A\Gamma_B}\Bigg\{ &[(\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{\mu}}_B) - (\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{r}}_{AB})(\hat{\boldsymbol{\mu}}_B\cdot\hat{\boldsymbol{r}}_{AB})]\frac{\sin(z)}{z} \\ &+[(\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{\mu}}_B) - 3(\hat{\boldsymbol{\mu}}_A\cdot\hat{\boldsymbol{r}}_{AB})(\hat{\boldsymbol{\mu}}_B\cdot\hat{\boldsymbol{r}}_{AB})] \\ &\times\left[\frac{\cos(z)}{z^2} - \frac{\sin(z)}{z^3}\right]\Bigg\}.\end{aligned} \tag{8}$$

The spontaneous emission rates Γ_n, $(n = A, B)$ read

$$\Gamma_n = \frac{(\omega_{\rm eg}^n)^3\boldsymbol{\mu}_n^2}{6\pi c^3\varepsilon_0\hbar}, \quad (n = A, B). \tag{9}$$

The generating function satisfies

$$\frac{\partial}{\partial t}\mathcal{G} = -\frac{\mathrm{i}}{\hbar}[\mathcal{H}, \mathcal{G}] + \mathcal{L}^r\mathcal{G} + \mathcal{L}^{+\Gamma_A}s_A\mathcal{G} + \mathcal{L}^{+\Gamma_B}s_B\mathcal{G}, \tag{10}$$

where $\mathcal{G} = \sum_n^\infty \rho^{(k+l)}s_A^k s_B^l$, with $\rho^{(k+l)}$ being the density matrix part corresponding to the case that the emitter A has emitted k photons and emitter B has emitted l photons; $\mathcal{L}^{+\Gamma_A}$ and $\mathcal{L}^{+\Gamma_B}$ are the spontaneous emission operators;[17] s_A and s_B are the auxiliary parameters for photon counting statistics.

The working generating function is defined as

$$\mathcal{Y}(s_A, s_B, t) = \sum_m \langle m|\mathcal{G}(s_A, s_B, t)|m\rangle, \tag{11}$$

where $|m\rangle = |g_A g_B\rangle, |g_A e_B\rangle, |e_A g_B\rangle$, and $|e_A e_B\rangle$.

The moments of emission photons from emitters A and B can be written as

$$\begin{aligned}\langle N_n^{(r)}\rangle &\equiv \langle N_n(N_n - 1)\cdots(N_n - r + 1)\rangle \\ &= \frac{\partial^r}{\partial s_n^r}\mathcal{Y}(s_A, s_B, t)\Big|_{s_A = s_B = 1}, \quad (n = A, B),\end{aligned} \tag{12}$$

the corresponding Mandel's Q parameters can be expressed as

$$Q_n = \frac{\langle N_n^{(2)}\rangle - \langle N_n^{(1)}\rangle^2}{\langle N_n^{(1)}\rangle}. \tag{13}$$

The moments of the total emission photons can also be expressed as

$$\begin{aligned}\langle N_{\rm tot}^{(r)}\rangle &\equiv \langle N_{\rm tot}(N_{\rm tot} - 1)\cdots(N_{\rm tot} - r + 1)\rangle \\ &= \frac{\partial^r}{\partial s^r}\mathcal{Y}(s_A = s_B = s, t)\Big|_{s=1},\end{aligned} \tag{14}$$

and the corresponding Mandel's Q parameter $Q_{\rm tot}$ can be written as

$$Q_{\rm tot} = \frac{\langle N_{\rm tot}^{(2)}\rangle - \langle N_{\rm tot}^{(1)}\rangle^2}{\langle N_{\rm tot}^{(1)}\rangle}. \tag{15}$$

The correlation of the photons, emission from emitters A and B, is[18]

$$\langle N_A N_B\rangle = \frac{\partial^2}{\partial s_A \partial s_B}\mathcal{Y}(s_A, s_B, t)\Big|_{s_A=s_B=1}, \tag{16}$$

and the correlation coefficent can be written as[19,20]

$$R_{AB} = \frac{\langle N_A N_B\rangle - \langle N_A\rangle\langle N_B\rangle}{\sqrt{(\langle N_A^2\rangle - \langle N_A\rangle^2)(\langle N_B^2\rangle - \langle N_B\rangle^2)}}, \tag{17}$$

where

$$\langle N_n^2\rangle - \langle N_n\rangle^2 = \langle N_n^{(2)}\rangle - \langle N_n^{(1)}\rangle^2 + \langle N_n^{(1)}\rangle, \tag{18}$$

with $n = A$ and B.

The probability of emitting m photons in time interval $[0,t]$ can be written as

$$P_m(t) = \frac{1}{m!}\frac{\partial}{\partial s^m}\mathcal{Y}(s_A = s_B = s, t)\Big|_{s=0}. \tag{19}$$

3. Results and discussion

In this section, we consider the single terrylene molecules in a paraterphenyl crystal.[11,12] The system has been used to demonstrate the high spatial resolution and two-photon resonance,[12] and implemented some quantum gates in the small quantum network.[11] The spontaneous emission rates of molecule A and molecule B are $\Gamma_A = \Gamma_B = 50$ MHz.[11,12] The cross damping rate $\Gamma_{AB} = 9$ MHz, and the interaction between molecules A and B is $\mathcal{V} = 950$ MHz, and the difference transition frequency between the molecules is $\Delta\omega = \omega_{\rm eg}^A - \omega_{\rm eg}^B = 2320$ MHz.[12]

The probabilities of emitting one, two, and more photons at $\Delta_A = \Delta\omega/2$ are shown in Fig. 4, while the central peak ($\Delta_A = \Delta\omega/2$) is known as the two-photon resonance peak. Figures 1(a)–1(c) correspond to the Rabi frequencies $\Omega_A = \Omega_B =$ 50, 100, and 150 MHz, respectively. The solid lines correspond to the probabilities of emitting one photon [$P_1(t)$], the dashed lines correspond to the probabilities of emitting two photons [$P_2(t)$], and the dash-dotted lines correspond to the probabilities of emitting multi- photons [$P_{\rm multi}(t)$]. From the figures, one could know that the maximum value of the probability of emitting two photons is two times greater than that of the probability of emitting one photon, for all the driving field strengths.

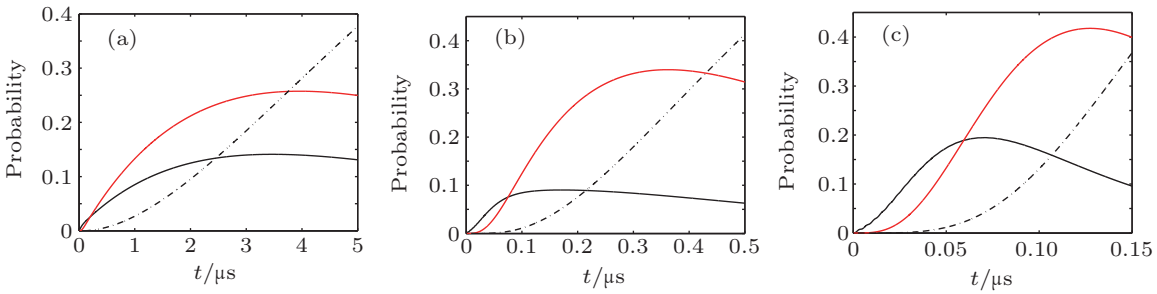


Fig. 1. (color online) Probabilities of emitting one, two, and more photons versus evolution time t, with the detuning frequency $\Delta_A = \Delta\omega/2$. Panels (a), (b), and (c) correspond to the Rabi frequencies $\Omega_A = \Omega_B = 50$, 100, and 150 MHz, respectively. The other parameters are the same as those in Fig. 1. The solid lines correspond to the probabilities of emitting one photon $P_1(t)$, dashed lines correspond to the probabilities of emitting two photons $P_2(t)$, and the dash-dotted lines correspond to the probabilities of emitting multi- photon $P_{\rm multi}(t) = 1 - P_0(t) - P_1(t) - P_2(t)$.

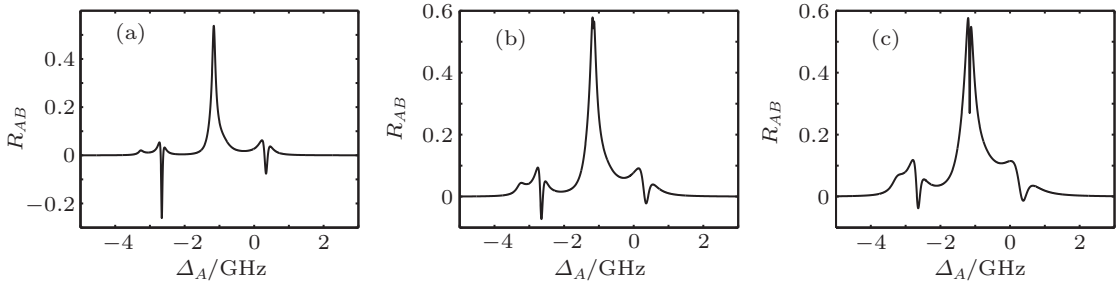


Fig. 2. The correlation coefficent R_{AB} versus the detuning frequency Δ_A. (a), (b), and (c) correspond to the Rabi frequencies $\Omega_A = \Omega_B = 50$, 100, and 150 MHz, respectively. The other parameters are the same as those in Fig. 3.

The values of correlation coefficent R_{AB} of the photons emitted from molecule A and molecule B are shown in Fig. 2. Figures 2(a)–2(c) correspond to the weak, medium, and strong driving fields, respectively. The correlation coefficent R_{AB} shows a similar behavior to Mandel's Q parameter $Q_{\rm tot}$. At the left and right resonance peak positions, the correlation coefficient R_{AB} is negative, the negative correlation, of the photons emitted from molecules A and B, means the molecules A and B cannot emit photons at the same time. On the weak driving field condition, the photons emitted from molecule A and molecule B show positive correlation at the peak position ($\Delta_A = \Delta\omega/2$), which means that molecule A emits a photon

at time interval $[t, t+\mathrm{d}t]$, while molecule B has a big probability to emit a photon at the same time interval $[t, t+\mathrm{d}t]$. The photons emitted from molecule A and molecule B are bunching together, which makes Mandel's Q parameter greater than zero. The correlation coefficient R_{AB} first increases and then decreases with the driving field strength inceasing, which means that the strong driving field destroys the correlation of the photons emitted from molecules A and B.

The emission photons $\langle N\rangle$ and Mandel's Q parameters versus the detuning frequency $\Delta_A = \omega_{\mathrm{L}} - \omega_{\mathrm{eg}}^{A}$ are plotted in Fig. 1. The first column [Figs. 3(a) and 3(d)] corresponds to the Rabi frequencies $\Omega_A = \Omega_B = 50$ MHz, the second column [Figs. 3(b) and 3(e)] corresponds to the Rabi frequencies $\Omega_A = \Omega_B = 100$ MHz, and the last column [Figs. 3(c) and 3(f)] corresponds to the Rabi frequencies $\Omega_A = \Omega_B = 150$ MHz. The red solid lines correspond to the total photons and Mandel's Q parameters of molecules A and B, the dashed-dotted lines correspond to the photons and Mandel's Q parameters of molecule A, and the dashed lines correspond to the photons and Mandel's Q parameters of molecule B.

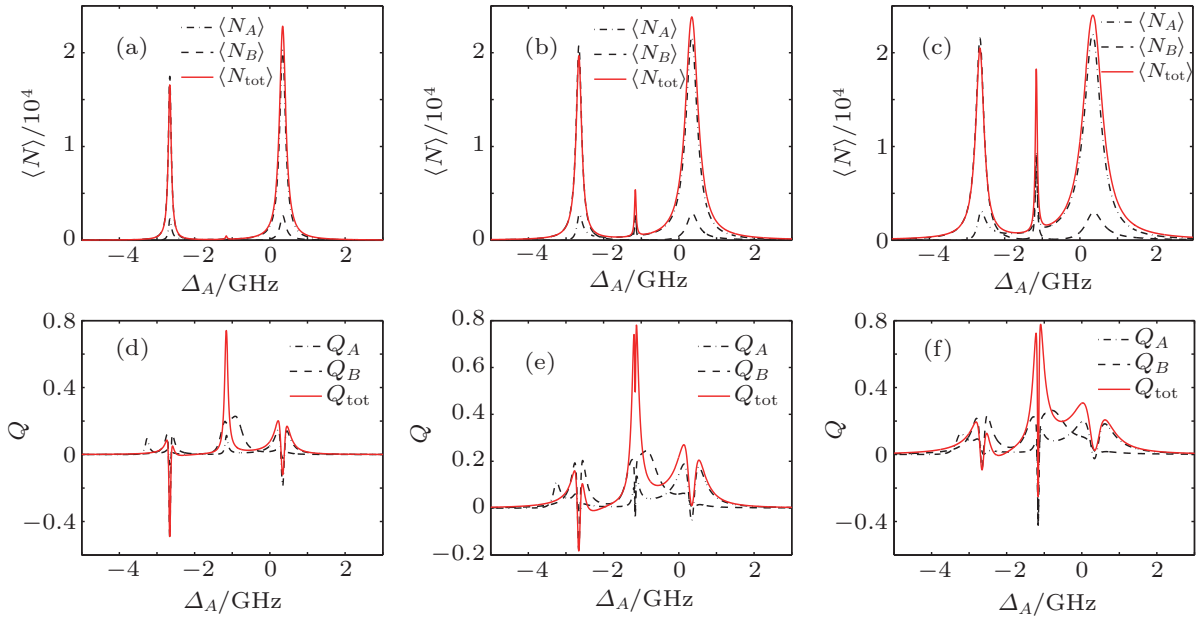


Fig. 3. (color online) The emission photons number $\langle N\rangle$ versus the detuning frequency Δ_A for different Mandel's parameters, with spontaneous emission rates $\Gamma_A = \Gamma_B = 50$ MHz, cross damping rate $\Gamma_{AB} = 9$ MHz, and the interaction $\mathcal{V} = 950$ MHz. Panels (a) and (d) correspond to the Rabi frequencies $\Omega_A = \Omega_B = 50$ MHz, panels (b) and (e) correspond to the Rabi frequencies $\Omega_A = \Omega_B = 100$ MHz, and panels (c) and (f) correspond to the Rabi frequencies $\Omega_A = \Omega_B = 150$ MHz. The red lines correspond to the total emission photons, dashed–dotted lines correspond to the photons emitted from molecule A, and the dashed lines correspond to the photons emitted from molecule B.

From Fig. 3, one can know that the total emission photon number $\langle N_{\mathrm{tot}}\rangle$ emitted from molecules A and B is a little smaller than the photon number $\langle N_B\rangle$ emitted from molecule B at the left peak, and a little greater than the photon number $\langle N_A\rangle$ emitted from molecule A at the right peak, for all the driving field strengths $\Omega_A = \Omega_B = 50$ MHz (weak condition), 100 MHz (medium condition), and 150 MHz (strong condition). The differences between the total emission photons $\langle N_{\mathrm{tot}}\rangle$ and the emission photons $\langle N_A\rangle$ and $\langle N_B\rangle$ are caused by the coherence of the states $|e_A g_B\rangle$ and $|g_A e_B\rangle$, and the virtual photons emission and absorption between the states.[21] The dipole–dipole interaction $\mathcal{V}$ causes the resonance peak to shift.[13] The Mandel's Q parameters Q_{tot}, Q_A, and Q_B are negative at the positions of left and right resonance peaks for all driving field strength conditions. That means that the photons emitted from molecule A, from molecule B, and from molecules A and B are anti-bunching at the resonance peak positions. On the weak driving condition, the emission photon number $\langle N_{\mathrm{tot}}\rangle$ is very small, and the corresponding Mandel's Q parameter Q_{tot} is positive at the position $\Delta_A = \Delta\omega/2$ (central peak), which corresponds to the emission photons showing bunching behavior, the result is the same as that in Ref. [12]. The emission photon number $\langle N_{\mathrm{tot}}\rangle$ increases, and the corresponding Mandel's Q parameter Q_{tot} first increases and then decreases with the driving field strength increasing. On the strong driving field condition, Mandel's Q parameter Q_{tot} decreases to a negative value, corresponding to the fact that the emission photons show anti-bunching behaviors.

4. Conclusions

In this paper, we consider a system with two coupling two-level single molecules as a photon pair source. The probabilities of emitting one, two-, and multi- photons: $P_1(t), P_2(t)$, and $P_{\mathrm{multi}}(t)$, are respectively calculated at $\Delta_A = \Delta\omega/2$ via generating function approach. In a short time region, the probability of emitting two photons is three times greater than the

probabilities of emission one and more photons. That means that in a short time, the system is a good two-photon source. In a long time region, the correlation coefficient R_{AB} is introduced to describe the coincidence of the emission photons, and on the weak driving field condition, the correlation coefficient and the Mandel's Q parameter $Q_{\rm tot}$ show that the emitted photons are bunching. The system could emit the photon pair train driven by a weak field. As the driving field increases, the correlation between the emission photons is destroyed by the driving field.

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