Quadripartite entanglement from a double three-level Λ-type-atom model

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We present a theoretical scheme for the generation of four quantized modes in a three-level double-Λ atomic system driven by two counterpropagating far-detuned pump beams and find the entanglement between the four modes. By using the second-order perturbation method and the phase-matching condition for the four-wave mixing processes, the effective Hamiltonian is derived, which clearly illustrates the generation of four light beams and their entanglement. The dependence of the four-mode entanglement on interaction time, pump detuning, strength of interaction force, and the ratio of Rabi frequency of two pump beams is analyzed with inseparability. The result presented here provides a method for the experimental generation of multipartite entanglement in an atomic system.

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I. INTRODUCTION

Quantum entanglement is a crucial resource in quantum network for large-scale quantum information processing [1]. In a quantum information network, light or photons are naturally used as carriers of entanglement to exchange the quantum information between the separated quantum nodes in remote locations [2] where the atoms generate, process, and store quantum information locally [3]. Fundamentally, this endeavor is the quantum interface that converts quantum states from one physical system to those of another in a reversible fashion. Such quantum connectivity can be achieved by optical interaction of photons and atoms [4]. Therefore, the quantum entanglement of light with the wavelength matching with the considered atomic transitions is necessarily required.

Up to now, many technologies, such as four-wave parametric interaction [5] and optical parametric oscillator [6], have been shown to result in the quantum effects of squeezed state and quantum entanglement [7]. These effects have been proved to be the basis for the research field of quantum information processing and communications [8]. Apart from the above-mentioned nonlinearities technologies, another interesting nonlinear process due to atomic coherence has attracted much attention [9], since it has potential application in the storage of quantum information and quantum memory [10,11], effective generation of squeezing without cavity [10], and multimode squeezing with possible applications to quantum imaging [12,13]. The experimental and theoretical studies have revealed that the atomic coherence of electromagnetically induced transparency (EIT) [14] played an important role in the generation of correlated photon pairs via the four-wave-mixing (FWM) process [15–19]. Thus, the combination of EIT and FWM opens the way for the generation of bright entangled beams at atomic wavelength [20]. Based on this process, a series of works for the preparation of two bright correlated beams with the wavelength of rubidium were implemented in a vapor cell [20–23]; a corresponding theoretical work was also presented to try to give the detailed effects of the FWM in an atomic system [24]. Moreover, with the development of the quantum teleportation network [25], controllable dense coding [26], and so on, the investigation of the multipartite entanglement was needed, and aroused a great deal of interest. To date, the scheme for generation of multipartite entanglement was mainly studied in the parametric process [27–31]. The investigation of multipartite entanglement with an atomic system is even important for developing the multinode quantum network. In this paper, we propose a scheme to realize the four-wave entanglement in two double-Λ atomic systems combining with FWM, and the parameters we used in the analysis show that the supposed system can be easily realized in experiment.

II. THEORETICAL MODEL

Consider a three-level system with one upper state $|3\rangle$ and two lower states $|1\rangle$ and $|2\rangle$, as shown in Fig. 1(a). A schematic experimental setup is shown in Fig. 1(b), where two strong pump beams (which can be split from one laser beam) with the same frequency ω_0 are counterpropagating through the system with the opposite wave vectors (denoted as $\vec{k}_{pF}$ and $-\vec{k}_{pB}$, respectively). A weak probe field with frequency ω_a propagates into the cell with a small angle θ. The frequencies of three levels $|l\rangle$ $(l = 1, 2, 3)$ are ω_l and their difference is $\omega_{lj} = \omega_l - \omega_j$. Both strong pump beams induce the transition $|3\rangle \rightarrow |1\rangle$ and $|3\rangle \rightarrow |2\rangle$ with detunings $\Delta = \omega_0 - \omega_{31}$ and $\delta + \Delta = \omega_0 - \omega_{32}$, where $\delta = \omega_{21}$ is the frequency difference between levels $|2\rangle$ and $|1\rangle$. The Rabi frequency of the forward (backward) pump beam (classical field) is $\bar{\Omega}_1$ $(\bar{\Omega}_2)$. We consider the generation of four quantized modes, a of frequency ω_a, b of frequency ω_b, c of frequency ω_c, and d of frequency ω_d. The wave vectors of the four quantized modes are denoted as $\vec{k}_a$, $\vec{k}_b$, $-\vec{k}_c$, and $-\vec{k}_d$ [see Fig. 1(b)]. Here we consider an ideal system in which the Doppler effects and Langevin noise operators will not be taken into account. Note that four new photons are generated when both the forward and backward pumping beams are applied.

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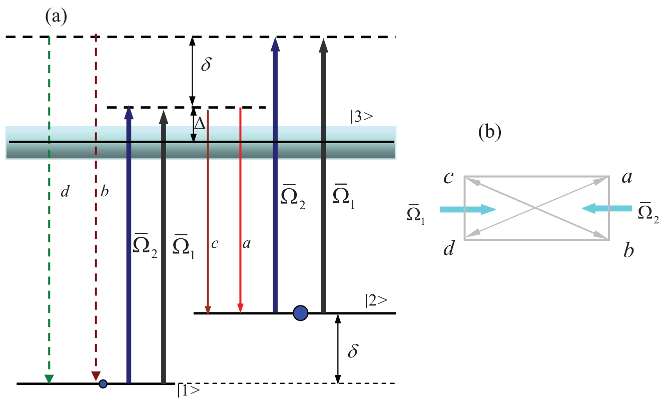

FIG. 1. (Color online) The level diagram.

If only the forward pumping beam ($\bar{\Omega}_2 = 0$) is applied, two photons will be generated (bipartite photon entanglement) [12].

A. Hamiltonian and equation of motion

We begin with the Hamiltonian under the rotation wave approximation ($\hbar = 1$),

$$H = \sum_{l=1}^{3} \omega_l \sigma_{ll} + H_V(t), \tag{1}$$

where the interaction Hamiltonian is

$$\begin{aligned} H_V(t) &= (\Omega e^{-i\omega_0 t} + B e^{-i\omega_b t})\sigma_{31} \\ &\quad + (\Omega e^{-i\omega_0 t} + A e^{-i\omega_a t})\sigma_{32} + \text{H.c.} \end{aligned} \tag{2}$$

In the above $\Omega = \bar{\Omega}_1 e^{i\vec{k}_{pF}\cdot\vec{r}} + \bar{\Omega}_2 e^{-i\vec{k}_{pB}\cdot\vec{r}}$, $A = d_{32} a e^{i\vec{k}_a\cdot\vec{r}} + d'_{32} c e^{-i\vec{k}_c\cdot\vec{r}}$, and $B = d_{31} b e^{i\vec{k}_b\cdot\vec{r}} + d'_{31} d e^{-i\vec{k}_d\cdot\vec{r}}$ with d_{31}, d_{32} and d'_{31}, d'_{32} being the coupling constants between the two atomic transitions and the four quantized modes (b,a and d,c), respectively. Here we have assumed $\omega_b = \omega_d$, $\omega_a = \omega_c$, $\vec{k}_a = \vec{k}_d$, and $\vec{k}_b = \vec{k}_c$ due to the symmetry consideration.

The dynamics of a single atom is described by the atomic operator, and satisfies the Heisenberg operator equation of motion under the dipole approximation, i.e.,

$$\dot{\sigma}_{mn} = i\omega_{mn}\sigma_{mn} + i[H_V(t),\sigma_{mn}], \quad m,n = 1,2,3. \tag{3}$$

Substituting Eq. (2) into Eq. (3), one can obtain a set of equations of motion:

$$\begin{aligned} \dot{\sigma}_{11} &= \gamma_1\sigma_{33} + i(\Omega + e^{-i\delta t}B)e^{-i\omega_0 t}\sigma_{31} \\ &\quad - i(\Omega^* + e^{i\delta t}B^+)e^{i\omega_0 t}\sigma_{13}, \end{aligned} \tag{4}$$

$$\begin{aligned} \dot{\sigma}_{22} &= \gamma_2\sigma_{33} + i(\Omega + e^{i\delta t}A)e^{-i\omega_0 t}\sigma_{32} \\ &\quad - i(\Omega^* + e^{-i\delta t}A^+)e^{i\omega_0 t}\sigma_{23}, \end{aligned} \tag{5}$$

$$\begin{aligned} \dot{\sigma}_{33} &= -(\gamma_1+\gamma_2)\sigma_{33} - i(\Omega + e^{-i\delta t}B)e^{-i\omega_0 t}\sigma_{31} \\ &\quad - i(\Omega + e^{i\delta t}A)e^{-i\omega_0 t}\sigma_{32} + i(\Omega^* + e^{i\delta t}B^+)e^{i\omega_0 t}\sigma_{13} \\ &\quad + i(\Omega^* + e^{-i\delta t}A^+)e^{i\omega_0 t}\sigma_{23}, \end{aligned} \tag{6}$$

$$\begin{aligned} \dot{\sigma}_{31} &= (i\omega_{31} - \gamma_{31})\sigma_{31} + i(\Omega^* e^{i\omega_0 t} + e^{i\omega_b t}B^+)(\sigma_{11} - \sigma_{33}) \\ &\quad + i(\Omega^* e^{i\omega_0 t} + e^{i\omega_a t}A^+)\sigma_{21}, \end{aligned} \tag{7}$$

$$\begin{aligned} \dot{\sigma}_{32} &= (i\omega_{32} - \gamma_{32})\sigma_{32} + i(\Omega^* e^{i\omega_0 t} + e^{i\omega_b t}B^+)\sigma_{12} \\ &\quad + i(\Omega^* e^{i\omega_0 t} + e^{i\omega_a t}A^+)(\sigma_{22} - \sigma_{33}), \end{aligned} \tag{8}$$

$$\begin{aligned} \dot{\sigma}_{21} &= (i\omega_{21} - \gamma_{21})\sigma_{21} - i(\Omega^* + e^{i\delta t}B^+)e^{i\omega_0 t}\sigma_{23} \\ &\quad + i(\Omega + e^{i\delta t}A)e^{-i\omega_0 t}\sigma_{31}, \end{aligned} \tag{9}$$

where $\delta = \omega_0 - \omega_a = \omega_b - \omega_0 = \omega_{21}$ and the atoms can decay from the excited state $|3\rangle$ into the two metastable states $|1\rangle$ and $|2\rangle$ with decay rates γ_1 and γ_2, respectively. γ_{31}, γ_{32}, and γ_{21} are the dephasing rates between levels $|3\rangle$ and $|1\rangle$, $|3\rangle$ and $|2\rangle$, and $|2\rangle$ and $|1\rangle$, respectively, with $\gamma_{31} = \gamma_{32} = (\gamma_1 + \gamma_2)/2$. Here the Langevin noise operators [32] are not taken into account, because their effect is small due to large single-photon detunings (δ and Δ). In Appendix A, we calculate the contribution of the Langevin noise for a two-mode case, and find it can be neglected. We can obtain the two-mode case, by setting $\bar{\Omega}_2 = d'_{32} = d'_{31} = 0$. The correlation for the two modes in the two-mode case is also presented in Appendix A.

To eliminate the fast oscillating phase terms in Eqs. (4)–(9), we introduce the following transformations:

$$\begin{aligned} Q_{mm} &= \sigma_{mm}, \quad (m = 1,2,3) \\ Q_{31} &= \sigma_{31}e^{-i\omega_0 t}, \\ Q_{32} &= \sigma_{32}e^{-i\omega_0 t}, \\ Q_{21} &= \sigma_{21}. \end{aligned} \tag{10}$$

Then the interaction Hamiltonian becomes

$$\begin{aligned} H_V(t) &= (\Omega e^{-i\omega_0 t} + B e^{-i\omega_b t})Q_{31}e^{i\omega_0 t} \\ &\quad + (\Omega e^{-i\omega_0 t} + A e^{-i\omega_a t})Q_{32}e^{i\omega_0 t} + \text{H.c.} \\ &= (\Omega + B e^{-i\delta t})Q_{31} + (\Omega + A e^{i\delta t})Q_{32} + \text{H.c.}, \end{aligned} \tag{11}$$

and the equations of atomic operators become

$$\begin{aligned} \dot{Q}_{11} &= \gamma_1 Q_{33} - i(\Omega^* + e^{i\delta t}B^+)Q_{13} + i(\Omega + e^{-i\delta t}B)Q_{31}, \\ \dot{Q}_{22} &= \gamma_2 Q_{33} - i(\Omega^* + e^{-i\delta t}A^+)Q_{23} + i(\Omega + e^{i\delta t}A)Q_{32}, \\ \dot{Q}_{33} &= -(\gamma_1+\gamma_2)Q_{33} - i(\Omega + e^{-i\delta t}B)Q_{31} \\ &\quad - i(\Omega + e^{i\delta t}A)Q_{32} + i(\Omega^* + e^{i\delta t}B^+)Q_{13} \\ &\quad + i(\Omega^* + e^{-i\delta t}A^+)Q_{23}, \end{aligned} \tag{12}$$

$$\begin{aligned} \dot{Q}_{31} &= i\Gamma_p Q_{31} + i(\Omega^* + e^{i\delta t}B^+)(Q_{11} - Q_{33}) \\ &\quad + i(\Omega^* + e^{-i\delta t}A^+)Q_{21}, \\ \dot{Q}_{32} &= -i\Gamma_a Q_{32} + i(\Omega^* + e^{i\delta t}B^+)Q_{12} \\ &\quad + i(\Omega^* + e^{-i\delta t}A^+)(Q_{22} - Q_{33}), \\ \dot{Q}_{21} &= i\Gamma_{21}Q_{21} - i(\Omega^* + e^{i\delta t}B^+)Q_{23} + i(\Omega + e^{i\delta t}A)Q_{31}, \end{aligned} \tag{13}$$

where $\Gamma_p = -\Delta + i\gamma_{31}$, $\Gamma_a = \delta + \Delta - i\gamma_{32}$, $\Gamma_{21} = \delta + i\gamma_{21}$, and $\Delta = \omega_0 - \omega_{31}$.

Here we use the perturbation method to solve Q_{31} and Q_{23}:

$$\begin{aligned} Q_{31} &= Q_{31}^{(0)} + Q_{31}^{(1)} + Q_{31}^{(2)} + \cdots, \\ Q_{23} &= Q_{23}^{(0)} + Q_{23}^{(1)} + Q_{23}^{(2)} + \cdots. \end{aligned} \tag{14}$$

Note that $Q_{31}^{(0)}$ or $Q_{23}^{(0)}$ does not involve the quantized mode operators, $Q_{31}^{(1)}$ and $Q_{23}^{(1)}$ involve a single operator, and $Q_{31}^{(2)}$ or $Q_{23}^{(2)}$ contains two operators. Substituting Eq. (14) into Eq. (11) and keeping the terms containing the quantized modes, A and B (i.e., a, b, c, d), to the second order (all orders for Ω), we have an effective Hamiltonian,

$$\begin{aligned} H_V(t) &= (\Omega + Be^{-i\delta t})\big(Q_{31}^{(0)} + Q_{31}^{(1)} + Q_{31}^{(2)}\big) \\ &\quad + (\Omega^* + A^+e^{-i\delta t})\big(Q_{23}^{(0)} + Q_{23}^{(1)} + Q_{23}^{(2)}\big) + \text{H.c.} \\ &\approx Be^{-i\delta t}Q_{31}^{(1)} + \Omega Q_{31}^{(2)} + A^+e^{-i\delta t}Q_{23}^{(1)} + \Omega^* Q_{23}^{(2)} + \text{H.c.} \end{aligned} \tag{15}$$

The last equals sign is present because the terms having a single quantized mode operator could not satisfy the phase-matching condition and can be neglected, while the constant term is just a shift.

B. Perturbation solution for Q_{31} and Q_{32}

As the generated four quantized modes and the probe are weak, we can use the perturbation method by keeping them to the second order and keeping the classical pump field to all orders. The steady solution of the zeroth order is (see Appendix B for details)

$$Q_{11}^{(0)} = \frac{1}{G_2}[\delta^2(\Delta^2 + \gamma^2) + \delta(\delta + 2\Delta)|\Omega|^2 + 4|\Omega|^4], \tag{16}$$

$$\begin{aligned} Q_{22}^{(0)} = \frac{1}{G_2}[&\delta^2(\Delta^2 + 2\Delta\delta + \gamma^2 + \delta^2) \\ &- \delta(\delta + 2\Delta)|\Omega|^2 + 4|\Omega|^4], \end{aligned} \tag{17}$$

$$Q_{33}^{(0)} = \frac{2\delta^2}{G_2}|\Omega|^2, \tag{18}$$

$$Q_{31}^{(0)} = \frac{\delta}{G_2}\Omega^*[(i\gamma + \Delta)\delta + 2|\Omega|^2] = \big(Q_{13}^{(0)}\big)^*, \tag{19}$$

$$Q_{23}^{(0)} = \frac{\delta}{G_2}\Omega[(\delta + \Delta - i\gamma)\delta - 2|\Omega|^2] = \big(Q_{32}^{(0)}\big)^*, \tag{20}$$

$$Q_{21}^{(0)} = \frac{|\Omega|^2}{G_2}[\delta(\delta - 2i\gamma) - 4|\Omega|^2] = \big(Q_{12}^{(0)}\big)^*, \tag{21}$$

where

$$G_2 = \delta^2(\delta^2 + 2\Delta\delta + 2\Delta^2 + 2\gamma^2) + 2\delta^2|\Omega|^2 + 8|\Omega|^4, \tag{22}$$

and we have taken $\gamma_1 = \gamma_2 = \gamma_{31} = \gamma_{32} = \gamma$ and set $\gamma_{21} = 0$. From Eqs. (16)–(21) one can clearly see, under the conditions $|\Delta/\delta|^2 \approx 0$ and $|\gamma/\delta|^2$, $|\Omega/\delta|^2$, $|\Delta\Omega/\delta^2| \to 0$, that $Q_{11}^{(0)} \approx Q_{33}^{(0)} \approx Q_{31}^{(0)} \approx Q_{23}^{(0)} \approx Q_{21}^{(0)} \approx 0$ and $Q_{22}^{(0)} \approx 1$, which indicates that the strong pump and weak probe configuration put all the atomic population approximately in the state of $|2\rangle$.

In a similar way, one can also obtain the first-order and the second-order steady solutions for $Q_{31}^{(1)}$, $Q_{23}^{(1)}$, and $Q_{31}^{(2)}$, $Q_{23}^{(2)}$ (see Appendix C for details), which are Eqs. (C6) and (C22). In the system of M three-level atoms, the phase-match conditions must be satisfied in order to have the output of the four modes.

C. Effective Hamiltonian

Now let us obtain an effective Hamiltonian which is equivalent to the Hamiltonian, Eq. (1), under the second-order perturbation approximation. Substituting Eq. (14), Eqs. (16)–(21), and Eqs. (C6), (C9), (C22), and (C25) in Appendix C into Eq. (2) and noticing $B = d_{31}be^{i\vec{k}_b\cdot\vec{r}} + d'_{31}de^{-i\vec{k}_d\cdot\vec{r}}$ and $\Omega = \bar{\Omega}_1 e^{i\vec{k}_{pF}\cdot\vec{r}} + \bar{\Omega}_2 e^{-i\vec{k}_{pB}\cdot\vec{r}}$, $A = d_{32}ae^{i\vec{k}_a\cdot\vec{r}} + d'_{32}ce^{-i\vec{k}_c\cdot\vec{r}}$, we can obtain the effective Hamiltonian in the interaction picture (see Appendix D):

$$H_{\text{eff}} = \kappa_1 a^+b^+ + \kappa_2 c^+d^+ + 2\sqrt{\kappa_1\kappa_2}(a^+d^+ + c^+b^+) + \text{H.c.}, \tag{23}$$

$$\kappa_1 = \kappa\bar{\Omega}_1^2 d_{31}^* d_{32}^*, \quad \kappa_2 = \kappa\bar{\Omega}_2^2 d_{31}^* d_{32}^*, \tag{24}$$

$$\begin{aligned} \kappa = \frac{4\delta^6(\delta + 2\Delta)}{[\delta^2(\delta^2 + 2\Delta\delta + 2\Delta^2)]^3}\Bigg\{&\frac{[\gamma^2 + (\delta + \Delta)^2](\gamma^2 + \Delta^2)}{(|\bar{\Omega}_1|^2 + |\bar{\Omega}_2|^2)} \\ &+ (\delta^2 + 6\delta\Delta + 6\Delta^2 - 2\gamma^2)\Bigg\}, \end{aligned} \tag{25}$$

where $d_{32} \approx d'_{32}$ and $d_{31} \approx d'_{31}$ have been used due to the very close frequencies of the four modes. Please note that we will have the two-mode case by setting $\bar{\Omega}_2 = 0$. In Eq. (23), we keep the correlation terms, and have dropped the constant terms which lead to an overall shift and the linear terms (containing one operator of the four modes) which lead to a frequency shift of the atomic levels. When the backward pump field is absent ($\bar{\Omega}_2 = 0$), Eq. (23) reduces to the two-mode case. For convenience, we set $\kappa_1 = |\kappa_1|\,e^{i\phi} = i\kappa_f^2 e^{i(\phi-\pi/2)}, \kappa_2 = |\kappa_2|\,e^{i\phi'} = i\kappa_b^2 e^{i(\phi'-\pi/2)}$, $a^+e^{i(\phi-\pi/2)/2} = \tilde{a}^+$, $b^+e^{i(\phi-\pi/2)/2} = \tilde{b}^+$, $c^+e^{i(\phi'-\pi/2)/2} = \tilde{c}^+$, and $d^+e^{i(\phi'-\pi/2)/2} = \tilde{d}^+$. Thus Eq. (23) can be rewritten as

$$H_{\text{eff}} = i\big[\kappa_f^2\tilde{a}^+\tilde{b}^+ + \kappa_b^2\tilde{c}^+\tilde{d}^+ + 2\kappa_f\kappa_b(\tilde{a}^+\tilde{d}^+ + \tilde{c}^+\tilde{b}^+)\big] + \text{H.c.}, \tag{26}$$

where

$$\begin{aligned} \kappa_f^2 &= \big|\kappa\bar{\Omega}_1^2 d_{31}^* d_{32}^*\big|, \\ \kappa_b^2 &= \big|\kappa\bar{\Omega}_2^2 d_{31}^* d_{32}^*\big|, \\ \kappa_f\kappa_b &= |\kappa\bar{\Omega}_1\bar{\Omega}_2 d_{31}^* d_{32}^*|. \end{aligned} \tag{27}$$

The generation of the four modes is due to the following four processes, which satisfy the phase-matching condition: the first, absorbing a forward pumping photon and then generating a photon of ω_b with an atom from level $|2\rangle$ to level $|1\rangle$ via level $|3\rangle$, and then absorbing a forward pumping photon and then generating a photon of ω_a with an atom from level $|1\rangle$ back to level $|2\rangle$ again via level $|3\rangle$; the second, absorbing a backward pumping photon and then generating a photon of ω_d with an atom from level $|2\rangle$ to level $|1\rangle$ via level $|3\rangle$, and then absorbing a backward pumping photon and then generating a photon of ω_c with an atom from level $|1\rangle$ back to level $|2\rangle$ again via level $|3\rangle$; the third, absorbing a forward pumping photon and then generating a photon of ω_b with an atom from level $|2\rangle$ to level $|1\rangle$ via level $|3\rangle$, and then absorbing a backward pumping photon and then generating a photon of ω_c with an atom from level $|1\rangle$ back to level $|2\rangle$ again via level $|3\rangle$; the fourth, absorbing a backward pumping photon and then generating a photon of ω_d with an atom from level $|2\rangle$ to level

$|1\rangle$ via level $|3\rangle$, and then absorbing a forward pumping photon and then generating a photon of ω_a with an atom from level $|1\rangle$ back to level $|2\rangle$ again via level $|3\rangle$.

III. EQUATIONS OF MOTION FOR THE FOUR QUANTIZED MODES

From Eq. (26) one can find the Heisenberg equations of motion for the four quantized modes,

$$\frac{d\tilde{a}}{dt} = \kappa_f^2\tilde{b}^+ + 2\kappa_f\kappa_b\tilde{d}^+, \quad \frac{d\tilde{b}}{dt} = \kappa_f^2\tilde{a}^+ + 2\kappa_f\kappa_b\tilde{c}^+,$$
$$\frac{d\tilde{c}}{dt} = \kappa_b^2\tilde{d}^+ + 2\kappa_f\kappa_b\tilde{b}^+, \quad \frac{d\tilde{d}}{dt} = \kappa_b^2\tilde{c}^+ + 2\kappa_f\kappa_b\tilde{a}^+. \tag{28}$$

We introduce the quadrature operators for each mode as $X_o = (o + o^+)/\sqrt{2}$, $P_o = (o - o^+)/(i\sqrt{2})$, $[X_o, P_o] = i$, $o = \tilde{a}, \tilde{b}, \tilde{c}, \tilde{d}$. Using the quadrature components X_o, P_o to re-form Eq. (28) into the following forms,

$$\frac{dX_{\tilde{a}}}{dt} = \kappa_f^2 X_{\tilde{b}} + 2\kappa_f\kappa_b X_{\tilde{d}}, \quad \frac{dX_{\tilde{b}}}{dt} = \kappa_f^2 X_{\tilde{a}} + 2\kappa_f\kappa_b X_{\tilde{c}},$$
$$\frac{dX_{\tilde{c}}}{dt} = \kappa_b^2 X_{\tilde{d}} + 2\kappa_f\kappa_b X_{\tilde{b}}, \quad \frac{dX_{\tilde{d}}}{dt} = \kappa_b^2 X_{\tilde{c}} + 2\kappa_f\kappa_b X_{\tilde{a}}, \tag{29}$$

and

$$\frac{dP_{\tilde{a}}}{dt} = -\kappa_f^2 P_{\tilde{b}} - 2\kappa_f\kappa_b P_{\tilde{d}}, \quad \frac{dP_{\tilde{b}}}{dt} = -\kappa_f^2 P_{\tilde{a}} - 2\kappa_f\kappa_b P_{\tilde{c}},$$
$$\frac{dP_{\tilde{c}}}{dt} = -\kappa_b^2 P_{\tilde{d}} - 2\kappa_f\kappa_b P_{\tilde{b}}, \quad \frac{dP_{\tilde{d}}}{dt} = -\kappa_b^2 P_{\tilde{c}} - 2\kappa_f\kappa_b P_{\tilde{a}}. \tag{30}$$

These equations (29) and (30) can be solved analytically with their initial values,

$$\begin{aligned}
X_{\tilde{a}}(t) &= \lambda_1 X_{\tilde{a}}(0) + \lambda_2 X_{\tilde{b}}(0) + \lambda_3 X_{\tilde{c}}(0) + \lambda_4 X_{\tilde{d}}(0),\\
X_{\tilde{b}}(t) &= \lambda_2 X_{\tilde{a}}(0) + \lambda_1 X_{\tilde{b}}(0) + \lambda_4 X_{\tilde{c}}(0) + \lambda_3 X_{\tilde{d}}(0),\\
X_{\tilde{c}}(t) &= \lambda_3 X_{\tilde{a}}(0) + \lambda_4 X_{\tilde{b}}(0) + \lambda_5 X_{\tilde{c}}(0) + \lambda_6 X_{\tilde{d}}(0),\\
X_{\tilde{d}}(t) &= \lambda_4 X_{\tilde{a}}(0) + \lambda_3 X_{\tilde{b}}(0) + \lambda_6 X_{\tilde{c}}(0) + \lambda_5 X_{\tilde{d}}(0),
\end{aligned} \tag{31}$$

and

$$\begin{aligned}
P_{\tilde{a}}(t) &= \lambda_1 P_{\tilde{a}}(0) - \lambda_2 P_{\tilde{b}}(0) + \lambda_3 P_{\tilde{c}}(0) - \lambda_4 P_{\tilde{d}}(0),\\
P_{\tilde{b}}(t) &= -\lambda_2 P_{\tilde{a}}(0) + \lambda_1 P_{\tilde{b}}(0) - \lambda_4 P_{\tilde{c}}(0) + \lambda_3 P_{\tilde{d}}(0),\\
P_{\tilde{c}}(t) &= \lambda_3 P_{\tilde{a}}(0) - \lambda_4 P_{\tilde{b}}(0) + \lambda_5 P_{\tilde{c}}(0) - \lambda_6 P_{\tilde{d}}(0),\\
P_{\tilde{d}}(t) &= -\lambda_4 P_{\tilde{a}}(0) + \lambda_3 P_{\tilde{b}}(0) - \lambda_6 P_{\tilde{c}}(0) + \lambda_5 P_{\tilde{d}}(0),
\end{aligned} \tag{32}$$

where

$$\begin{aligned}
\lambda_1 &= \frac{1}{2\xi}[\xi_+\cosh(\eta_+ t) + \xi_-\cosh(\eta_- t)],\\
\lambda_2 &= \frac{1}{2\xi}[\xi_+\sinh(\eta_+ t) + \xi_-\sinh(\eta_- t)],
\end{aligned} \tag{33}$$

$$\begin{aligned}
\lambda_3 &= \frac{2\kappa_f\kappa_b}{\xi}[\cosh(\eta_+ t) - \cosh(\eta_- t)],\\
\lambda_4 &= \frac{2\kappa_f\kappa_b}{\xi}[\sinh(\eta_+ t) - \sinh(\eta_- t)],\\
\lambda_5 &= \frac{1}{2\xi}[\xi_-\cosh(\eta_+ t) + \xi_+\cosh(\eta_- t)],\\
\lambda_6 &= \frac{1}{2\xi}[\xi_-\sinh(\eta_+ t) + \xi_+\sinh(\eta_- t)],
\end{aligned} \tag{34}$$

with $\xi = \sqrt{\kappa_f^4 + 14\kappa_f^2\kappa_b^2 + \kappa_b^4}$, $\xi_\pm = \xi \pm (\kappa_f^2 - \kappa_b^2)$, and $\eta_\pm = (\kappa_f^2 + \kappa_b^2 \pm \xi)/2$. All the information needed to calculate the van Loock–Furusawa (VLF) correlation [25] is contained in Eqs. (31) and (32).

IV. FOUR-MODE ENTANGLEMENT

For any quantum system, a set of sufficient conditions for multimode entanglement is derived by VLF. For the four-mode system under consideration and our definitions for quadrature $[X_o = (o + o^+)/\sqrt{2},\ P_o = (o - o^+)/(i\sqrt{2}),$ $[X_o, P_o] = i,\ o = \tilde{a}, \tilde{b}, \tilde{c}, \tilde{d}]$, the VLF inequalities can be given by

$$\begin{aligned}
V_{\tilde{a}\tilde{b}} &= \delta^2(X_{\tilde{a}} - X_{\tilde{b}}) + \delta^2(P_{\tilde{a}} + P_{\tilde{b}} + g_c P_{\tilde{c}} + g_d P_{\tilde{d}}) < 2,\\
V_{\tilde{a}\tilde{c}} &= \delta^2(X_{\tilde{a}} - X_{\tilde{c}}) + \delta^2(P_{\tilde{a}} + g_b P_{\tilde{b}} + P_{\tilde{c}} + g_d' P_{\tilde{d}}) < 2,\\
V_{\tilde{a}\tilde{d}} &= \delta^2(X_{\tilde{a}} - X_{\tilde{d}}) + \delta^2(P_{\tilde{a}} + g_b' P_{\tilde{b}} + g_c' P_{\tilde{c}} + P_{\tilde{d}}) < 2,\\
V_{\tilde{b}\tilde{c}} &= \delta^2(X_{\tilde{b}} - X_{\tilde{c}}) + \delta^2(g_a P_{\tilde{a}} + P_{\tilde{b}} + P_{\tilde{c}} + g_d'' P_{\tilde{d}}) < 2,\\
V_{\tilde{b}\tilde{d}} &= \delta^2(X_{\tilde{b}} - X_{\tilde{d}}) + \delta^2(g_a' P_{\tilde{a}} + P_{\tilde{b}} + g_c'' P_{\tilde{c}} + P_{\tilde{d}}) < 2,\\
V_{\tilde{c}\tilde{d}} &= \delta^2(X_{\tilde{c}} - X_{\tilde{d}}) + \delta^2(g_a'' P_{\tilde{a}} + g_b'' P_{\tilde{b}} + P_{\tilde{c}} + P_{\tilde{d}}) < 2,
\end{aligned} \tag{35}$$

where $\delta^2(X) = \langle X^2\rangle - \langle X\rangle^2$ and the parameters g_i, g_i', g_i'' are arbitrary real numbers. As shown in Refs. [25,29], among the above six inequalities, three conditions are sufficient to verify the full inseparability of a four-mode, four-party state. Thus we may choose

$$V_{\tilde{a}\tilde{b}} < 2, \quad V_{\tilde{b}\tilde{c}} < 2, \quad V_{\tilde{c}\tilde{d}} < 2. \tag{36}$$

The system is fully inseparable and there is genuine fourfold entanglement if the three inequalities are all satisfied. Please note that Eq. (35) or Eq. (36) is the sufficient condition, but not the necessary condition for a fourfold entanglement. For further obtaining the expression, say $V_{\tilde{a}\tilde{b}}$, these average values $\langle\tilde{a}^2\rangle$, $\langle\tilde{a}^{+2}\rangle$, $\langle\tilde{b}^2\rangle$, $\langle\tilde{b}^{+2}\rangle$, $\langle\tilde{a}\tilde{b}^+\rangle$, $\langle\tilde{a}^+\tilde{b}\rangle$, and $\langle\tilde{a}\rangle$, $\langle\tilde{a}^+\rangle$, $\langle\tilde{b}\rangle$, $\langle\tilde{b}^+\rangle$ are needed. For this system, at the initial time $t = 0$, all the four modes are in vacuum; thus we have $\langle\tilde{a}(0)\rangle = \langle\tilde{a}^+(0)\rangle = \langle\tilde{b}(0)\rangle = \langle\tilde{b}^+(0)\rangle = 0$, which leads to the average values of the amplitude all being zero, i.e., $\langle X_o(0)\rangle = \langle P_o(0)\rangle = 0$ and $\langle X_o(t)\rangle = \langle P_o(t)\rangle = 0$. Due to the bosonic communication relations, not all the moments vanish, such as $\langle X_o(0)X_{o'}(0)\rangle = \langle P_o(0)P_{o'}(0)\rangle = \delta_{oo'}/2$, and $\langle X_o(0)P_{o'}(0)\rangle = -\langle P_{o'}(0)X_o(0)\rangle = i\delta_{oo'}/2$. The variances at time t can be obtained based on Eqs. (31) and (32),

$$\begin{aligned}
\delta^2 X_{\tilde{a}} &= \delta^2 X_{\tilde{b}} = \delta^2 P_{\tilde{a}} = \delta^2 P_{\tilde{b}}\\
&= \frac{1}{4\xi}[\xi_+\cosh(2\eta_+ t) + \xi_-\cosh(2\eta_- t)],
\end{aligned} \tag{37}$$

$$\begin{aligned}
\delta^2 X_{\tilde{c}} &= \delta^2 X_{\tilde{d}} = \delta^2 P_{\tilde{c}} = \delta^2 P_{\tilde{d}}\\
&= \frac{1}{4\xi}[\xi_-\cosh(2\eta_+ t) + \xi_+\cosh(2\eta_- t)],
\end{aligned} \tag{38}$$

and

$$\begin{aligned}
\langle X_{\tilde{a}}X_{\tilde{b}}\rangle &= -\langle P_{\tilde{a}}P_{\tilde{b}}\rangle = \frac{1}{4\xi}[\xi_+\sinh(2\eta_+ t) + \xi_-\sinh(2\eta_- t)],\\
\langle X_{\tilde{c}}X_{\tilde{d}}\rangle &= -\langle P_{\tilde{c}}P_{\tilde{d}}\rangle = \frac{1}{4\xi}[\xi_-\sinh(2\eta_+ t) + \xi_+\sinh(2\eta_- t)],
\end{aligned} \tag{39}$$

$$\begin{aligned}\langle X_{\tilde{a}} X_{\tilde{c}}\rangle &= \langle P_{\tilde{a}} P_{\tilde{c}}\rangle = \langle X_{\tilde{b}} X_{\tilde{d}}\rangle = \langle P_{\tilde{b}} P_{\tilde{d}}\rangle \\ &= \frac{\kappa_f \kappa_b}{\xi}[\cosh(2\eta_+ t) - \cosh(2\eta_- t)], \\ \langle X_{\tilde{a}} X_{\tilde{d}}\rangle &= -\langle P_{\tilde{a}} P_{\tilde{d}}\rangle = \langle X_{\tilde{b}} X_{\tilde{c}}\rangle = -\langle P_{\tilde{b}} P_{\tilde{c}}\rangle \\ &= \frac{\kappa_f \kappa_b}{\xi}[\sinh(2\eta_+ t) - \sinh(2\eta_- t)].\end{aligned} \tag{40}$$

As the average values of the amplitudes are all zero, the above average values are actually the covariances.

Next, we investigate optimize of the VLF criteria by using the freedom allowed in the choice of g_i, g_i', g_i'', which are arbitrary real. Using the relations $\langle X_o(t)\rangle = \langle P_o(t)\rangle = 0$, the required variance can be rewritten as, taking $V_{\tilde{a}\tilde{b}}$ as an example,

$$\begin{aligned}V_{\tilde{a}\tilde{b}} &= 4\delta^2 X_{\tilde{a}} + \left(g_c^2 + g_d^2\right)\delta^2 P_{\tilde{c}} - 4\langle X_{\tilde{a}} X_{\tilde{b}}\rangle \\ &\quad + 2(g_c + g_d)\langle P_{\tilde{b}} P_{\tilde{c}} + P_{\tilde{a}} P_{\tilde{c}}\rangle + 2 g_c g_d \langle P_{\tilde{c}} P_{\tilde{d}}\rangle.\end{aligned} \tag{41}$$

Following Ref. [33], a simple minimization of the right-hand sides of Eq. (41) with respect to the g_c and g_d gives

$$g_c = g_d = -\frac{\langle P_{\tilde{a}} P_{\tilde{c}}\rangle + \langle P_{\tilde{b}} P_{\tilde{c}}\rangle}{\langle P_{\tilde{c}} P_{\tilde{d}}\rangle + \left\langle P_{\tilde{d}}^2\right\rangle}, \tag{42}$$

where we have used the relations in Eqs. (37)–(40). Once this optimization process has taken place, one can find

$$\begin{aligned}V_{\tilde{a}\tilde{b}} &= 4\delta^2 X_{\tilde{a}} + 2 g_c^2 \delta^2 P_{\tilde{c}} - 4\langle X_{\tilde{a}} X_{\tilde{b}}\rangle \\ &\quad + 4 g_c \langle P_{\tilde{b}} P_{\tilde{c}} + P_{\tilde{a}} P_{\tilde{c}}\rangle + 2 g_c^2 \langle P_{\tilde{c}} P_{\tilde{d}}\rangle.\end{aligned} \tag{43}$$

Similarly, one can obtain that

$$\begin{aligned}V_{\tilde{b}\tilde{c}} &= \left(2 + g_a^2\right)\delta^2 X_{\tilde{b}} + \left(2 + g_d''^2\right)\delta^2 X_{\tilde{c}} - 4\langle X_{\tilde{b}} X_{\tilde{c}}\rangle \\ &\quad + 2 g_a g_d'' \langle P_{\tilde{a}} P_{\tilde{d}}\rangle + 2 g_a(\langle P_{\tilde{a}} P_{\tilde{b}}\rangle + \langle P_{\tilde{a}} P_{\tilde{c}}\rangle) \\ &\quad + 2 g_d'' \langle P_{\tilde{b}} P_{\tilde{d}} + P_{\tilde{c}} P_{\tilde{d}}\rangle, \\ V_{\tilde{c}\tilde{d}} &= 4\delta^2 X_{\tilde{c}} + 2 g_a''^2 \delta^2 P_{\tilde{a}} - 4\langle X_{\tilde{c}} X_{\tilde{d}}\rangle \\ &\quad + 4 g_a'' \langle P_{\tilde{a}} P_{\tilde{c}} + P_{\tilde{a}} P_{\tilde{d}}\rangle + 2 g_a''^2 \langle P_{\tilde{a}} P_{\tilde{b}}\rangle,\end{aligned} \tag{44}$$

where we have set

$$\begin{aligned}g_a &= [(\langle P_{\tilde{a}} P_{\tilde{b}}\rangle + \langle P_{\tilde{a}} P_{\tilde{c}}\rangle)\delta^2 P_{\tilde{d}} \\ &\quad - (\langle P_{\tilde{b}} P_{\tilde{d}} + P_{\tilde{c}} P_{\tilde{d}}\rangle)\langle P_{\tilde{a}} P_{\tilde{d}}\rangle]/[\langle P_{\tilde{a}} P_{\tilde{d}}\rangle^2 - \delta^2 P_{\tilde{a}} \delta^2 P_{\tilde{d}}], \\ g_d'' &= [\langle P_{\tilde{b}} P_{\tilde{d}} + P_{\tilde{c}} P_{\tilde{d}}\rangle \delta^2 P_{\tilde{a}} - (\langle P_{\tilde{a}} P_{\tilde{b}}\rangle \\ &\quad + \langle P_{\tilde{a}} P_{\tilde{c}}\rangle)\langle P_{\tilde{a}} P_{\tilde{d}}\rangle]/[\langle P_{\tilde{a}} P_{\tilde{d}}\rangle^2 - \delta^2 P_{\tilde{a}} \delta^2 P_{\tilde{d}}], \\ g_a'' &= -\langle P_{\tilde{a}} P_{\tilde{c}} + P_{\tilde{a}} P_{\tilde{d}}\rangle/(\langle P_{\tilde{a}} P_{\tilde{b}}\rangle + \delta^2 P_{\tilde{a}}).\end{aligned}$$

V. DISCUSSIONS

First, let us consider the two-mode entanglement case. As mentioned above, we can obtain the two-mode entanglement case by taking $\bar{\Omega}_2 = d_{32}' = d_{31}' = 0$. In this case, we have $\kappa_b = 0$, $\xi = \kappa_f^2$ and $\eta_+ = \xi$, $\eta_- = 0$, and from Eqs. (37), (39), and (43) we can find that $V_{\tilde{a}\tilde{b}} = 2e^{-2\kappa_f^2 t} < 2$, which demonstrates the genuine bipartite entanglement, as expected [12]. In Fig. 2, for the two-mode case, we plot the graph of squeezing parameter κ_f^2 as the function of $\Omega(=\bar{\Omega}_1)$ and Δ for several given values of δ: $G \equiv \sqrt{M} d_{31} = \sqrt{M} d_{32} = -250$ MHz (M is the atom number density), and $\gamma = 4.56$ MHz, (a) $\delta = 3.0$ GHz, (b) $\delta = 9.2$ GHz. From Fig. 2 one can clearly see that κ_f^2 increases with the decreasing value of δ and the increasing absolute value of Δ (especially for $\Delta > 0$), respectively. In addition, for the case of $\Delta > 0$, κ_f^2 becomes bigger as the increasing value Ω. This case is not true for the case of $\Delta < 0$. It is interesting to notice that the effect of Δ on κ_f^2 is unsymmetrical, i.e., one can obtain a bigger value of κ_f^2 with the increasing value of $\Delta > 0$ than that with the increasing absolute value of $\Delta < 0$ for a given value of δ. Especially, κ_f^2 is very small when the resonance is present ($\Delta = 0$), that is to say, an appreciable value of $\Delta \neq 0$ is necessary to generate the observable entanglement.

Next, we turn to the general four-mode case with asymmetric pumping strengths and arbitrary detuning. Substituting Eqs. (37)–(40) into Eqs. (43) and (44), the analytical expressions can be found, but the expression will be very complicated. Here we use numerical calculation to present the results. In Fig. 3, we plot the VLF correlations of the four quantized modes for different pumping detunings Δ: (a) $\Delta = 34.78\gamma$, (b) $\Delta = 104.35\gamma$, (c) $\Delta = 173.91\gamma$, (d) $\Delta = 347.83\gamma$, with $\delta = 0.52 \times 10^3\gamma$, $\bar{\Omega}_1 = 104.35\gamma$ ($\bar{\Omega}_2 = 0.8\bar{\Omega}_1$), and $G = -43.48\gamma$. These parameters are based on the Rb atoms [34–36]. The black line is for $V_{\tilde{c}\tilde{d}}$, red line is for $V_{\tilde{a}\tilde{b}}$, and blue line is for $V_{\tilde{b}\tilde{c}}$ (for simplicity, the tilt is neglected in all figures). From Fig. 3, we find that three correlations fall below 2 in a certain interaction time range, which demonstrates the quadripartite entanglement. Furthermore, the correlation $V_{\tilde{b}\tilde{c}}$ between $\tilde{b}$ and $\tilde{c}$ modes seems stronger than those involved in the other two modes. The interaction time range, within which quadripartite entanglement is present, decreases with the increasing detune Δ. In order to observe the quadripartite entanglement with other parameters fixed, small

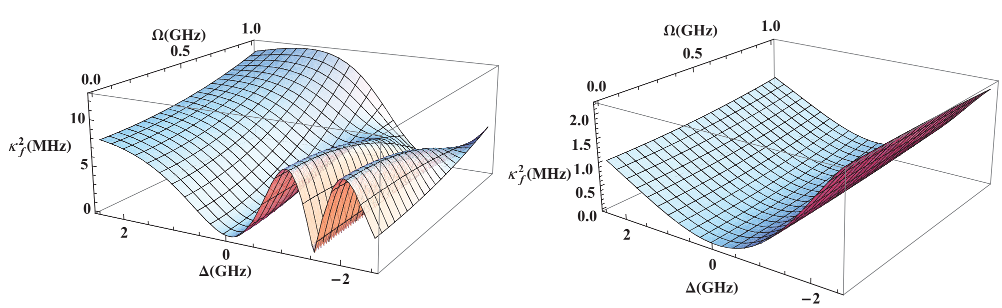

FIG. 2. (Color online) The squeezing parameter κ_f^2 as the function of $\Omega(=\bar{\Omega}_1)$, and Δ with $G = -250$ MHz and $\gamma = 4.56$ MHz; (a) $\delta = 3.0$ GHz, (b) $\delta = 9.2$ GHz.

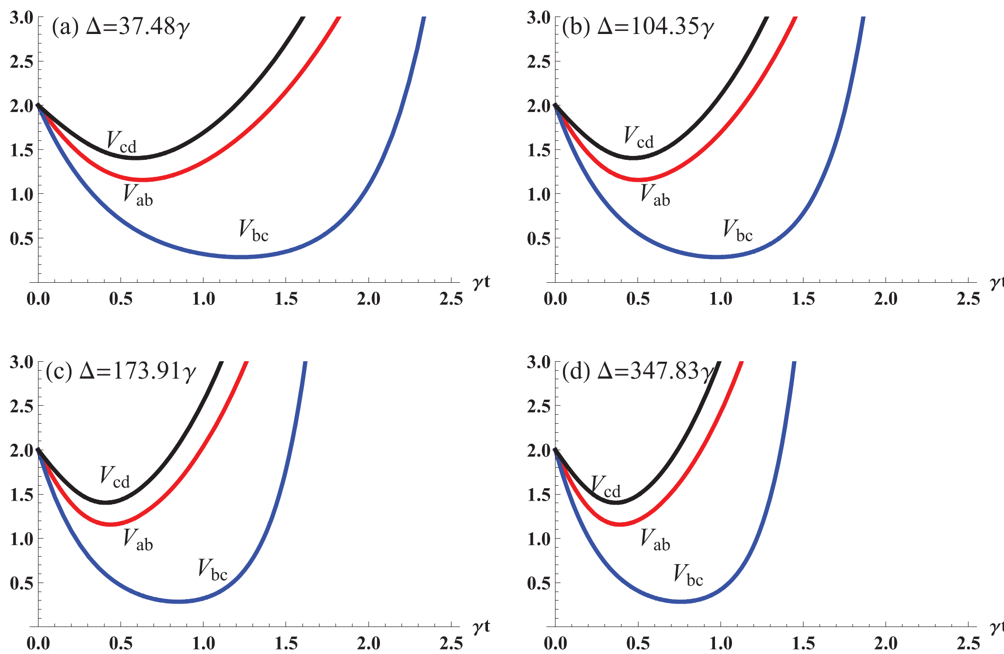

FIG. 3. (Color online) The VLF correlations for (a) $\Delta = 34.78\gamma$, (b) $\Delta = 104.35\gamma$, (c) $\Delta = 173.91\gamma$, (d) $\Delta = 347.83\gamma$ with $\delta = 0.52 \times 10^3\gamma$ and $\bar{\Omega}_1 = 104.35\gamma$, as well as $G = -43.48\gamma$.

Δ corresponds to a long interaction time (or long vapor cell), and large Δ corresponds to a short interaction time (or short vapor cell).

In Fig. 4, we plot the VLF correlations for different combination of δ, Δ, and the pumping Rabi frequency. Comparing Figs. 4(a) and 4(b) with Figs. 4(c) and 4(d), we find that we can have the quadripartite entanglement over a longer range of interaction time for a bigger δ/γ. Therefore, the interaction time to observe the entanglement is different for different δ. The bigger δ/γ is, the longer interaction time is needed. As shown in Figs. 3 and 4, there is an unambiguous demonstration of the inseparability of the four modes as soon as the interaction begins. In Fig. 5, we present the dependence of the VLF correlation of the four modes on the coupling constant G ranging from -43.86γ to -76.75γ, which tells us that the quadripartite entanglement can exist over a long

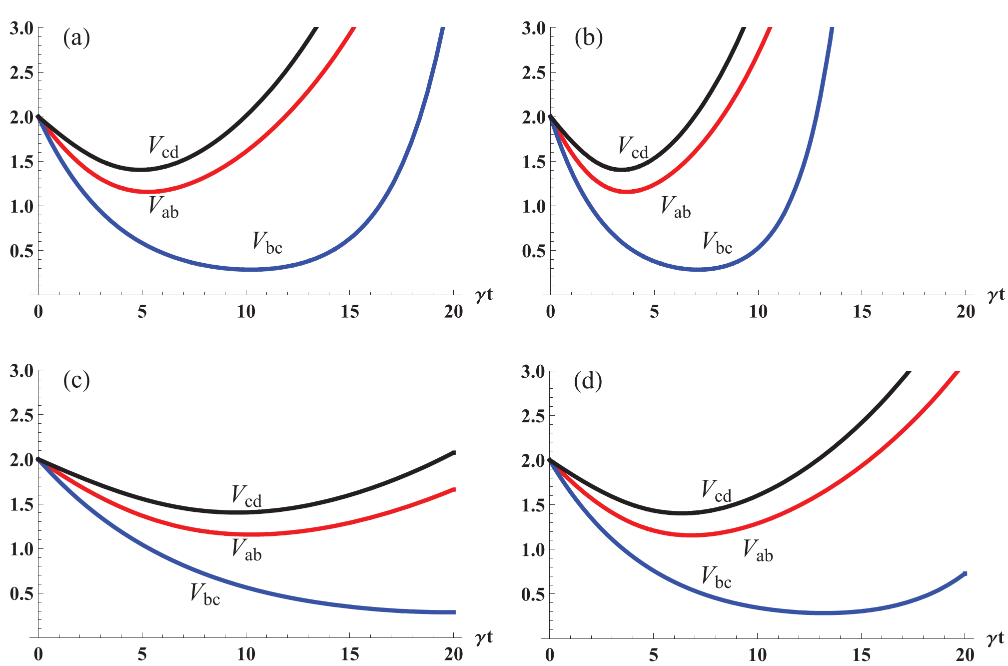

FIG. 4. (Color online) The VLF correlations for (a) $\Delta = 104.35\gamma$, and (b) $\Delta = 173.91\gamma$ with $\delta = 1182.6\gamma$, $\bar{\Omega}_1 = 104.35\gamma$; (c) $\Delta = 131.58\gamma$, and (d) $\Delta = 219.30\gamma$, with $\delta = 2017.5\gamma$, $\bar{\Omega}_1 = 131.58\gamma$. Here $\bar{\Omega}_2 = 0.8\bar{\Omega}_1$ and $G = -43.48\gamma$ are the same as in Fig. 3. These parameters in (a)–(d) are based on the Rb and Cs atoms, respectively.

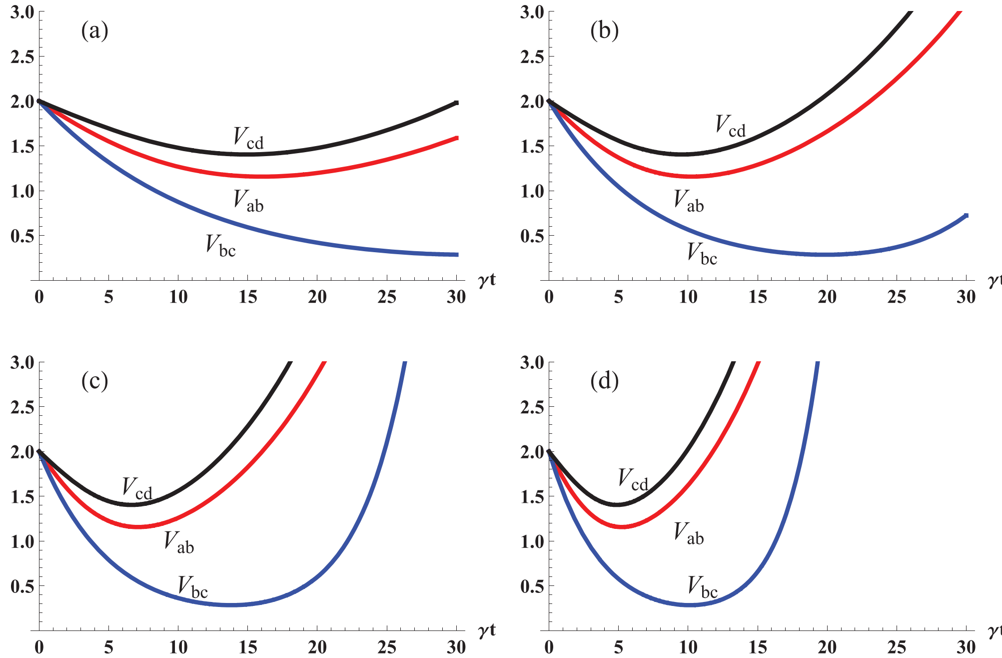

FIG. 5. (Color online) The VLF correlations for (a) $G = -43.48\gamma$, (b) $G = -54.83\gamma$, (c) $G = -65.80\gamma$, (d) $G = -76.75\gamma$ with $\delta = 2017.5\gamma$, $\bar{\Omega}_2 = 0.8\bar{\Omega}_1$, and $\Delta = \bar{\Omega}_1 = 131.58\gamma$.

interaction time range, which is shortened with the increase of $|G|$.

In general, the Rabi frequencies of the forward and backward pump fields are unsymmetrical. Here we consider the effect of the ratio ($\bar{\Omega}_2/\bar{\Omega}_1$) of forward and backward pump Rabi frequencies on the four-mode entanglement, which is plotted in Fig. 6. For $\bar{\Omega}_2/\bar{\Omega}_1 < 1$, the degree of inequality violation for $\tilde{b}$ and $\tilde{c}$ modes becomes stronger, and the curves of $V_{\tilde{c}\tilde{d}}$ and $V_{\tilde{a}\tilde{b}}$ tend to coincide, when $\bar{\Omega}_2/\bar{\Omega}_1$ increases to 1. For $\bar{\Omega}_2 = \bar{\Omega}_1$, the curves of $V_{\tilde{c}\tilde{d}}$ and $V_{\tilde{a}\tilde{b}}$ are completely overlapped. On the contrary, for $\bar{\Omega}_2/\bar{\Omega}_1 > 1$, the curves of $V_{\tilde{c}\tilde{d}}$ and $V_{\tilde{a}\tilde{b}}$ are separated and the degree of the inequality violation for $\tilde{b}$ and $\tilde{c}$ modes becomes weaker, when $\bar{\Omega}_2/\bar{\Omega}_1$ increases.

VI. CONCLUSIONS

We have examined the three-order nonlinear interaction scheme of Boyer *et al.* [9], which motivates us to propose a

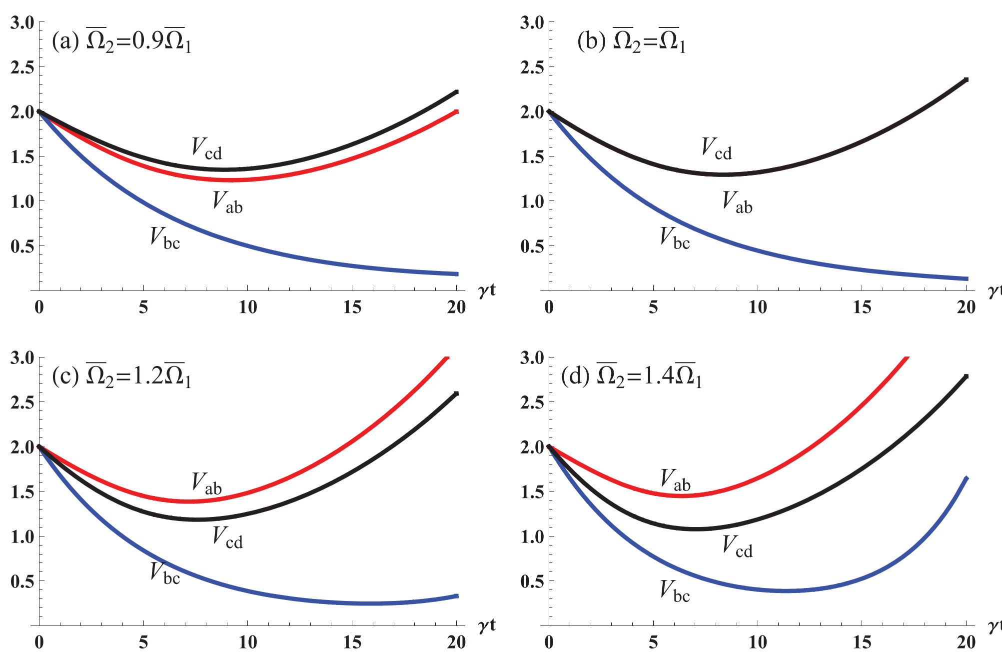

FIG. 6. (Color online) The VLF correlations for (a) $\bar{\Omega}_2 = 0.9\bar{\Omega}_1$, (b) $\bar{\Omega}_2 = 1.0\bar{\Omega}_1$, (c) $\bar{\Omega}_2 = 1.2\bar{\Omega}_1$, (d) $\bar{\Omega}_2 = 1.4\bar{\Omega}_1$, with $\delta = 2017.5\gamma$, $G = -54.83\gamma$, and $\Delta = \bar{\Omega}_1 = 131.58\gamma$.

theoretical scheme to generate the entanglement of four light beams in a three-level double-Λ atomic system derived by two counterpropagating far-detuned pump beams. The degree of entanglement between the four-mode is evaluated by using the sufficient inseparability criterion proposed by van Loock and Furusawa and the effective Hamiltonian. The dependence of entanglement on the detuning of pump beams, the ratio of counterpropagating pump field, as well as the coupling constant of four generated fields with the atomic system is also discussed. It is shown that a nonzero detuning is necessary for generating the observable entanglement under the current atomic configuration. These results supply a concrete method for preparation of multipartite entanglement in an atomic system experimentally.

ACKNOWLEDGMENTS

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APPENDIX A: LANGEVIN NOISE EFFECT IN A TWO-MODE CASE

In this Appendix, we consider the Langevin noise effect to the quantum entanglement (correlation). For simplicity, here we only consider the two-mode case, whose effective Hamiltonian can be given by taking $\bar{\Omega}_2 = 0$ ($\bar{\Omega}_1 = \Omega$) in Eq. (23), i.e.,

$$H_{\text{eff}} = \kappa_1 a^+ b^+ + \kappa_1 ab, \tag{A1}$$

where $\kappa_1 = \kappa\Omega^2 d_{31}^* d_{32}^*$ and κ is defined in Eq. (25) with $\bar{\Omega}_2 = 0$.

Taking the Langevin noise operators into account, the equations of atomic operators become

$$\begin{aligned}\dot{Q}_{31} &= i\Gamma_p Q_{31} + i(\Omega^* + d_{31}^* e^{i\delta t} b^+)(Q_{11} - Q_{33})\\ &\quad + i(\Omega^* + d_{32}^* e^{-i\delta t} a^+) Q_{21} + F_{31},\\ \dot{Q}_{23} &= i\Gamma_a^* Q_{23} - i(\Omega + d_{31} e^{-i\delta t} b) Q_{21}\\ &\quad - i(\Omega + d_{32} e^{i\delta t} a)(Q_{22} - Q_{33}) + F_{23},\\ \dot{Q}_{21} &= i\Gamma_{21} Q_{21} + i(\Omega + d_{32} e^{i\delta t} a) Q_{31}\\ &\quad - i(\Omega^* + d_{31}^* e^{i\delta t} b^+) Q_{23}, + F_{21},\end{aligned} \tag{A2}$$

$$\begin{aligned}\dot{Q}_{13} &= -i\Gamma_p^* Q_{13} - i(\Omega + d_{31} e^{-i\delta t} b)(Q_{11} - Q_{33})\\ &\quad - i(\Omega + d_{32} e^{i\delta t} a) Q_{12} + F_{13},\\ \dot{Q}_{32} &= -i\Gamma_a Q_{32} + i(\Omega^* + d_{31}^* e^{i\delta t} b^+) Q_{12}\\ &\quad + i(\Omega^* + d_{32}^* e^{-i\delta t} a^+)(Q_{22} - Q_{33}) + F_{32},\\ \dot{Q}_{12} &= -i\Gamma_{21}^* Q_{12} - i(\Omega^* + d_{32}^* e^{-i\delta t} a^+) Q_{13}\\ &\quad + i(\Omega + d_{31} e^{-i\delta t} b) Q_{32} + F_{12},\end{aligned} \tag{A3}$$

where F_{ij} are the Langevin operators, characterized by

$$\langle F_{ij}(t)\rangle = 0, \quad \text{and } \langle F_{ij}^+(t) F_{i'j'}(t')\rangle = 2D_{ij,i'j'}\delta(t - t'),$$

which defines the diffusion coefficients $D_{ij,i'j'}$.

For the contribution of the Langevin noise to the field, first order is enough [37,38]. The terms containing the field operators in Q_{31} and Q_{23} have already been included in the effective Hamiltonian, so that here we only consider the Langevin noise contribution (neglecting higher-order terms such as $F_{ij}a, F_{ij}b$ and $F_{ij}a^+, F_{ij}b^+$, and only keeping the contribution of F_{ij}). Dropping the terms containing field operators and substituting the zero-order solution for the atomic operators Q_{11}, Q_{22}, Q_{33} into Eqs. (A2) and (A3), we obtain the steady state solution [37,38],

$$\begin{aligned}0 &= i\Gamma_p Q_{31} + i\Omega^*\big(Q_{11}^{(0)} - Q_{33}^{(0)}\big) + i\Omega^* Q_{21} + F_{31},\\ 0 &= -i\Gamma_a Q_{32} + i\Omega^*\big(Q_{22}^{(0)} - Q_{33}^{(0)}\big) + i\Omega^* Q_{12} + F_{32},\\ 0 &= i\Gamma_{21} Q_{21} - i\Omega^* Q_{23} + i\Omega Q_{31} + F_{21}.\end{aligned} \tag{A4}$$

with

$$\begin{aligned}iF_{31} &= \Gamma_p Q_{31}^{(1)} + \Omega^* Q_{21}^{(1)},\\ iF_{23} &= \Gamma_a^* Q_{23}^{(1)} - \Omega Q_{21}^{(1)},\\ iF_{21} &= \Gamma_{21} Q_{21}^{(1)} - \Omega^* Q_{23}^{(1)} + \Omega Q_{31}^{(1)}.\end{aligned} \tag{A5}$$

Consequently, we obtain the Lagevin noise contribution to the field through

$$\begin{aligned}Q_{31} &\to F_a \equiv \alpha_{31}F_{31} + \alpha_{23}F_{23} + \alpha_{21}F_{21}, \quad \text{for } Q_{31},\\ Q_{23} &\to F_b \equiv \beta_{31}F_{31} + \beta_{23}F_{23} + \beta_{21}F_{21}, \quad \text{for } Q_{23},\end{aligned} \tag{A6}$$

where we have set

$$\begin{aligned}\alpha_{23} &= \frac{i}{T}(\Omega^*)^2, \quad \alpha_{21} = \frac{i}{T}(i\gamma + \delta + \Delta)\Omega^*,\\ \alpha_{31} &= \frac{i}{T}(-i\gamma\delta - \delta^2 - \delta\Delta + |\Omega|^2),\end{aligned} \tag{A7}$$

and

$$\begin{aligned}\beta_{31} &= \frac{i}{T}\Omega^2, \quad \beta_{21} = \frac{i}{T}(-i\gamma + \Delta)\Omega,\\ \beta_{23} &= \frac{i}{T}(-i\gamma\delta + \delta\Delta + |\Omega|^2),\end{aligned} \tag{A8}$$

with the definition of the denominator as

$$T = \delta(\gamma^2 - i\gamma\delta + \delta\Delta + \Delta^2) + (2i\gamma + \delta)|\Omega|^2. \tag{A9}$$

With considering the noise, the effective Hamiltonian, Eq. (A1), is modified as

$$\begin{aligned}H_{\text{eff}} &= \kappa_1 a^+ b^+ + \kappa_1 ab\\ &\quad + \{d_{31}e^{-i\delta t} b F_a + d_{32}^* e^{-i\delta t} a^+ F_b + \text{H.c.}\},\end{aligned} \tag{A10}$$

and the Heisenberg motion equations for optical fields are given by

$$\frac{d}{dt}a = -i\kappa_1 b^+ - i\tilde{F}_b, \quad \frac{d}{dt}b^+ = i\kappa_1 a + i\tilde{F}_a, \tag{A11}$$

where we have set $\tilde{F}_b = d_{32}^* e^{-i\delta t} F_b$, $\tilde{F}_a = d_{31} e^{-i\delta t} F_a$.

Here we introduce $ib = \tilde{b}, -ib^+ = \tilde{b}^+$, and write the quadrature amplitude and phase operators as $X_a = (a + a^+)/\sqrt{2}$, $P_a = (a - a^+)/(\sqrt{2}i)$, $X_b = (\tilde{b} + \tilde{b}^+)/\sqrt{2}$, $P_b = (\tilde{b} - \tilde{b}^+)/(\sqrt{2}i)$, and then the equations for the quadrature amplitude and phase are

$$\begin{aligned}\frac{d}{dt}X_a &= \kappa_1 X_b + i\frac{(\tilde{F}_b)^+ - \tilde{F}_b}{\sqrt{2}},\\ \frac{d}{dt}X_b &= \kappa_1 X_a + \frac{(\tilde{F}_a)^+ + \tilde{F}_a}{\sqrt{2}},\end{aligned} \tag{A12}$$

$$\frac{d}{dt}P_a = -\kappa_1 P_b - \frac{(\tilde{F}_b)^+ + \tilde{F}_b}{\sqrt{2}},$$
$$\frac{d}{dt}P_b = -\kappa_1 P_a + \frac{(\tilde{F}_a)^+ - \tilde{F}_a}{\sqrt{2}i}. \quad \text{(A13)}$$

In order to solve Eqs. (A12) and (A13), we rewrite Eq. (A12) as

$$\frac{d}{dt}u(t) = Ju(t) + F(t), \quad \text{(A14)}$$

where

$$u(t) = \begin{bmatrix} X_a(t) \\ X_b(t) \end{bmatrix}, \quad J = \begin{pmatrix} 0 & \kappa_1 \\ \kappa_1 & 0 \end{pmatrix},$$
$$F(t) = \frac{1}{\sqrt{2}}\begin{bmatrix} i(\tilde{F}_b)^+ - i\tilde{F}_b \\ \tilde{F}_b + (\tilde{F}_a)^+ \end{bmatrix} \equiv \begin{bmatrix} f_a(t) \\ f_b(t) \end{bmatrix}, \quad \text{(A15)}$$

The solution of Eq. (A15) is

$$u(t) = e^{Jt}\left[u(0) + \int_0^t e^{-Jt'}F(t')dt'\right]. \quad \text{(A16)}$$

In order to solve Eq. (A14), we can diagonalize the matrix J as

$$J = UJ_aU^{-1}, \quad \text{(A17)}$$

with

$$J_a = \begin{pmatrix} -\kappa_1 & 0 \\ 0 & \kappa_1 \end{pmatrix},$$
$$U = \begin{pmatrix} -1 & 1 \\ 1 & 1 \end{pmatrix},$$
$$U^{-1} = \frac{1}{2}\begin{pmatrix} -1 & 1 \\ 1 & 1 \end{pmatrix}. \quad \text{(A18)}$$

Consequently, we have

$$X_a(t) = \cosh[\kappa_1 t]X_{a0} + \sinh[\kappa_1 t]X_{b0} + \int_0^t \{\cosh[\kappa_1(t-t')]f_a(t') + \sinh[\kappa_1(t-t')]f_b(t')\}dt', \quad \text{(A19)}$$

$$X_b(t) = \sinh[\kappa_1 t]X_{a0} + \cosh[\kappa_1 t]X_{b0} + \int_0^t \{\sinh[\kappa_1(t-t')]f_a(t') + \cosh[\kappa_1(t-t')]f_b(t')\}dt', \quad \text{(A20)}$$

where X_{a0} and X_{b0} are the initial quadrature amplitude. From Eqs. (A19) and (A20), we obtain

$$X_a(t) - X_b(t) = (X_{a0} - X_{b0})\,e^{-\kappa_1 t} + \int_0^t \{[f_a(t') - f_b(t')]e^{-\kappa_1(t-t')}\}dt'. \quad \text{(A21)}$$

In a similar way, we obtain

$$P_a(t) = \cosh[\kappa_1 t]\,P_{a0} - \sinh[\kappa_1 t]\,P_{b0} + \int_0^t \{\cosh[\kappa_1(t-t')]f'_a(t') - \sinh[\kappa_1(t-t')]f'_b(t')\}dt', \quad \text{(A22)}$$

and

$$P_b(t) = -\sinh[\kappa_1 t]\,P_{a0} + \cosh[\kappa_1 t]\,P_{b0} + \int_0^t \{-\sinh[\kappa_1(t-t')]f'_a(t') + \cosh[\kappa_1(t-t')]f'_b(t')\}dt', \quad \text{(A23)}$$

as well as

$$P_a(t) + P_b(t) = (P_{a0} + P_{b0})\,e^{-\kappa_1 t} + \int_0^t \{[f'_b(t') + f'_a(t')]e^{-\kappa_1(t-t')}\}dt', \quad \text{(A24)}$$

where P_{a0} and P_{b0} are the initial quadrature phase and

$$f'_a(t) = -\frac{1}{\sqrt{2}}[(\tilde{F}_b)^+ + \tilde{F}_b], \quad f'_b(t) = \frac{1}{\sqrt{2}i}[(\tilde{F}_b)^+ - \tilde{F}_b]. \quad \text{(A25)}$$

For the initial input vacuum state, we have $\langle X_{a,b}(0)\rangle = \langle P_{a,b}(0)\rangle = 0$, $\langle F_{a,b}(t')\rangle = 0$, and $\langle X_o(0)X_{o'}(0)\rangle = \langle P_o(0)P_{o'}(0)\rangle = \delta_{oo'}/2$, so that we have $\langle X_a(t) - X_b(t)\rangle = \langle P_b(t) + P_a(t)\rangle = 0$. Then we obtain

$$\langle[X_a(t) - X_b(t)]^2\rangle = \langle\{A_1(t)\}^2\rangle + \int_0^t \langle B_1(t,t')B_1(t,t'')\rangle dt'dt'',$$
$$\langle[P_a(t) + P_b(t)]^2\rangle = \langle\{A_2(t)\}^2\rangle + \int_0^t \langle B_2(t,t')B_2(t,t'')\rangle dt'dt'', \quad \text{(A26)}$$

where we have set

$$A_1(t) = e^{-\kappa_1 t}\,(X_{a0} - X_{b0}),$$
$$A_2(t) = e^{-\kappa_1 t}\,(P_{a0} + P_{b0}),$$
$$B_1(t,t') = e^{-\kappa_1(t-t')}[f_a(t') - f_b(t')],$$
$$B_2(t,t') = e^{-\kappa_1(t-t')}[f'_b(t') + f'_a(t')], \quad \text{(A27)}$$

and assumed $\langle X_{a,c}(0)f_{a,c}(t')\rangle = \langle P_{a,c}(0)f_{a,c}(t')\rangle = 0$. Finally we can express the correlation as

$$I = \frac{1}{2}\{\langle[X_a(t) - X_b(t)]^2\rangle + \langle[P_a(t) + P_b(t)]^2\rangle\}$$
$$= e^{-2\kappa_1 t} + \frac{1}{2}\int_0^t \langle B_1(t,t')B_1(t,t'')\rangle dt'dt''$$
$$+ \frac{1}{2}\int_0^t \langle B_2(t,t')B_2(t,t'')\rangle dt\prime dt''. \quad \text{(A28)}$$

To derive the second and third terms in Eq. (A28), we need to calculate some correlation items, such as $\langle f_o(t')f_{o'}(t'')\rangle$ and $\langle f'_o(t')f'_o(t'')\rangle$ $(o = a,b)$. Using Eqs. (A15) and (A25), and denoting $d_{32}^*\beta_{ij} \to \beta_{ij}$, $d_{31}\alpha_{ij} \to \alpha_{ij}$, we obtain

$$\begin{aligned}\langle f_a(t')f_a(t'')\rangle &= -\frac{1}{2}\{e^{i2\delta t'}D_{b^+,b^+} - D_{b^+,b} - D_{b,b^+} + e^{-i2\delta t'}D_{b,b}\}\delta(t'-t''),\\ \langle f_a(t')f_b(t'')\rangle &= i\frac{1}{2}\{e^{i2\delta t'}D_{b^+,a^+} + D_{b^+,a} - D_{b,a^+} - e^{-i2\delta t'}D_{b,a}\}\delta(t'-t''),\end{aligned} \tag{A29}$$

$$\begin{aligned}\langle f_b(t')f_a(t'')\rangle &= \frac{i}{2}\langle D_{a,b^+} - D_{a,b}e^{-i2\delta t} + D_{a^+,b^+}e^{i2\delta t} - D_{a^+,b}\rangle\delta(t'-t''),\\ \langle f_b(t')f_b(t'')\rangle &= \frac{1}{2}\{e^{i2\delta t'}D_{a^+,a^+} - D_{a^+,a} - D_{a,a^+} + e^{-i2\delta t'}D_{a,a}\}\delta(t'-t''),\end{aligned} \tag{A30}$$

and

$$\begin{aligned}\langle f_a'(t')f_a'(t'')\rangle &= \frac{1}{2}\{e^{i2\delta t'}D_{b^+,b^+} + D_{b^+,b} + D_{b,b^+} + e^{-i2\delta t'}D_{b,b}\}\delta(t'-t''),\\ \langle f_a'(t')f_b'(t'')\rangle &= \frac{i}{2}\{e^{i2\delta t'}D_{b^+,a^+} - D_{b^+,a} + D_{b,a^+} - e^{-i2\delta t'}D_{b,a}\}\delta(t'-t''),\\ \langle f_b'(t')f_a'(t'')\rangle &= \frac{i}{2}\{e^{i2\delta t'}D_{a^+,b^+} + D_{a^+,b} - D_{a,b^+} - e^{-i2\delta t'}D_{a,b}\}\delta(t'-t''),\\ \langle f_b'(t')f_b'(t'')\rangle &= -\frac{1}{2}\{e^{i2\delta t'}D_{a^+,a^+} - D_{a^+,a} - D_{a,a^+} + e^{-i2\delta t'}D_{a,a}\}\delta(t'-t'') = -\langle f_b(t')f_b(t'')\rangle,\end{aligned} \tag{A31}$$

where $D_{oo'}$ is defined by $D_{o,o'} = \langle F_oF_{o'}\rangle$, $D_{o^+,o'} = \langle (F_o)^+F_{o'}\rangle$, and $D_{o,o'^+} = \langle F_o(F_{o'})^+\rangle$, whose expressions will be determined later.

Substituting Eqs. (A29)–(A31) into Eq. (A28) we finally obtain the variance

$$\begin{aligned}I = e^{-2\kappa_1 t} + \frac{1-e^{-2\kappa_1 t}}{4\kappa_1}\{D_{b^+,b} + D_{b,b^+}\\ + i(D_{a^+,b} - D_{b^+,a} + D_{b,a^+} - D_{a,b^+})\},\end{aligned} \tag{A32}$$

where $D_{b,b^+}\delta(t-t') = \langle F_b(t)F_{b^+}(t')\rangle$, $D_{b^+,b}\delta(t-t') = \langle [F_b(t)]^+F_b(t')\rangle$, $D_{b,a^+}\delta(t-t') = \langle F_b(t)[F_a(t')]^+\rangle$, $D_{a^+,b}\delta(t-t') = \langle [F_a(t)]^+F_b(t')\rangle$, and $D_{a^+,b} = (D_{b^+,a})^*$, $D_{a,b^+} = (D_{b,a^+})^*$. In Eq. (A32), the first item $e^{-2\kappa_1 t}$ corresponds to the entanglement without the Langevin noise, and the second item is the contribution of the Langevin noise (the effect of atom spontaneous emission) to the entanglement.

Using the generalized Einstein relation [37,38] and the Heisenberg-Langevin equations (A2) and (A3), the Langevin diffusion coefficients for atomic operators can be obtained. Here we are interested in the diffusion coefficients due to the Langevin noise operators, F_{31}, F_{23}, F_{21}, and their adjoint F_{13}, F_{32}, F_{12},

$$\langle [K(t)][K(t)]^T\rangle = \gamma\begin{pmatrix} 0 & \langle Q_{33}\rangle & 0 & 0 & 0 & 0\\ \langle Q_{33}\rangle & 0 & 0 & 0 & 0 & 0\\ 0 & 0 & 0 & 2\langle Q_{11}\rangle + \langle Q_{33}\rangle & 0 & 2\langle Q_{12}\rangle\\ 0 & 0 & 0 & 0 & 0 & 0\\ 0 & 0 & 0 & 2\langle Q_{21}\rangle & 0 & 2\langle Q_{22}\rangle + \langle Q_{33}\rangle\\ 0 & 0 & 0 & 0 & 0 & 0\end{pmatrix}\cdot\delta(t-t'), \tag{A33}$$

where $[K(t)] \equiv [F_{12}(t)F_{21}(t)F_{13}(t)F_{31}(t)F_{23}(t)F_{32}(t)]^T$. Using Eqs. (A33) and (A6), we can finally obtain (here we recover $\beta_{ij} \to d_{32}^*\beta_{ij}$, $\alpha_{ij} \to d_{31}\alpha_{ij}$)

$$\begin{aligned}D_{b^+,b} &= \gamma|d_{32}|^2\{2|\beta_{31}|^2\langle Q_{11}\rangle + (|\beta_{21}|^2 + |\beta_{31}|^2)\langle Q_{33}\rangle\},\\ D_{b,b^+} &= \gamma|d_{32}|^2\{2|\beta_{23}|^2\langle Q_{22}\rangle + (|\beta_{21}|^2 + |\beta_{23}|^2)\langle Q_{33}\rangle\},\\ D_{b^+,a} &= \gamma d_{31}d_{32}\{2\beta_{31}^*\alpha_{31}\langle Q_{11}\rangle + (\beta_{21}^*\alpha_{21} + \beta_{31}^*\alpha_{31})\langle Q_{33}\rangle\},\\ D_{b,a^+} &= \gamma d_{32}^*d_{31}^*\{2\beta_{23}\alpha_{23}^*\langle Q_{22}\rangle + (\beta_{21}\alpha_{21}^* + \beta_{23}\alpha_{23}^*)\langle Q_{33}\rangle\}.\end{aligned} \tag{A34}$$

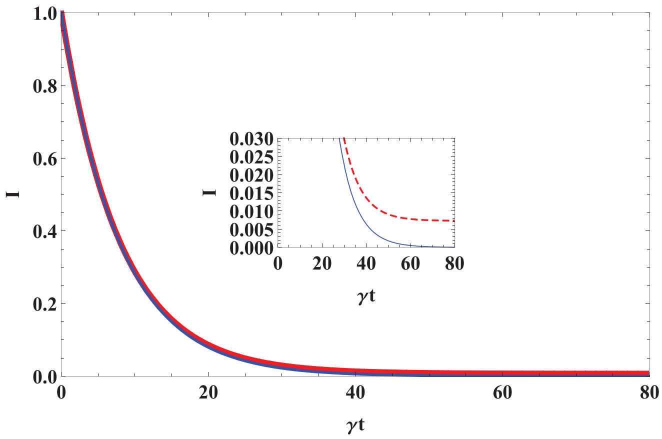

FIG. 7. (Color online) The entanglement I as the function of time. Here we have chosen the parameters $G = -250$ MHz, $\gamma = 4.56$ MHz, and $\Omega = 0.4$ GHz, $\delta = 9$ GHz, $\Delta = 0.8$ GHz.

Here we have assumed $\gamma_1 = \gamma_2 = \gamma$. It is easy to see that $D_{i,j}$ in Eq. (A34) satisfy the phase-matching condition. Note $\langle Q_{ii}\rangle (i = 1,2,3)$ are replaced with their zero-order values of $Q_{ii}^{(0)}$ in Eqs. (16)–(22). The special expressions for $D_{i,j}$ can be derived by substituting Eqs. (A5) and (A6) into Eq. (A34). In Fig. 7, we plot the entanglement I in Eq. (A32) as the function of time (see the red curve) where we also plot the entanglement without considering the Langevin noise by dropping the second term in right-hand side of Eq. (A32). It is clear that the effect of Langevin noise on the entanglement I is very small; thus we can neglect it in our further calculations.

APPENDIX B: ZERO-ORDER SOLUTION

Taking the zero-order steady solution of Eqs. (10)–(13), we can derive

$$\begin{aligned}
1 &= Q_{11}^{(0)} + Q_{22}^{(0)} + Q_{33}^{(0)},\\
0 &= \gamma_1 Q_{33}^{(0)} - i\Omega^* Q_{13}^{(0)} + i\Omega Q_{31}^{(0)},\\
0 &= \gamma_2 Q_{33}^{(0)} - i\Omega^* Q_{23}^{(0)} + i\Omega Q_{32}^{(0)},\\
0 &= \Gamma_p Q_{31}^{(0)} + \Omega^*\big(Q_{11}^{(0)} - Q_{33}^{(0)}\big) + \Omega^* Q_{21}^{(0)},\\
0 &= \Gamma_a Q_{32}^{(0)} - \Omega^*\big(Q_{22}^{(0)} - Q_{33}^{(0)}\big) - \Omega^* Q_{12}^{(0)},\\
0 &= \Gamma_{21} Q_{21}^{(0)} - \Omega^* Q_{23}^{(0)} + \Omega Q_{31}^{(0)},\\
0 &= \Gamma_p^* Q_{13}^{(0)} + \Omega\big(Q_{11}^{(0)} - Q_{33}^{(0)}\big) + \Omega Q_{12}^{(0)},\\
0 &= \Gamma_a^* Q_{23}^{(0)} - \Omega\big(Q_{22}^{(0)} - Q_{33}^{(0)}\big) - \Omega Q_{21}^{(0)},\\
0 &= \Gamma_{21}^* Q_{12}^{(0)} - \Omega Q_{32}^{(0)} + \Omega^* Q_{13}^{(0)}.
\end{aligned} \tag{B1}$$

The solution of Eq. (B1) can be written in the matrix form

$$\begin{pmatrix}
1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & \gamma_1 & i\Omega & -i\Omega^* & 0 & 0 & 0 & 0\\
0 & 0 & \gamma_2 & 0 & 0 & i\Omega & -i\Omega^* & 0 & 0\\
-\Omega^* & 0 & \Omega^* & -\Gamma_p & 0 & 0 & 0 & -\Omega^* & 0\\
0 & -\Omega^* & \Omega^* & 0 & 0 & \Gamma_a & 0 & 0 & -\Omega^*\\
0 & 0 & 0 & -\Omega & 0 & 0 & \Omega^* & -\Gamma_{21} & 0\\
-\Omega & 0 & \Omega & 0 & -\Gamma_p^* & 0 & 0 & 0 & -\Omega\\
0 & -\Omega & \Omega & 0 & 0 & 0 & \Gamma_a^* & -\Omega & 0\\
0 & 0 & 0 & 0 & -\Omega^* & \Omega & 0 & 0 & -\Gamma_{21}^*
\end{pmatrix}^{-1}
\begin{pmatrix}1\\0\\0\\0\\0\\0\\0\\0\\0\end{pmatrix}
=
\begin{pmatrix}Q_{11}^{(0)}\\Q_{22}^{(0)}\\Q_{33}^{(0)}\\Q_{31}^{(0)}\\Q_{13}^{(0)}\\Q_{32}^{(0)}\\Q_{23}^{(0)}\\Q_{21}^{(0)}\\Q_{12}^{(0)}\end{pmatrix}. \tag{B2}$$

Taking $\gamma_1 = \gamma_2 = \gamma_{32} = \gamma_{31} = \gamma$ and $\gamma_{21} = 0$, then solving Eq. (B2), one can obtain the zero-order steady solutions for $Q_{jk}^{(0)}$ $(j,k = 1,2,3)$ shown in Eqs. (16)–(21).

APPENDIX C: FIRST-ORDER SOLUTION

In a similar way, the first-order steady equations are given by

$$\begin{aligned}
0 &= Q_{11}^{(1)} + Q_{22}^{(1)} + Q_{33}^{(1)},\\
m_1 &= i\gamma Q_{33}^{(1)} + \Omega^* Q_{13}^{(1)} - \Omega Q_{31}^{(1)},\\
m_2 &= i\gamma Q_{33}^{(1)} + \Omega^* Q_{23}^{(1)} - \Omega Q_{32}^{(1)},\\
m_3 &= \Gamma_p Q_{31}^{(1)} + \Omega^*\big(Q_{11}^{(1)} - Q_{33}^{(1)}\big) + \Omega^* Q_{21}^{(1)},\\
m_4 &= \Gamma_a Q_{32}^{(1)} - \Omega^* Q_{12}^{(1)} - \Omega^*\big(Q_{22}^{(1)} - Q_{33}^{(1)}\big),\\
m_5 &= \Gamma_{21} Q_{21}^{(1)} - \Omega^* Q_{23}^{(1)} + \Omega Q_{31}^{(1)},
\end{aligned} \tag{C1}$$

where we have set

$$\begin{aligned}
m_1 &= e^{-i\delta t} B Q_{31}^{(0)} - e^{i\delta t} B^+ Q_{13}^{(0)},\\
m_2 &= e^{i\delta t} A Q_{32}^{(0)} - e^{-i\delta t} A^+ Q_{23}^{(0)},\\
m_3 &= -e^{-i\delta t} A^+ Q_{21}^{(0)} - e^{i\delta t} B^+\big(Q_{11}^{(0)} - Q_{33}^{(0)}\big),\\
m_4 &= e^{i\delta t} B^+ Q_{12}^{(0)} + e^{-i\delta t} A^+\big(Q_{22}^{(0)} - Q_{33}^{(0)}\big),\\
m_5 &= e^{i\delta t} B^+ Q_{23}^{(0)} - e^{i\delta t} A Q_{31}^{(0)}.
\end{aligned} \tag{C2}$$

The solutions of $Q_{jk}^{(1)}$ $(j,k=1,2,3)$ are given by

$$\begin{pmatrix} Q_{11}^{(1)} \\ Q_{22}^{(1)} \\ Q_{33}^{(1)} \\ Q_{13}^{(1)} \\ Q_{31}^{(1)} \\ Q_{23}^{(1)} \\ Q_{32}^{(1)} \\ Q_{21}^{(1)} \\ Q_{12}^{(1)} \end{pmatrix} = U_1^{-1} \begin{pmatrix} 0 \\ m_1 \\ m_2 \\ m_3 \\ m_4 \\ m_5 \\ m_3^* \\ m_4^* \\ m_5^* \end{pmatrix}, \tag{C3}$$

where

$$U_1 = \begin{pmatrix} 1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & \Omega^* & -\Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & 0 & 0 & \Omega^* & -\Omega & 0 & 0 \\ \Omega^* & 0 & -\Omega^* & 0 & (-\Delta+i\gamma) & 0 & 0 & \Omega^* & 0 \\ 0 & -\Omega^* & \Omega^* & 0 & 0 & 0 & (\delta+\Delta-i\gamma) & 0 & -\Omega^* \\ 0 & 0 & 0 & 0 & \Omega & -\Omega^* & 0 & \delta & 0 \\ \Omega & 0 & -\Omega & (-\Delta-i\gamma) & 0 & 0 & 0 & 0 & \Omega \\ 0 & -\Omega & \Omega & 0 & 0 & (\delta+\Delta+i\gamma) & 0 & -\Omega & 0 \\ 0 & 0 & 0 & \Omega^* & 0 & 0 & -\Omega & 0 & \delta \end{pmatrix}, \tag{C4}$$

and

$$N = \det U_1 = 2\gamma^2|\Omega|^2\{[2\Delta^2 + (\delta+2\Delta)\delta + 2\gamma^2]\delta^2 + 2\delta^2|\Omega|^2 + 8|\Omega|^4\}. \tag{C5}$$

We write $Q_{31}^{(1)}$ and $Q_{23}^{(1)}$ as

$$Q_{31}^{(1)} = \frac{D_1}{N}, \quad Q_{23}^{(1)} = \frac{E_1}{N}, \tag{C6}$$

where

$$D_1 = \det \begin{pmatrix} 1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & \Omega^* & m_1 & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & 0 & m_2 & \Omega^* & -\Omega & 0 & 0 \\ \Omega^* & 0 & -\Omega^* & 0 & m_3 & 0 & 0 & \Omega^* & 0 \\ 0 & -\Omega^* & \Omega^* & 0 & m_4 & 0 & (\delta+\Delta-i\gamma) & 0 & -\Omega^* \\ 0 & 0 & 0 & 0 & m_5 & -\Omega^* & 0 & \delta & 0 \\ \Omega & 0 & -\Omega & (-\Delta-i\gamma) & m_3^* & 0 & 0 & 0 & \Omega \\ 0 & -\Omega & \Omega & 0 & m_4^* & (\delta+\Delta+i\gamma) & 0 & -\Omega & 0 \\ 0 & 0 & 0 & \Omega^* & m_5^* & 0 & -\Omega & 0 & \delta \end{pmatrix}, \tag{C7}$$

$$E_1 = \det \begin{pmatrix} 1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & \Omega^* & -\Omega & m_1 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & 0 & 0 & m_2 & -\Omega & 0 & 0 \\ \Omega^* & 0 & -\Omega^* & 0 & (-\Delta+i\gamma) & m_3 & 0 & \Omega^* & 0 \\ 0 & -\Omega^* & \Omega^* & 0 & 0 & m_4 & (\delta+\Delta-i\gamma) & 0 & -\Omega^* \\ 0 & 0 & 0 & 0 & \Omega & m_5 & 0 & \delta & 0 \\ \Omega & 0 & -\Omega & (-\Delta-i\gamma) & 0 & m_3^* & 0 & 0 & \Omega \\ 0 & -\Omega & \Omega & 0 & 0 & m_4^* & 0 & -\Omega & 0 \\ 0 & 0 & 0 & \Omega^* & 0 & m_5^* & -\Omega & 0 & \delta \end{pmatrix}. \tag{C8}$$

Keeping the terms including only single operator and satisfying the phase-matching condition, we write

$$D_1 \to \gamma A e^{it\delta}(\Omega^*)^2\mu_a, \quad E_1 \to -\Omega^2\gamma B^+ e^{it\delta}\mu_c, \tag{C9}$$

where

$$\mu_a = -iQ_{12}^{(0)}\{\delta^2[\Delta^2+\gamma^2+(\delta+2\Delta)\delta]+2\delta(\delta-2\Delta)|\Omega|^2+4|\Omega|^4\}$$
$$+\left(Q_{22}^{(0)}-Q_{33}^{(0)}\right)[i\delta^2(\Delta^2+\gamma^2)-(\gamma-i\Delta)\delta^3+2\delta(2\gamma+i\delta)|\Omega|^2-4i|\Omega|^4]$$
$$+\frac{Q_{32}^{(0)}}{\Omega^*}\{\delta^2(\gamma-i\Delta)(\Delta^2+2\Delta\delta+\gamma^2+\delta^2)+(2i\Delta^2\delta-2\Delta\gamma\delta+i\Delta\delta^2-3\gamma\delta^2-i\delta^3)|\Omega|^2+4(\gamma+2i\delta)|\Omega|^4\}$$
$$-\Omega Q_{31}^{(0)}[2i\Delta^2\delta+2\Delta\gamma\delta+3i\Delta\delta^2+4i\gamma^2\delta-\gamma\delta^2+i\delta^3+4(-i\Delta+2\gamma+i\delta)|\Omega|^2], \quad \text{(C10)}$$

and

$$\mu_b = \left(Q_{11}^{(0)}-Q_{33}^{(0)}\right)\{\delta^2[i(\Delta^2+\gamma^2)-(\gamma-i\Delta)\delta]+2\delta(2\gamma+i\delta)|\Omega|^2-4i|\Omega|^4\}-iQ_{12}^{(0)}[(\Delta^2+\gamma^2)\delta^2+2(3\delta+2\Delta)\delta|\Omega|^2+4|\Omega|^4]$$
$$-\frac{Q_{13}^{(0)}}{\Omega}[\delta^2(\Delta^2+\gamma^2)(\gamma+i\Delta+i\delta)+\delta(2i\Delta^2+2\Delta\gamma+3i\Delta\delta-\gamma\delta)|\Omega|^2+4(\gamma+2i\delta)|\Omega|^4]$$
$$+Q_{23}^{(0)}\Omega^*[\delta(2i\Delta^2-2\Delta\gamma+i\Delta\delta+4i\gamma^2-3\gamma\delta)+4(2\gamma+2i\delta+i\Delta)|\Omega|^2]. \quad \text{(C11)}$$

Substituting the zero solutions Eqs. (16)–(21) into Eqs. (C10) and (C11), we have

$$\mu_a = -\frac{2\gamma\delta^2}{G_2}[\delta^2(-\Delta-i\gamma)(\Delta^2+2\Delta\delta+\gamma^2+\delta^2)-\delta(4\gamma^2-2i\gamma\delta+4i\Delta\gamma+3\delta^2+4\Delta\delta)|\Omega|^2+2(2i\gamma+6\Delta+\delta)|\Omega|^4],$$
$$\mu_b = \frac{2\gamma\delta^2}{G_2}[\delta^2(\Delta^2+\gamma^2)(i\gamma-\Delta-\delta)+\delta(4\gamma^2+2i\gamma\delta-4i\Delta\gamma-\delta^2-4\Delta\delta)|\Omega|^2-2(2i\gamma-6\Delta-5\delta)|\Omega|^4], \quad \text{(C12)}$$

where

$$G_2 = \delta^2(\delta^2+2\Delta\delta+2\Delta^2+2\gamma^2)+2\delta^2|\Omega|^2+8|\Omega|^4. \quad \text{(C13)}$$

Finally, Eq. (C9) becomes

$$D_1 \to \gamma Ae^{it\delta}(\Omega^*)^2\mu_a = -Ae^{(it\delta)}(\Omega^*)^2\frac{2\gamma^2\delta^2}{G_2}\{\delta^2(-\Delta-i\gamma)(\Delta^2+2\Delta\delta+\gamma^2+\delta^2)$$
$$-\delta(4\gamma^2-2i\gamma\delta-4i\Delta\gamma+3\delta^2+4\Delta\delta)|\Omega|^2+2(2i\gamma+6\Delta+\delta)|\Omega|^4\}, \quad \text{(C14)}$$

$$E_1 \to -\Omega^2\gamma B^+e^{it\delta}\mu_b = -B^+e^{(it\delta)}\Omega^2\frac{2\gamma^2\delta^2}{G_2}\{\delta^2(\Delta^2+\gamma^2)(-\Delta+i\gamma-\delta)$$
$$+\delta(4\gamma^2+2i\gamma\delta+4i\Delta\gamma-\delta^2-4\Delta\delta)|\Omega|^2-2(-6\Delta+2i\gamma-5\delta)|\Omega|^4\}. \quad \text{(C15)}$$

The correlation items resulting from the first-order steady solutions, $Q_{31}^{(1)}$ and $Q_{23}^{(1)}$, are

$$Be^{-i\delta t}Q_{31}^{(1)}+A^+e^{-i\delta t}Q_{23}^{(1)}+\text{H.c.} = Be^{-i\delta t}\frac{D_1}{N}+A^+e^{-i\delta t}\frac{E_1}{N}+\text{H.c.} \to \Lambda_aAB+\Lambda_bA^+B^++\text{H.c.}$$
$$= (\Lambda_a+\Lambda_b^*)AB+(\Lambda_a^*+\Lambda_b)A^+B^+ = \Lambda AB+\Lambda^*A^+B^+, \quad \text{(C16)}$$

where we have set

$$\Lambda_a = \frac{2\delta^2\gamma^2}{-NG_2}(\Omega^*)^2[\delta^2(-\Delta-i\gamma)(\Delta^2+2\Delta\delta+\gamma^2+\delta^2)-\delta(4\gamma^2-2i\gamma\delta-4i\Delta\gamma+3\delta^2+4\Delta\delta)|\Omega|^2+2(2i\gamma+6\Delta+\delta)|\Omega|^4],$$
$$\Lambda_b = \frac{2\delta^2\gamma^2}{-NG_2}\Omega^2[\delta^2(\Delta^2+\gamma^2)(-\Delta+i\gamma-\delta)+\delta(4\gamma^2+2i\gamma\delta+4i\Delta\gamma-\delta^2-4\Delta\delta)|\Omega|^2-2(-6\Delta+2i\gamma-5\delta)|\Omega|^4], \quad \text{(C17)}$$

$$\Lambda = \Lambda_a+\Lambda_b^* = \frac{2\delta^2\gamma\gamma}{NG_2}(\Omega^*)^2\{i\delta^2[2\gamma^2-i\gamma\delta+(\delta+2\Delta)(\delta+\Delta)](\gamma-i\Delta)+4\delta^2(\delta+2\Delta)|\Omega|^2-4(2i\gamma+6\Delta+3\delta)|\Omega|^4\}. \quad \text{(C18)}$$

Note $N = 2\gamma^2|\Omega|^2G_2$, so

$$\Lambda = \frac{\delta^2}{(G_2)^2}\left\{i\delta^2[2\gamma^2-i\gamma\delta+(\delta+2\Delta)(\delta+\Delta)](\gamma-i\Delta)\frac{\Omega^*}{\Omega}+4\delta^2(\delta+2\Delta)(\Omega^*)^2-4(2i\gamma+6\Delta+3\delta)|\Omega|^2(\Omega^*)^2\right\}, \quad \text{(C19)}$$

which is just the contribution to correlation from the first-order solutions.

Next, we consider the second-order solutions. From Eqs. (12) and (13), the matrix form of second-order steady equations is given by

$$\begin{pmatrix} Q_{11}^{(2)} \\ Q_{22}^{(2)} \\ Q_{33}^{(2)} \\ Q_{13}^{(2)} \\ Q_{31}^{(2)} \\ Q_{23}^{(2)} \\ Q_{32}^{(2)} \\ Q_{21}^{(2)} \\ Q_{12}^{(2)} \end{pmatrix} = U_1^{-1} \begin{pmatrix} 0 \\ n_1 \\ n_2 \\ n_3 \\ n_4 \\ n_5 \\ n_3^* \\ n_4^* \\ n_5^* \end{pmatrix}, \tag{C20}$$

where U_1 is defined in Eq. (C4), and

$$\begin{aligned} n_1 &= e^{-i\delta t} B Q_{31}^{(1)} - e^{i\delta t} B^+ Q_{13}^{(1)}, \\ n_2 &= e^{i\delta t} A Q_{32}^{(1)} - e^{-i\delta t} A^+ Q_{23}^{(1)}, \\ n_3 &= -e^{-i\delta t} A^+ Q_{21}^{(1)} - e^{i\delta t} B^+ \big(Q_{11}^{(1)} - Q_{33}^{(1)}\big), \\ n_4 &= e^{i\delta t} B^+ Q_{12}^{(1)} + e^{-i\delta t} A^+ \big(Q_{22}^{(1)} - Q_{33}^{(1)}\big), \\ n_5 &= e^{i\delta t} B^+ Q_{23}^{(1)} - e^{i\delta t} A Q_{31}^{(1)}. \end{aligned} \tag{C21}$$

The solutions of $Q_{31}^{(2)}$ and $Q_{23}^{(2)}$ are written as

$$Q_{31}^{(2)} = \frac{H_{31}^{(2)}}{N}, \quad Q_{23}^{(2)} = \frac{H_{23}^{(2)}}{N}, \tag{C22}$$

where

$$H_{31}^{(2)} = \det \begin{pmatrix} 1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & \Omega^* & n_1 & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & 0 & n_2 & \Omega^* & -\Omega & 0 & 0 \\ \Omega^* & 0 & -\Omega^* & 0 & n_3 & 0 & 0 & \Omega^* & 0 \\ 0 & -\Omega^* & \Omega^* & 0 & n_4 & 0 & (\delta+\Delta-i\gamma) & 0 & -\Omega^* \\ 0 & 0 & 0 & 0 & n_5 & -\Omega^* & 0 & \delta & 0 \\ \Omega & 0 & -\Omega & (-\Delta-i\gamma) & n_3^* & 0 & 0 & 0 & \Omega \\ 0 & -\Omega & \Omega & 0 & n_4^* & (\delta+\Delta+i\gamma) & 0 & -\Omega & 0 \\ 0 & 0 & 0 & \Omega^* & n_5^* & 0 & -\Omega & 0 & \delta \end{pmatrix}, \tag{C23}$$

$$H_{23}^{(2)} = \det \begin{pmatrix} 1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & \Omega^* & -\Omega & n_1 & 0 & 0 & 0 \\ 0 & 0 & i\gamma & 0 & 0 & n_2 & -\Omega & 0 & 0 \\ \Omega^* & 0 & -\Omega^* & 0 & (-\Delta+i\gamma) & n_3 & 0 & \Omega^* & 0 \\ 0 & -\Omega^* & \Omega^* & 0 & 0 & n_4 & (\delta+\Delta-i\gamma) & 0 & -\Omega^* \\ 0 & 0 & 0 & 0 & \Omega & n_5 & 0 & \delta & 0 \\ \Omega & 0 & -\Omega & (-\Delta-i\gamma) & 0 & n_3^* & 0 & 0 & \Omega \\ 0 & -\Omega & \Omega & 0 & 0 & n_4^* & 0 & -\Omega & 0 \\ 0 & 0 & 0 & \Omega^* & 0 & n_5^* & -\Omega & 0 & \delta \end{pmatrix}. \tag{C24}$$

We are only interested in these terms AB and A^+B^+ involved in $H_{31}^{(2)}$ and $H_{23}^{(2)}$, because they might satisfy the phase-matching condition. Substituting the zero-order and the first-order solutions into Eqs. (C23) and (C24) and only extracting these terms only including AB and A^+B^+, we obtain

$$\Omega Q_{31}^{(2)} = \Omega \frac{H_{31}^{(2)}}{N} \rightarrow \mu_1 AB + \mu_2 A^+B^+,$$
$$\Omega^* Q_{23}^{(2)} = \Omega^* \frac{H_{23}^{(2)}}{N} \rightarrow \mu_3 AB + \mu_4 A^+B^+. \tag{C25}$$

where we have set

$$\mu_1 = -\frac{8\gamma^6\delta|\Omega|^4\Omega^{*2}}{N^3}\{2\delta^5[\gamma + i(\delta+\Delta)]^2(i\gamma+\delta+\Delta)(\gamma^2+\Delta^2)$$
$$+\delta^4[8\gamma^4 - 4i\gamma^3\delta - \delta^4 - 10\delta^3\Delta - 12\delta^2\Delta^2 + 4\delta\Delta^3 + 8\Delta^4 + 4\gamma^2(3\delta^2+5\delta\Delta+4\Delta^2) + 2i\gamma\delta(\delta^2+6\delta\Delta+6\Delta^2)]|\Omega|^2$$
$$+2\delta^3[11\delta^3 - 4i\gamma^3 + 2\gamma^2(\delta+2\Delta) + 42\delta^2\Delta + 54\delta\Delta^2 + 20\Delta^3 - 4i\gamma(2\delta^2+5\delta\Delta+5\Delta^2)]|\Omega|^4$$
$$-8\delta^2(2\gamma^2 + i\gamma\delta + 3\delta^2 + 9\delta\Delta + 10\Delta^2)|\Omega|^6 + 32\delta(2i\gamma - 3\delta - 2\Delta)|\Omega|^8 + 64|\Omega|^{10}\}, \tag{C26}$$

$$\mu_2 = -\frac{8i\gamma^6\delta\Omega^2|\Omega|^4}{N^3}\{\delta^6[\gamma + i(\delta+\Delta)](\delta+2\Delta)(\gamma^2+\Delta^2)$$
$$-2\delta^4[\gamma^3(5\delta+4\Delta) - \gamma\Delta(2\delta^2 - \delta\Delta - 4\Delta^2) + i\gamma^2(6\delta^2+11\delta\Delta+4\Delta^2) + i(-2\delta^3\Delta + 7\delta\Delta^3 + 4\Delta^4)]|\Omega|^2$$
$$-2\delta^3[4\gamma^3 - 4i\gamma^2(2\delta+\Delta) + 4\gamma(2\delta^2+5\delta\Delta+5\Delta^2) + i(7\delta^3 + 30\delta^2\Delta + 36\delta\Delta^2 + 12\Delta^3)]|\Omega|^4$$
$$+4i\delta^2[12\gamma^2 + 9\delta^2 + 4i\gamma(\delta+2\Delta) + 16\delta\Delta + 20\Delta^2]|\Omega|^6 - 16i\delta(6i\gamma - 11\delta - 10\Delta)|\Omega|^8 - 64i|\Omega|^{10}\}, \tag{C27}$$

$$\mu_3 = -\frac{8i\gamma^6\delta(\Omega^*)^2|\Omega|^4}{N^3}\{\delta^6[\gamma^2 + (\delta+\Delta)^2](\delta+2\Delta)(\gamma - i\Delta) + 2\delta^4[-i\gamma^2(\delta+4\Delta)(\delta-\Delta) + \gamma^3(\delta-4\Delta)$$
$$-\gamma(\delta+\Delta)(\delta^2+7\delta\Delta+4\Delta^2) - i(\delta+\Delta)(\delta^3 + 2\delta^2\Delta - 5\delta\Delta^2 - 4\Delta^3)]|\Omega|^2$$
$$+2\delta^3[4\gamma^3 - 4i\gamma^2(\delta-\Delta) + 4\gamma(2\delta^2+5\delta\Delta+5\Delta^2) + i(\delta^3 + 6\delta^2\Delta - 12\Delta^3)]|\Omega|^4$$
$$-4i\delta^2[12\gamma^2 + 13\delta^2 - 4i\gamma(\delta+2\Delta) + 24\delta\Delta + 20\Delta^2]|\Omega|^6 - 16i\delta(-6i\gamma + \delta - 10\Delta)|\Omega|^8 + 64i|\Omega|^{10}, \tag{C28}$$

$$\mu_4 = \frac{8\gamma^6\delta|\Omega|^4\Omega^2}{N^3}\{2\delta^5[\gamma^2 + (\delta+\Delta)^2](\gamma - i\Delta)^2(i\gamma - \Delta)$$
$$+\delta^4[8\gamma^4 - 4i\gamma^3\delta + \delta^4 + 6\delta^3\Delta + 24\delta^2\Delta^2 + 28\delta\Delta^3 + 8\Delta^4 + 4\gamma^2(2\delta^2+3\delta\Delta+4\Delta^2) + 2i\gamma\delta(\delta^2+6\delta\Delta+6\Delta^2)]|\Omega|^2$$
$$+2\delta^3[3\delta^3 - 4i\gamma^3 - 2\gamma^2(\delta+2\Delta) + 6\delta^2\Delta - 6\delta\Delta^2 - 20\Delta^3 - 4i\gamma(2\delta^2+5\delta\Delta+5\Delta^2)]|\Omega|^4$$
$$-8\delta^2(2\gamma^2 + i\gamma\delta + 4\delta^2 + 11\delta\Delta + 10\Delta^2)|\Omega|^6 - 32\delta(\delta - 2i\gamma - 2\Delta)|\Omega|^8 + 64|\Omega|^{10}\}. \tag{C29}$$

Thus substituting Eqs. (C25) and (C16) into Eq. (15), we have

$$H_V \rightarrow Be^{-i\delta t}Q_{31}^{(1)} + \Omega Q_{31}^{(2)} + A^+e^{-i\delta t}Q_{23}^{(1)} + \Omega^* Q_{23}^{(2)} + \text{H.c.}$$
$$= \Lambda AB + \Lambda^* A^+B^+ + \{\mu_1 AB + \mu_2 A^+B^+ + \mu_3 AB + \mu_4 A^+B^+ + \text{H.c.}\}$$
$$= (\Lambda + \mu_1 + \mu_3 + \mu_2^* + \mu_4^*)AB + \text{H.c.} = \lambda_z AB + \lambda_z^* A^+B^+, \tag{C30}$$

where

$$\lambda_z = \Lambda + \mu_1 + \mu_3 + \mu_2^* + \mu_4^*$$
$$= \frac{4\delta^6}{G_2^3}(\delta+2\Delta)\left\{[\gamma^2 + (\delta+\Delta)^2](\gamma^2+\Delta^2)\frac{\Omega^*}{\Omega} + (\delta^2 - 2\gamma^2 + 6\delta\Delta + 6\Delta^2)(\Omega^*)^2\right\}$$
$$-\frac{16\delta^2(\delta+2\Delta)}{G_2^3}[\delta^2(\gamma^2 + 2\delta^2 + 5\delta\Delta + 5\Delta^2) + \delta^2|\Omega|^2 - 8|\Omega|^4]|\Omega|^2(\Omega^*)^2. \tag{C31}$$

Considering the assumption condition $|\Omega/\delta|^2 \rightarrow 0$, then we have

$$\lambda_z \simeq \frac{4\delta^6}{G_2^3}(\delta+2\Delta)\left\{[\gamma^2 + (\delta+\Delta)^2](\gamma^2+\Delta^2)\frac{\Omega^*}{\Omega} + (\delta^2 - 2\gamma^2 + 6\delta\Delta + 6\Delta^2)(\Omega^*)^2\right\}, \tag{C32}$$

$$G_2 \simeq \delta^2(\delta^2 + 2\Delta\delta + 2\Delta^2 + 2\gamma^2). \tag{C33}$$

APPENDIX D: EFFECTIVE HAMILTONIAN

From Eqs. (C32) and (C30) and noting $\Omega = \bar{\Omega}_1 e^{i\vec{k}_{pF}\cdot\vec{r}} + \bar{\Omega}_2 e^{-i\vec{k}_{pB}\cdot\vec{r}}$,

$$A = d_{32} a e^{i\vec{k}_a\cdot\vec{r}} + d'_{32} c e^{-i\vec{k}_c\cdot\vec{r}}, \quad B = d_{31} b e^{i\vec{k}_b\cdot\vec{r}} + d'_{31} d e^{-i\vec{k}_d\cdot\vec{r}},$$

we have

$$\begin{aligned}\Omega^2 A^+ B^+ &= (\bar{\Omega}_1 e^{i\vec{k}_{pF}\cdot\vec{r}} + \bar{\Omega}_2 e^{-i\vec{k}_{pB}\cdot\vec{r}})^2 (d^*_{32} a^+ e^{-i\vec{k}_a\cdot\vec{r}} + d'^*_{32} c^+ e^{i\vec{k}_c\cdot\vec{r}})(d^*_{31} b^+ e^{-i\vec{k}_b\cdot\vec{r}} + d'^*_{31} d^+ e^{i\vec{k}_d\cdot\vec{r}})\\
&= d^*_{32} d^*_{31} a^+ b^+ \big[\bar{\Omega}_1^2 e^{i(2\vec{k}_{pF}-\vec{k}_a-\vec{k}_b)\cdot\vec{r}} + 2\bar{\Omega}_1\bar{\Omega}_2 e^{i(\vec{k}_{pF}-\vec{k}_{pB}-\vec{k}_a-\vec{k}_b)\cdot\vec{r}} + \bar{\Omega}_2^2 e^{-i(\vec{k}_a+\vec{k}_b+2\vec{k}_{pB})\cdot\vec{r}}\big]\\
&\quad + d^*_{32} d'^*_{31} a^+ d^+ \big[\bar{\Omega}_1^2 e^{i(2\vec{k}_{pF}-\vec{k}_a+\vec{k}_d)\cdot\vec{r}} + 2\bar{\Omega}_1\bar{\Omega}_2 e^{i(\vec{k}_{pF}-\vec{k}_{pB}-\vec{k}_a+\vec{k}_d)\cdot\vec{r}} + \bar{\Omega}_2^2 e^{-i(\vec{k}_a-\vec{k}_d+2\vec{k}_{pB})\cdot\vec{r}}\big]\\
&\quad + d'^*_{32} d^*_{31} b^+ c^+ \big[\bar{\Omega}_1^2 e^{i(2\vec{k}_{pF}-\vec{k}_b+\vec{k}_c)\cdot\vec{r}} + 2\bar{\Omega}_1\bar{\Omega}_2 e^{i(\vec{k}_{pF}-\vec{k}_{pB}-\vec{k}_b+\vec{k}_c)\cdot\vec{r}} + \bar{\Omega}_2^2 e^{-i(\vec{k}_b-\vec{k}_c+2\vec{k}_{pB})\cdot\vec{r}}\big]\\
&\quad + d'^*_{32} d'^*_{31} c^+ d^+ \big[\bar{\Omega}_1^2 e^{i(2\vec{k}_{pF}+\vec{k}_c+\vec{k}_d)\cdot\vec{r}} + 2\bar{\Omega}_1\bar{\Omega}_2 e^{i(\vec{k}_{pF}-\vec{k}_{pB}+\vec{k}_c+\vec{k}_d)\cdot\vec{r}} + \bar{\Omega}_2^2 e^{-i(2\vec{k}_{pB}-\vec{k}_c-\vec{k}_d)\cdot\vec{r}}\big],\end{aligned} \tag{D1}$$

and

$$|\bar{\Omega}_1 e^{i\vec{k}_{pF}\cdot\vec{r}} + \bar{\Omega}_2 e^{-i\vec{k}_{pB}\cdot\vec{r}}|^2 = |\bar{\Omega}_1|^2 + |\bar{\Omega}_2|^2 + \bar{\Omega}_1\bar{\Omega}_2^* e^{i(\vec{k}_{pF}+\vec{k}_{pB})\cdot\vec{r}} + \bar{\Omega}_1^*\bar{\Omega}_2 e^{-i(\vec{k}_{pF}+\vec{k}_{pB})\cdot\vec{r}}. \tag{D2}$$

We only examine the phase-matching items, and notice $\vec{k}_{pF} = \vec{k}_{pB}$, $\vec{k}_a = \vec{k}_d$, $\vec{k}_b = \vec{k}_c$. Under the phase-matching condition,

$$\begin{aligned}0 &= \vec{k}_a + \vec{k}_b - 2\vec{k}_{pF},\\ 0 &= \vec{k}_d + \vec{k}_c - 2\vec{k}_{pF},\\ 0 &= \vec{k}_b - \vec{k}_c - \vec{k}_{pF} + \vec{k}_{pB},\\ 0 &= \vec{k}_d - \vec{k}_a + \vec{k}_{pF} - \vec{k}_{pB},\end{aligned} \tag{D3}$$

Equations (D1) and (D2) can be rewritten as

$$\Omega^2 A^+ B^+ = \big[\bar{\Omega}_1^2 d^*_{32} d^*_{31} a^+ b^+ + 2\bar{\Omega}_1\bar{\Omega}_2 d^*_{32} d'^*_{31} a^+ d^+ + 2\bar{\Omega}_1\bar{\Omega}_2 d'^*_{32} d^*_{31} b^+ c^+ + \bar{\Omega}_2^2 d'^*_{32} d'^*_{31} c^+ d^+\big], \tag{D4}$$

and

$$|\bar{\Omega}_1 e^{i\vec{k}_{pF}\cdot\vec{r}} + \bar{\Omega}_2 e^{-i\vec{k}_{pB}\cdot\vec{r}}|^2 = |\bar{\Omega}_1|^2 + |\bar{\Omega}_2|^2. \tag{D5}$$

Substituting Eqs. (C32) and (C33), and (D4) and (D5) into (C30) yields the effective Hamiltonian

$$H_{\text{eff}} = \kappa_1 a^+ b^+ + \kappa_2 c^+ d^+ + 2\sqrt{\kappa_1\kappa_2}(a^+ d^+ + b^+ c^+) + \text{H.c.}, \tag{D6}$$

where κ_1 and κ_2 are defined in Eqs. (24) and (25). For simplicity, here we have taken $d_{31} \approx d'_{31}$, $d_{32} \approx d'_{32}$, because the frequencies are almost the same.

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