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Photon emission from a single molecule in a solid matrix: Phase-delay control

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In this paper, we study the influence of time-dependent phase delay on the emission of few photons from a single molecule in a solid matrix. The results demonstrate that a time-dependent phase of the pulses can be used to control the photon emission and strongly affects the spectrum of single molecules. The spectrum of the photon emission probabilities reflects the fast dynamics of the states of the system when the duration of the driven laser pulses is short. Also, it is shown that the coupling between the system and the rf field strongly affects the spectrum of the photon emission probabilities in space.

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I. INTRODUCTION

Single quantum systems (single atoms, single ions, single molecules, and single quantum dots) have been widely investigated in recent years [1–20]. Not only can the single quantum systems be considered as effective quantum bits in quantum computing, but also the photon emission from the single quantum systems can be considered as effective quantum bits in quantum communication. The photon has relatively longer coherent time and its transition speed is fast and it is less affected by its surrounding environment molecules [5–7]. The deterministic single and pair photon source is important to the quantum computing and quantum communication. One can modify the strength of the external laser pulses to control the photon emission from single quantum systems [4]. Or, one can modify the delay phase between the sequence pulses to control the photon emission from single quantum systems. Generally, the surrounding environments of the single molecule could break the symmetry of the electron cloud of the single molecule states, which makes the single molecule present permanent dipole moments. The external radio frequency (rf) field couples with the permanent dipole moments of a single molecule; the electron could oscillate following the external rf field according to the laws of electrodynamics. This means that one can modify the rf field or the environment (which could change the permanent dipole moments of the single molecule states) to control photon emission from single molecules [21–25].

The single dibenzantanthrene (DBATT) molecule in a matrix is widely studied [21–28]. This single molecule in the matrix can be described by a two-level system: a ground state $|g\rangle$ and an excited state $|e\rangle$, since its transition rate to the triplet state can be omitted at low temperature. The transition dipole moment between the states $|g\rangle$ and $|e\rangle$ is $\boldsymbol{\mu}_{ge}$, the permanent dipole moments of the states $|g\rangle$ and $|e\rangle$ are $\boldsymbol{\mu}_{gg}$ and $\boldsymbol{\mu}_{ee}$, respectively. The laser pulse couples with the transition dipole moment and the rf field couples with the permanent dipole moments. The DBATT molecule is usually fixed in the matrix, but sometimes the single molecule diffuses in the matrix in the laser spot, then the different positions of the single molecule in the laser spot may present different emission effects, such as the photon emission frequency, emission intensity, etc.

In this paper, we investigate the control of few photons emitted from the single DBATT molecule in a solid matrix via the delay time-dependent phase of driven external fields. The single DBATT molecule in a solid matrix system is driven by external laser pulses and the rf field. If the external fields have a time-dependent delay phase (or a time-dependent initial phase) between the sequence pulses, we show that there is a new coupling "path" between the coherence and populations. This new coupling path has an influence on photon emission, and can be used to control the photon emission from the single molecule system.

The generating function approach [23–25,29–40] is employed in this paper. The generating function approach has been used to study the quantum beats, coherent population trapping of the single-V-type three-level system, and the electromagnetically induced transparency (EIT) in the Λ three-level system, etc. The statistical properties of x and y polarized photons and their correlation have also been studied via the generating function approach [35–40].

This paper is organized as follows: the generating function approach is briefly reviewed in Sec. II. The results and discussion are presented in Sec. III. The conclusion is given in Sec. IV.

II. THEORY

The Hamiltonian of the single molecule in the solid matrix driven by external fields can be written as

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2, \tag{1}$$

where $\mathcal{H}_0$ is the Hamiltonian of the "bare" single molecule, $\mathcal{H}_1$ is the interaction between the single molecule and the external laser pulses, and $\mathcal{H}_2$ is the interaction between the single molecule and the rf field.

The Hamiltonian of the bare single molecule can be expressed as

$$\mathcal{H}_0 = \hbar\omega_g|g\rangle\langle g| + \hbar\omega_e|e\rangle\langle e|, \tag{2}$$

where $|g\rangle$ and $|e\rangle$ are the ground state and the excited state of the single molecule, and ω_g and ω_e are the eigenfrequencies corresponding to the states of $|g\rangle$ and $|e\rangle$, respectively.

The interaction between the single molecule and the external laser field, in the dipole approximation, can be expressed as

$$\mathcal{H}_1 = -\boldsymbol{\mu}_{ge}\cdot\boldsymbol{E}(x,y;t)(|e\rangle\langle g| + |g\rangle\langle e|), \tag{3}$$

where $\boldsymbol{\mu}_{ge}$ is the transition dipole moment between the states of $|e\rangle$ and $|g\rangle$; $\boldsymbol{E}(x,y;t)$ is the external laser field, which is chosen as the classical electromagnetic field [41]. In this paper the external laser field is taken as the Gaussian distribution in spaces, and is taken as [26]

$$\boldsymbol{E}(x,y;t) = \boldsymbol{E}_1(x,y;t) + \boldsymbol{E}_2(x,y;t), \tag{4}$$

and

$$\begin{aligned} \boldsymbol{E}_1(x,y;t) &= \boldsymbol{\mathcal{E}}_1(x,y;t)\cos(\omega_L t), \\ \boldsymbol{E}_2(x,y;t) &= \boldsymbol{\mathcal{E}}_2(x,y;t)\cos[\omega_L t + \varphi(t)], \end{aligned} \tag{5}$$

where

$$\boldsymbol{\mathcal{E}}_\kappa(x,y;t) = \boldsymbol{\mathcal{E}}_0 e^{-[(x-x_0)^2+(y-y_0)^2]/2\sigma^2} e^{-(t-t_0)^2/\tau_\kappa^2} \ (\kappa = 1,2). \tag{6}$$

where $\boldsymbol{\mathcal{E}}_0$ is the electric amplitude of the laser pulses, and t_0 is the pulse center. σ is the spatial half width of the laser spot, x and y are the positions of the single molecule in the laser spot, x_0 and y_0 are the spatial center of the pulse, $\varphi(t)$ is the time-dependent phase of the laser field [or the phase difference between the two laser pulses as shown in Eq. (5)], ω_L is the angular frequency of laser field, and τ_1 and τ_2 are the time half width of the first and second laser pulse, respectively.

The interaction between the single molecule and the rf field can be expressed as

$$\mathcal{H}_2 = -\boldsymbol{\mu}_{gg}\cdot\boldsymbol{E}_{rf}(t)|g\rangle\langle g| - \boldsymbol{\mu}_{ee}\cdot\boldsymbol{E}_{rf}(t)|e\rangle\langle e|, \tag{7}$$

where $\boldsymbol{\mu}_{gg}$ and $\boldsymbol{\mu}_{ee}$ are the permanent dipole moments of the states $|g\rangle$ and $|e\rangle$, respectively, and $\boldsymbol{E}_{rf}(t)$ is the amplitude of the external rf field. The rf field is taken as follows:

$$\boldsymbol{E}_{rf}(t) = \boldsymbol{\mathcal{E}}_{rf}\cos(\omega_{rf}t), \tag{8}$$

where ω_{rf} and $\boldsymbol{\mathcal{E}}_{rf}$ are the angular frequency and electric amplitude of the rf field, respectively.

The time evolution of the single molecule in the matrix can be described by the Liouville–von Neumann equation [42,43],

$$\frac{\partial}{\partial t}\rho(t) = -\frac{i}{\hbar}[\mathcal{H},\rho(t)] + \mathcal{L}\rho(t), \tag{9}$$

where $\rho(t)$ is the density matrix of the single molecule, and $\mathcal{L}$ is the superoperator including the spontaneous emission rates which are caused by the vacuum field.

The generating function approach, as a useful method to extract information of photon emission, is defined as [29–33]

$$\mathcal{G}_{ij}(s,t) = \sum_n \rho_{ij}^{(n)} s^n, \tag{10}$$

where $\rho_{ij}^{(n)}$ $(i,j = e,g)$ is the portion of the reduced density matrix stemming from a system with a prior history of n photon emission events. s is an auxiliary parameter; it can be used to count the photon emission events in the time interval $[0,t]$.

We define

$$\Omega_\kappa(x,y;t) = -\frac{1}{\hbar}\boldsymbol{\mu}_{ge}\cdot\boldsymbol{\mathcal{E}}_\kappa(x,y;t), \tag{11}$$

and

$$\omega_0 = \omega_{eg} + \zeta\omega_{rf}\cos(\omega_{rf}t), \tag{12}$$

where $\omega_{eg} = \omega_e - \omega_g$ is the transition frequency, and $\zeta = (\boldsymbol{\mu}_{ee} - \boldsymbol{\mu}_{gg})\cdot\boldsymbol{\mathcal{E}}_{rf}/\hbar\omega_{rf}$ is the modulation index of the rf field. We assume $x_0 = y_0 = 0$ in the following numerical calculations.

In the following discussion, the laser pulse area at (x,y) is defined as

$$\Theta_\kappa(x,y) = \int_{-\infty}^{\infty}\Omega_\kappa(x,y;\tau)d\tau \quad (\kappa = 1,2). \tag{13}$$

Introducing the Bloch vectors

$$\begin{aligned} \mathcal{U} &= \mathrm{Re}\mathcal{G}_{ge}e^{-i\omega_L t}, \\ \mathcal{V} &= \mathrm{Im}\mathcal{G}_{ge}e^{-i\omega_L t}, \\ \mathcal{W} &= (\mathcal{G}_{ee} - \mathcal{G}_{gg})/2, \\ \mathcal{Y} &= (\mathcal{G}_{ee} + \mathcal{G}_{gg})/2, \end{aligned} \tag{14}$$

the generating function can, under the rotating wave approximation (RWA), be written as

$$\begin{aligned} \dot{\mathcal{U}} &= -\frac{\Gamma}{2}\mathcal{U} + \delta(t)\mathcal{V} - \Omega^{(i)}\mathcal{W}, \\ \dot{\mathcal{V}} &= -\delta(t)\mathcal{U} - \frac{\Gamma}{2}\mathcal{V} - \Omega^{(r)}\mathcal{W}, \\ \dot{\mathcal{W}} &= \Omega^{(r)}\mathcal{V} - \Omega^{(i)}\mathcal{U} - \frac{\Gamma}{2}(1+s)\mathcal{W} - \frac{\Gamma}{2}(1+s)\mathcal{Y}, \\ \dot{\mathcal{Y}} &= -\frac{\Gamma}{2}(1-s)\mathcal{W} - \frac{\Gamma}{2}(1-s)\mathcal{Y}, \end{aligned} \tag{15}$$

where $\delta(t) = \Delta - \zeta\omega_{rf}\cos(\omega_{rf}t)$, $\Delta = \omega_L - \omega_{eg}$ is the detuning frequency of the laser field, and

$$\Omega^{(r)} = \Omega_1 + \Omega_2\cos[\varphi(t)], \quad \Omega^{(i)} = \Omega_2\sin[\varphi(t)]. \tag{16}$$

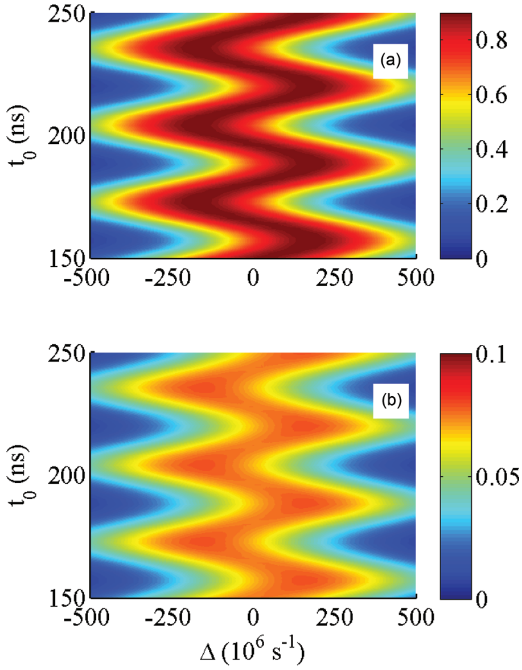

FIG. 1. (Color online) The probabilities of the emission of one (a) and two (b) photons as a function of the detuning frequency $\Delta = \omega_L - \omega_{eg}$ and t_0. The parameters are $\zeta = 1$, $\omega_{rf} = 2\times10^8$ s^{-1}, $\Gamma/2\pi = 1.7\times10^7$ s^{-1}, the pulse area $\Theta_1 = 0$ and $\Theta_2 = \pi$, and $\omega_c = 0$, $x = y = 0$, $\sigma = 120$ nm, $\varphi_0 = 0$, and $\delta = 0$. The calculation time $t = 400$ ns.

We can see that the Rabi frequency $\Omega^{(i)}$ comes from the phase difference $\varphi(t)$ between the two laser pulses. Clearly, if $\Omega^{(i)} = 0$, Eq. (15) will back up our previous results [25]. Also, Eq. (15) shows that there is a new coupling path between the coherence and populations (that is, the terms $\mathcal{U}$ and $\mathcal{W}$), and this new coupling path has an influence on photon emission. We can control the photon emission from the single molecule system using this new path.

We can extract information on the statistical properties of the photon emission, such as n-photon probability,

$$p_n(t) = \frac{2}{n!}\frac{\partial^n}{\partial s^n}\mathcal{Y}(s,t)\bigg|_{s=0}, \tag{17}$$

and the average photon emission numbers,

$$\langle N\rangle(t) = 2\frac{\partial}{\partial s}\mathcal{Y}(s,t)\bigg|_{s=1}, \tag{18}$$

etc.

III. RESULTS AND DISCUSSION

We consider the case of DBATT embedded in a n-tetradecane Shpol'skiĭ matrix. The fluorescence lifetime of the first excited state of DBATT in the matrix is $T_1 = 9.5$ ns [26,44], and it corresponds to the spontaneous emission rate $\Gamma/2\pi = 1.7 \times 10^7\ \mathrm{s}^{-1}$.

A. Single pulse

In this subsection, we consider the external laser field with a time-dependent initial phase. The external laser field is taken as $\boldsymbol{E}(x,y,t) = \boldsymbol{E}_2(x,y,t)$ (i.e., $\Omega_1 = 0$), and the time-dependent phase is

$$\varphi(t) = \int_0^t a(\tau)d\tau + \varphi_0, \tag{19}$$

where φ_0 is a constant. The detuning frequency is $\delta = \Delta + a(t)$ with $\Delta = \omega_L - \omega_{eg}$.

In this paper, we consider the case of the linear chirped laser field, namely, $a(t) = \omega_c$ is taken as a constant. This similar linear chirped laser field is easily obtained experimentally. In this case, the time-dependent phase $\varphi(t) = \omega_c t + \varphi_0$ is a linear function with time t. In this case, the resonance condition is $\delta = \Delta + \omega_c = 0$. This means that the spectrum of the single molecule system has a frequency shift, and the shift frequency is $\omega_{shift} = \omega_c$ [27].

We consider the case of the laser pulse duration $\tau_2 \ll 2\pi/\omega_{rf}$. This means that the pulse duration is much less than the rf field periodic time; the photon emission number (by taking the average time over the pulse duration) cannot cover the "fluctuation" of the transition frequency which is modified by the rf field. In other words, the laser pulse center t_0 is the time that one uses to detect the dynamics of the energy levels of

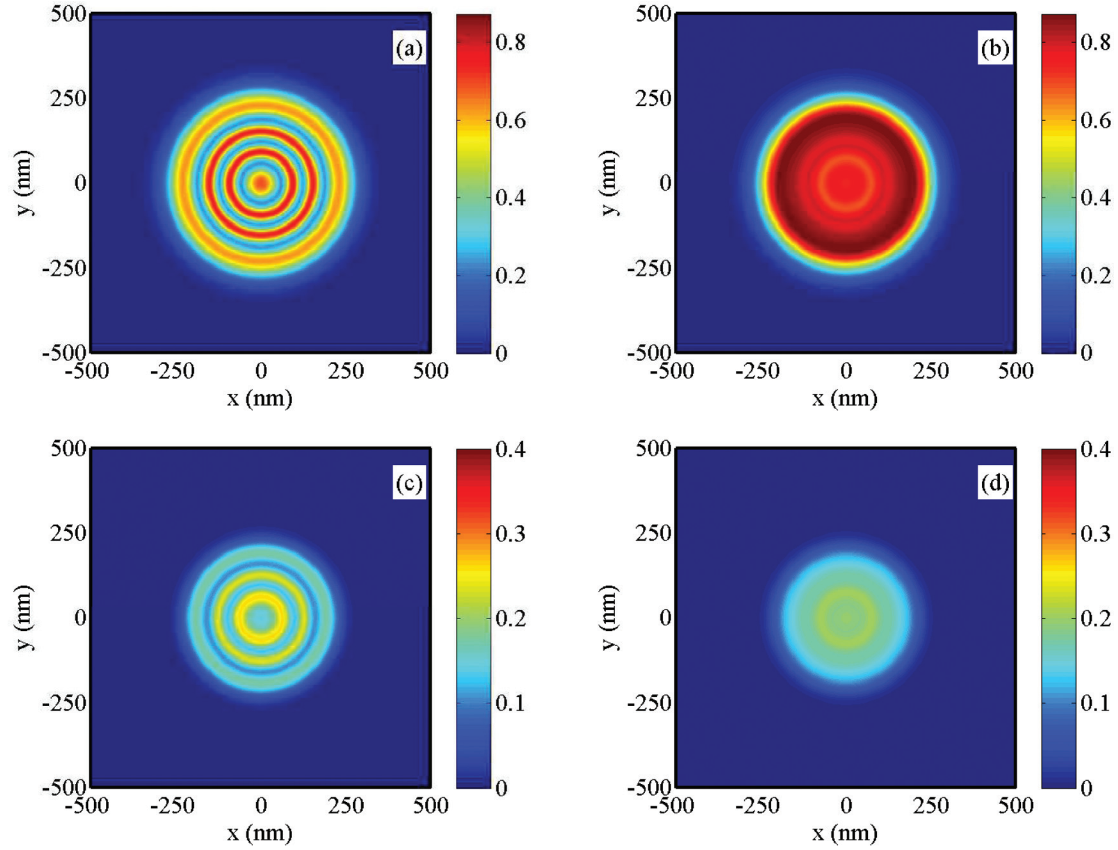

FIG. 2. (Color online) The probabilities of the emission of one [panels (a) and (b)] and two [panels (c) and (d)] photons as a function of the spatial coordinates x and y. The parameters in our calculation are $\tau_2 = 4$ ns, $\Omega = 3 \times 10^9\ \mathrm{s}^{-1}$, $\Gamma/2\pi = 1.7 \times 10^7\ \mathrm{s}^{-1}$, $\omega_c = 0$, $\Theta_1 = 0$, $\zeta = 2$, and $\omega_{rf} = 2 \times 10^8\ \mathrm{s}^{-1}$ [for (a) and (c)], $\zeta = 2$, $\omega_{rf} = 4 \times 10^8\ \mathrm{s}^{-1}$ [for (b) and (d)], $\varphi_0 = 0$, $t_0 = 200$ ns, $\Delta = 0$, and the calculation time $t = 400$ ns.

the system. The influence of the laser center t_0 and the detuning frequency $\Delta = \omega_L - \omega_{eg}$ on the emission probabilities is demonstrated in Fig. 1 for the pulse duration $\tau_2 = 4.0$ ns. As shown in the figure, the excited and ground states of the single molecule oscillate following the rf field. However, the amplitudes of the excited state and the ground state are different; the transition frequency of the single molecule becomes $\omega'_{eg} = \omega_{eg} + (\boldsymbol{\mu}_{ee} - \boldsymbol{\mu}_{gg}) \cdot \mathcal{E}_{rf} \cos(\omega_{rf} t) = \omega_0$, where ω_0 is the same as that in Eq. (12). When τ_2 is long enough, such as $\omega_{rf}\tau_2 \gg 2\pi$, the oscillation is eliminated by the time average. If this happens, it recovers the result of the single molecule driven by the continuous wave field [23,25].

The probabilities p_1 and p_2 as a function of the coordinates (x,y) of a single molecule in the laser spot are demonstrated in Fig. 2. In Figs. 2(a) and 2(c), p_1 and p_2 oscillate with the coordinates x and y for the case of $\omega_{rf} = 2 \times 10^8$ s^{-1}, $\zeta = 2$. When the pulse area $\Theta_2 \sim (2n+1)\pi$ $(n = 0,1,2,\ldots)$, the system gives the maximum value of p_1. When the pulse area $\Theta_2 \sim 2(n+1)\pi$ $(n = 0,1,2,\ldots)$, however, the system gives the maximum value of p_2. In Figs. 2(b) and 2(d), we show the case that p_1 and p_2 have almost no oscillation with the spatial coordinates x and y ($\omega_{rf} = 4 \times 10^8$ s^{-1} and $\zeta = 2$). Physically, this means that the rf frequency ω_{rf} can eliminate the spatial Rabi oscillation. In this aspect, one can delete the effect of the spatial positions of the single molecule by choosing a suitable rf field.

B. Two pulses

In this subsection, we consider the single molecule driven by two pulses. We choose the case of $\tau_1 = 50$ ns and $\tau_2 = 4$ ns.

As we noted in Sec. II, $\Omega^{(i)}$ shows us a new path to control the photon emission from the single molecule system; to more deeply understand the effect of $\Omega^{(i)}$ on p_1 and p_2, in Fig. 3 we show p_1 and p_2 as a function of detuning frequency Δ and pulse center t_0 for different $\Omega^{(i)}$. Figures 3(a) and 3(c) show the results of $\Omega^{(i)} = 0$ (we let $\varphi_0 = 0$).

It is shown in Figs. 3(a) and 3(c) that p_1 and p_2 are not the (co)sine functions of the driven pulse center t_0; the maximum values of p_1 and p_2 show discrete behavior. The reason for this case is that the first pulse width $\tau_1 = 50$ ns is long enough, which can be thought of as a continuous wave field.

In Figs. 3(b) and 3(d), we show the effect of the Rabi frequency $\Omega^{(i)}$ on p_1 and p_2 (when $\varphi_0 = \pi/2$, $\omega_c = 0$, and the Rabi frequencies $\Omega^{(r)} = \Omega_1$ and $\Omega^{(i)} = \Omega_2$, this gives the maximum value of $\Omega^{(i)}$). The position of the maximum values of the probability p_1 are shifted in the case $\Omega^{(i)} = \Omega_2$ compared with that in the case $\Omega^{(i)} = 0$. The maximum values of p_2 are concentrated in a small range; this contracts to the case of $\Omega^{(i)} = 0$, as shown in Figs. 3(a) and 3(c).

Figure 4 shows p_1 and p_2 as a function of the detuning frequency Δ and the phase changing rate ω_c. It demonstrates the trajectory of the maximal value of p_1 and p_2 at $\Delta = \omega_c$, which corresponds to the resonance emission condition of

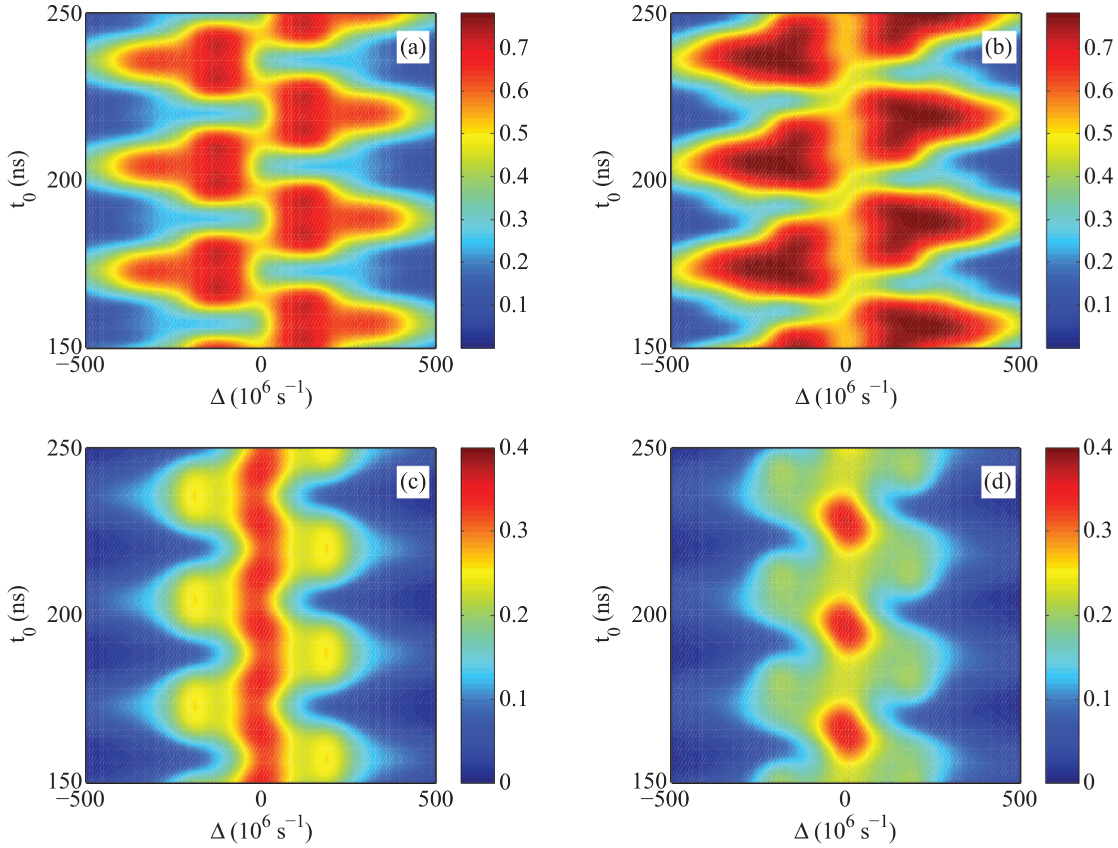

FIG. 3. (Color online) The probabilities of the emission of one [panels (a) and (b)] and two [panels (c) and (d)] photons as a function of the detuning frequency $\Delta = \omega_L - \omega_{eg}$ and t_0. The left column is for $\varphi_0 = 0$, and the right column is for $\varphi_0 = \pi/2$. The other parameters are $\zeta = 1$, $\omega_{rf} = 2 \times 10^8$ s^{-1}, $\Gamma/2\pi = 1.7 \times 10^7$ s^{-1}, the pulse area $\Theta_1 = 1.4\pi$ and $\Theta_2 = \pi$, and $\omega_c = 0$, $x = y = 0$, $\sigma = 120$ nm, and $\delta = 0$, and the calculation time $t = 400$ ns.

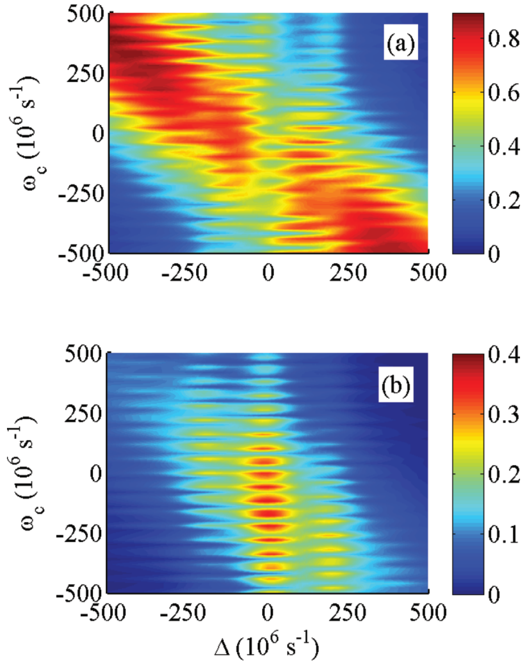

FIG. 4. (Color online) The probabilities of the emission of one (a) and two (b) photons as a function of the detuning frequency Δ and the time-dependent phase changing rate ω_c, $\varphi_0 = 0$, $t_0 = 200$ ns, and the other parameters as in Fig. 3.

the second pulse. Both p_1 and p_2 oscillate with ω_c; their period is $T_\varphi = 2n\pi$ $(n = 0, \pm1, \pm2, \ldots)$. The reason is that p_1 and p_2 depend on the driven laser strength $I \propto |\Omega|^2 = \Omega^{(r)2} + \Omega^{(i)2} = \Omega_1^2 + \Omega_2^2 + 2\Omega_1\Omega_2\cos(\omega_c t)$. In this aspect, the probabilities oscillate with ω_c. Also, the interesting thing is that for the case of near resonance excitation (detuning frequency $\Delta \sim 0$), p_1 gives a minimum value, corresponding to the Rabi oscillation in a small value (the total pulse area $\Theta = \Theta_1 + \Theta_2 \sim 2.4\pi$), and the probability p_2 reaches its maximum value. This give us a method to generate photon pairs via a single molecule system.

The effect of the rf field modulation index ζ and the pulse area Θ_2 to the probability on p_1 and p_2 $(\Theta_1 = 1.4\pi)$ are demonstrated in Fig. 5. As shown in the figure, p_1 and p_2 also oscillate with the pulse area Θ_2 and the modulation index ζ. When the strength of the pulse is strong enough, the photon emission probabilities show the well-known Rabi oscillation. And, when the driven pulse width is long enough, the photon emission probabilities oscillate with the modulation index ζ [21–23]. As the pulse area Θ_2 and the modulation index ζ increase, p_1 increases wiggly. It reaches its maximum value $p_1^{(\max)}$ at $\zeta^{(\max)}$ and $\Theta_2^{(\max)}$. The probability of p_2, however, shows decreasing behavior as the pulse area Θ_2 and the modulation index ζ increase. The maximum value of the emission of the two-photon probability $p_2^{(\max)}$ is at $\Theta_2 \sim 0.7\pi$ and $\zeta = 0$.

IV. CONCLUSIONS

In this paper, we studied the control of one- and two-photon emissions from the single DBATT molecule system in the matrix environment driven by laser pulses and continuous rf field. The results demonstrate that the time-dependent phase of the laser pulses strongly affects the probabilities of the photon emission. Our calculations show that the short pulse of the laser field can be used to detect the fast dynamics of the single molecule states. With the increase of laser pulse width, the behavior of the fast dynamics of the single molecule in the matrix is covered by the time averaging. The probabilities p_1 and p_2 oscillate with rf modulation index ζ. The probability p_1 increases the oscillation with the increase of modulation index ζ and the second laser pulse area Θ_2. This shows that the maximum value of p_1 can be found in the greatest modulation index ζ and pulse area Θ_2. The probability p_2, however, decreases the oscillation with the increase of modulation index ζ and the second pulse area Θ_2, which shows that the maximum value of the probability p_2 exists at a small modulation index ζ and pulse area Θ_2. The photon emission probabilities p_1 and p_2 oscillate with the rf field. This property presents a way to modify and control the photon emission via employing the rf field. Also, the different rf fields show the spatial Rabi oscillations.

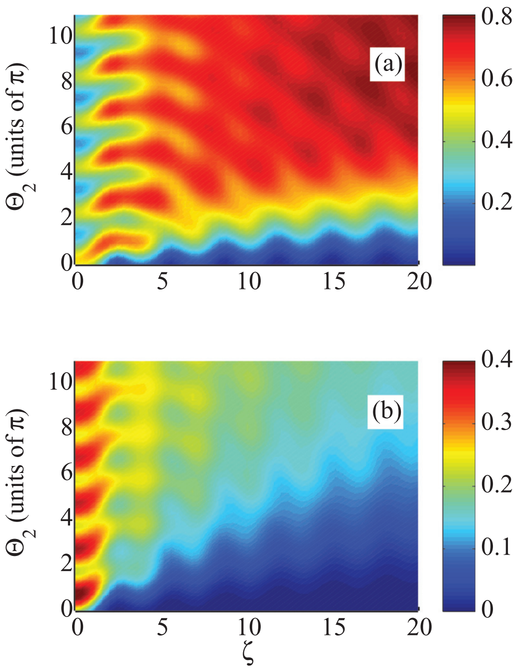

FIG. 5. (Color online) The probabilities of the emission of one (a) and two (b) photons as a function of the modulation index ζ and the pulse area Θ_2 with $\Theta_1 = 1.4\pi$. The other parameters are $x = y = 0, \sigma = 120\,\text{nm}, \omega_{rf} = 2 \times 10^8\ \text{s}^{-1}, \Delta = 0, \omega_c = 0, \varphi_0 = 0$, and $\Gamma/2\pi = 1.7 \times 10^7\ \text{s}^{-1}$, $t_0 = 200$ ns, and the calculation time $t = 400$ ns.

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[1] F. Treussart, R. Alléaume, V. LeFloc'h, L. T. Xiao, J.-M. Courty, and J.-F. Roch, Phys. Rev. Lett. **89**, 093601 (2002).
[2] B. E. Cole, J. B. Williams, B. T. King, M. S. Sherwin, and C. R. Stanley, Nature (London) **410**, 60 (2001).
[3] B. Lounis and W. E. Moerner, Nature (London) **407**, 491 (2000).
[4] A. Zrenner, E. Beham, S. Stufler, F. Findeis, M. Bichler, and G. Abstreiter, Nature (London) **418**, 612 (2002).
[5] J. Kim, O. Benson, H. Kan, and Y. Yamamoto, Nature (London) **397**, 500 (1999).
[6] C. Santori, D. Fattal, J. Vučković, G. S. Solomon, and Y. Yamamoto, Nature (London) **419**, 594 (2002).
[7] C. Santori, D. Fattal, J. Vuckovic, G. S. Solomon, and Y. Yamamoto, New J. Phys. **6**, 89 (2004).
[8] B. Darquié, M. P. A. Jones, J. Dingjan, J. Beugnon, S. Bergamini, Y. Sortais, G. Messin, A. Browaeys, and P. Grangier, Science **309**, 454 (2005).
[9] J. R. Guest, T. H. Stievater, G. Chen, E. A. Tabak, B. G. Orr, D. G. Steel, D. Gammon, and D. S. Katzer, Science **293**, 2224 (2001).
[10] N. H. Bonadeo, J. Erland, D. Gammon, D. Park, D. S. Katzer, and D. G. Steel, Science **282**, 1473 (1998).
[11] M. Bayer, O. Stern, P. Hawrylak, S. Fafard, and A. Forchel, Nature (London) **405**, 923 (2000).
[12] F. Shikerman and E. Barkai, Phys. Rev. Lett. **99**, 208302 (2007).
[13] L. P. Watkins and H. Yang, Biophys. J. **86**, 4015 (2004).
[14] X. Xu, B. Sun, P. R. Berman, D. G. Steel, A. S. Bracker, D. Gammon, and L. J. Sham, Science **317**, 929 (2007).
[15] R. Alléaume, F. Treussart, J. M. Courty, and J. F. Roch, New J. Phys. **6**, 85 (2004).
[16] R. Hildner, D. Brinks, and N. F. van Hulst, Nat. Phys. **7**, 172 (2011).
[17] J. Cao, Chem. Phys. Lett. **327**, 38 (2000).
[18] S. Yang and J. Cao, J. Phys. Chem. B **105**, 6536 (2001).
[19] S. Yang and J. Cao, J. Chem. Phys. **117**, 10996 (2002).
[20] J. Cao, J. Phys. Chem. B **110**, 19040 (2006).
[21] I. Rozhkov and E. Barkai, J. Chem. Phys. **123**, 074703 (2005).
[22] I. Rozhkov and E. Barkai, Phys. Rev. A **71**, 033810 (2005).
[23] B. Han and Y. Zheng, Phys. Rev. A **78**, 015402 (2008).
[24] B. P. Han, L.-S. Gu, Z.-W. Ji, and Y.-J. Zheng, J. At. Mol. Sci. **1**, 280 (2010).
[25] B. Han, Z. Ji, and Y. Zheng, J. Chem. Phys. **130**, 244502 (2009).
[26] I. Gerhardt, G. Wrigge, J. Hwang, G. Zumofen, and V. Sandoghdar, Phys. Rev. A **82**, 063823 (2010).
[27] I. Gerhardt, G. Wrigge, G. Zumofen, J. Hwang, A. Renn, and V. Sandoghdar, Phys. Rev. A **79**, 011402 (2009).
[28] D. E. Makarov and H. Metiu, J. Chem. Phys. **115**, 5989 (2001).
[29] F. L. H. Brown, Phys. Rev. Lett. **90**, 028302 (2003).
[30] I. V. Gopich and A. Szabo, J. Chem. Phys. **118**, 454 (2003).
[31] Y. Zheng and F. L. H. Brown, Phys. Rev. Lett. **90**, 238305 (2003).
[32] Y. Zheng and F. L. H. Brown, J. Chem. Phys. **119**, 11814 (2003).
[33] Y. Zheng and F. L. H. Brown, J. Chem. Phys. **121**, 7914 (2004).
[34] Y. Zheng and F. L. H. Brown, J. Chem. Phys. **121**, 3238 (2004).
[35] Y. Peng, Y. Zheng, and F. L. H. Brown, J. Chem. Phys. **126**, 104303 (2007).
[36] Y. Peng and Y. Zheng, Appl. Phys. Lett. **92**, 092120 (2008).
[37] Y. Peng and Y. Zheng, Phys. Rev. A **80**, 043831 (2009).
[38] Y. Peng, S. Xie, Y. Zheng, and F. L. H. Brown, J. Chem. Phys. **131**, 214107 (2009).
[39] Y. Peng, D. Wang, Y. Zheng, and S. Xie, Physica E **42**, 2242 (2010).
[40] Y. Peng and Y. Zheng, Chem. Phys. Lett. **516**, 272 (2011).
[41] As the average photon number of the laser field is greater than 1, the classical description of the electromagnetic field is a good approximation; the quantum description and the classical description would give the same results [43,45–47].
[42] S. Mukamel, *Principle of Nonlinear Optical Spectroscopy* (Oxford University Press, New York, 1995).
[43] R. Loudon, *The Quantum Theory of Light*, 3rd ed. (Oxford University Press, New York, 2000).
[44] R. Lettow, V. Ahtee, R. Pfab, A. Renn, E. Ikonen, S. Götzinger, and V. Sandoghdar, Opt. Express **15**, 15842 (2007).
[45] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications* (John Wiley and Sons, New York, 1992).
[46] M. H. Mittleman, *Introduction to the Theory of Laser-Atom Interactions*, 2nd ed. (Plenum, New York, 1993).
[47] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University Press, Cambridge, England, 1995).