Quadripartite entanglement from a double three-level $\Lambda$-type-atom model

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We present a theoretical scheme for the generation of four quantized modes in a three-level double-$\Lambda$ 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 with an atomic system is even important for developing the multimode quantum network. In this paper, we propose a scheme to realize the four-wave entanglement in two double-$\Lambda$ 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 $\omega_0$ are counterpropagating through the system with the opposite wave vectors (denoted as $\vec{k}_{pf}$ and $-\vec{k}_{pf}$, respectively). A weak probe field with frequency $\omega_p$ propagates into the cell with a small angle $\theta$. The frequencies of three levels $|l\rangle$ ($l = 1, 2, 3$) are $\omega_l$ and their difference is $\omega_{ij} = \omega_i - \omega_j$. Both strong pump beams induce the transition $|3\rangle \rightarrow |1\rangle$ and $|3\rangle \rightarrow |2\rangle$ with detunings $\Delta = \omega_0 - \omega_3$ and $\delta + \Delta = \omega_0 - \omega_2$, where $\delta = \omega_{23}$ is the frequency difference between levels $|2\rangle$ and $|1\rangle$. The Rabi frequency of the forward (backward) pump beam (classical field) is $\Omega_1 (\Omega_2)$. We consider the generation of four quantized modes, $a$ of frequency $\omega_a$, $b$ of frequency $\omega_b$, $c$ of frequency $\omega_c$, and $d$ of frequency $\omega_d$. The wave vectors of the four quantized modes are denoted as $\vec{k}_a$, $\vec{k}_b$, $-\vec{k}_a$, and $-\vec{k}_b$ [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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If only the forward pumping beam ($\Omega_2 = 0$) 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 ($\hbar = 1$),

$$H = \sum_{i=1}^{3} \omega_i \sigma_i + H_V(t),$$

where the interaction Hamiltonian is

$$H_V(t) = (\Omega e^{-i\omega t} + Be^{-i\omega t})\sigma_{31}$$

$$+ (\Omega^* e^{i\omega t} + Ae^{i\omega t})\sigma_{32} + H.c.$$  (2)

In the above $\Omega = \Omega_1 e^{i\delta} + \Omega_2 e^{-i\delta}$, $A = d_{32} e^{i\theta} + d_{31} e^{-i\theta}$, and $B = d_{33} e^{i\theta} + d_{31} e^{-i\theta}$. The $d_{ij}$ being the coupling constants between the two atomic transitions and the four quantized modes ($b, a$ and $c, d$), respectively. Here we have assumed $\omega_b = \omega_d$, $\omega_a = \omega_c$, $\delta_a = \delta_d$, and $\delta_b = \delta_c$ 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.,

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

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

$$\dot{\sigma}_{11} = \gamma_1 \sigma_{33} + i(\Omega e^{-i\omega t} + B) e^{-i\omega t} \sigma_{31}$$

$$- i(\Omega^* e^{i\omega t} + A) e^{i\omega t} \sigma_{13},$$  (4)

$$\dot{\sigma}_{22} = \gamma_2 \sigma_{33} + i(\Omega e^{i\omega t} + A) e^{i\omega t} \sigma_{32}$$

$$- i(\Omega^* e^{-i\omega t} + B) e^{-i\omega t} \sigma_{23},$$  (5)

$$\dot{\sigma}_{33} = - (\gamma_1 + \gamma_2) \sigma_{33} - i(\Omega e^{-i\omega t} + B) e^{-i\omega t} \sigma_{31}$$

$$- i(\Omega^* e^{i\omega t} + A) e^{i\omega t} \sigma_{32} + i(\Omega^* e^{i\omega t} + \theta_{31}^* B^+) e^{i\omega t} \sigma_{13}$$

$$+ i(\Omega^* e^{-i\omega t} A^+) e^{-i\omega t} \sigma_{23},$$  (6)

where $\gamma_1$ and $\gamma_2$ are the decay rates of the excited states $|3\rangle$ and $|2\rangle$, respectively. $\gamma_1, \gamma_2$, and $\gamma_{31}$ 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 ($\delta$ and $\Delta$). 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 $\Omega_2 = d_{32} = d_{31} = 0$. 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:

$$Q_{mm} = \sigma_{mm}, \quad (m = 1, 2, 3)$$

$$Q_{31} = \gamma_1 \sigma_{33} e^{-i\omega t}$$

$$Q_{32} = \gamma_2 \sigma_{33} e^{-i\omega t},$$  (10)

$$Q_{21} = \sigma_{21}.$$

Then the interaction Hamiltonian becomes

$$H_V(t) = (\Omega e^{-i\omega t} + B e^{-i\omega t})Q_{31} e^{i\omega t}$$

$$+ (\Omega^* e^{i\omega t} + A e^{i\omega t})Q_{32} e^{i\omega t} + H.c.$$  (11)

and the equations of atomic operators become

$$\dot{Q}_{11} = \gamma_1 Q_{33} - i(\Omega^* e^{i\omega t} + \theta_{31}^* B^+) Q_{13} + i(\Omega + e^{i\omega t} B) Q_{31},$$

$$\dot{Q}_{22} = \gamma_2 Q_{33} - i(\Omega^* e^{i\omega t} + \theta_{31}^* A^+) Q_{23} + i(\Omega + e^{i\omega t} A) Q_{32},$$

$$\dot{Q}_{33} = -(\gamma_1 + \gamma_2) Q_{33} - i(\Omega + e^{i\omega t} A^+) Q_{33}$$

$$- i(\Omega + e^{i\omega t} A) Q_{33} + i(\Omega^* e^{i\omega t} + \theta_{31}^* B^+) Q_{33}$$

$$+ i(\Omega^* e^{-i\omega t} A^+) e^{-i\omega t} Q_{33},$$  (12)

$$\dot{Q}_{31} = i\Gamma_p Q_{31} + i(\Omega^* e^{i\omega t} + \theta_{31}^* B^+) (Q_{11} - Q_{33})$$

$$+ i(\Omega + e^{i\omega t} A^+) Q_{21},$$

$$\dot{Q}_{32} = -i\Gamma_a Q_{33} + i(\Omega^* e^{i\omega t} + \theta_{31}^* B^+) Q_{21}$$

$$+ i(\Omega + e^{i\omega t} A^+) (Q_{32} - Q_{33}),$$  (13)

$$\dot{Q}_{21} = i\Gamma_2 Q_{32} - i(\Omega + e^{i\omega t} B) Q_{23} + i(\Omega + e^{i\omega t} A) Q_{31},$$

where $\Gamma_p = -\Delta + i\gamma_3, \Gamma_a = \delta + \Delta - i\gamma_2, \Gamma_2 = \delta + i\gamma_2$, and $\Delta = \omega_0 - \omega_3$.

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

$$Q_{31} = Q_{31}^{(0)} + Q_{31}^{(1)} + Q_{31}^{(2)} + \cdots,$$

$$Q_{23} = Q_{23}^{(0)} + Q_{23}^{(1)} + Q_{23}^{(2)} + \cdots.$$  (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 $\Omega$), we have an effective Hamiltonian,

$$H_V(t) = (\Omega + Be^{-it})(Q_{31}^{(0)} + Q_{31}^{(1)} + Q_{31}^{(2)})$$
$$+ (\Omega^\ast + A^\ast e^{-it})(Q_{23}^{(0)} + Q_{23}^{(1)} + Q_{23}^{(2)}) + \text{H.c.}$$
$$\approx Be^{-it}Q_{31}^{(0)} + \Omega Q_{31}^{(2)} + A^\ast e^{-it}Q_{23}^{(1)} + \Omega^\ast Q_{23}^{(2)} + \text{H.c.}$$

(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 $Q_{23}$

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)

$$Q_{31}^{(0)} = \frac{1}{G_2}[\delta^2(\Delta^2 + \gamma^2) + \delta(\Delta + 2\Delta)|\Omega|^2 + 4|\Omega|^4],$$

(16)

$$Q_{23}^{(0)} = \frac{1}{G_2}[\delta^2(\Delta^2 + 2\Delta^2 + \gamma^2 + 2\delta^2) - \delta(\Delta + 2\Delta)|\Omega|^2 + 4|\Omega|^4],$$

(17)

$$Q_{31}^{(0)} = \frac{2\delta^2}{G_2}|\Omega|^2,$$

(18)

$$Q_{23}^{(0)} = \frac{\delta}{G_2}\Omega^2[(i\gamma + \Delta)\delta + 2|\Omega|^2] = (Q_{31}^{(0)})^\ast,$$

(19)

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

(20)

$$Q_{23}^{(0)} = \frac{|\Omega|^2}{G_2}[\delta(\Delta + 2\gamma - 4|\Omega|^2] = (Q_{21}^{(0)})^\ast,$$

(21)

where

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

(22)

and we have taken $\gamma_1 = \gamma_2 = \gamma_3 = \gamma_4 = \gamma$ and set $\gamma_1 = 0$. From Eqs. (16)–(21) one can clearly see, under the conditions $|\Delta/\delta|^2 \approx 0$ and $|\gamma/\delta|^2$, $|\Omega/\delta|^2$, $|\Delta/\delta|^2$ $\rightarrow$ 0, that $Q_{31}^{(0)} \approx Q_{31}^{(0)} \approx Q_{31}^{(0)} \approx Q_{23}^{(0)} \approx 0$ and $Q_{23}^{(2)} \approx 1$, 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 $B = d_{31}be^{-ik_3\tau} + d_{31}d_{12}e^{-ik_2\tau}$ and $\Omega = \Omega_1e^{i\xi_{1}\tau} + \Omega_2e^{-i\xi_{2}\tau} + \Omega_3e^{i\xi_{3}\tau} + \Omega_4e^{i\xi_{4}\tau}$, we can obtain the effective Hamiltonian in the interaction picture (see Appendix D):

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

(23)

$$\kappa_1 = \kappa \Omega_1^2d_{31}^\dagger d_{31}^\ast, \quad \kappa_2 = \kappa \Omega_2^2d_{31}^\dagger d_{31}^\ast,$$

(24)

$$\kappa = \frac{4\delta^6(\delta + 2\Delta)}{[\delta^2(\delta^2 + 2\Delta^2 + 2\Delta^2)]^3} \left\{ \frac{|\gamma|^2 + (\delta + \Delta)^2}{|\Omega|^2} + \frac{|\Omega|^2}{|\Omega|^2} \right\},$$

(25)

where $d_{31} \approx d_{31}^\dagger$ and $d_{31} \approx d_{31}^\dagger$ 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 $\Omega_2 = 0$. 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 ($\Omega_2 = 0$), Eq. (23) reduces to the two-mode case. For convenience, we set $\kappa_1 = |\kappa_1| e^{i\phi} = i\kappa_2^2 e^{i(\phi - \pi/2)}, \kappa_2 = |\kappa_2| e^{i\phi} = i\kappa_2^2 e^{i(\phi - \pi/2)}$, $a_+ e^{i(\phi - \pi/2)} = \bar{a}_+, b_+ e^{i(\phi - \pi/2)} = \bar{b}_+, c_+ e^{i(\phi - \pi/2)} = \bar{c}_+$, and $d_+ e^{i(\phi - \pi/2)} = \bar{d}_+$. Thus Eq. (23) can be rewritten as

$$H_{eff} = i\left[\kappa_2^2\bar{a}_+ \bar{b}_+ + \kappa_2^2\bar{c}_+ \bar{d}_+ + 2\kappa_2\kappa_2\kappa_2^2(\bar{a}_+ \bar{d}_+ + \bar{c}_+ \bar{b}_+)\right] + \text{H.c.},$$

(26)

where

$$\kappa_2^2 = |\kappa_2| \Omega_2^2d_{31}^\dagger d_{31}^\ast,$$

(27)

$$\kappa_2^2 = |\kappa_2| \Omega_2^2d_{31}^\dagger d_{31}^\ast,$$

$$\kappa_2 \kappa_2 = |\kappa_2| \Omega_2^2d_{31}^\dagger d_{31}^\ast.$$

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 $\omega_0$ 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 $\omega_0$ with an atom from level $|2\rangle$ to level $|1\rangle$ via level $|3\rangle$; the second, absorbing a backward pumping photon and then generating a photon of $\omega_{ad}$ 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 $\omega_{ad}$ with an atom from level $|2\rangle$ to level $|1\rangle$ via level $|3\rangle$; the third, absorbing a forward pumping photon and then generating a photon of $\omega_{ad}$ 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 $\omega_{ad}$ with an atom from level $|2\rangle$ to level $|1\rangle$ via level $|3\rangle$; the fourth, absorbing a backward pumping photon and then generating a photon of $\omega_{ad}$ with an atom from level $|2\rangle$ to level $|1\rangle$ via level $|3\rangle$. 


with } \xi = \sqrt{\kappa_1^2 + 14\kappa_2^2\kappa_b^2 + \kappa_b^4}, \quad \eta_\pm = \pm (\xi - \kappa_b^2)/2. \text{ All the information needed to calculate the van Looock–Furusawa (VLF) correlation [25] is contained in Eqs. (31) and (32).}

\section{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, P_o\} = i, o = a, b, c, d. Using the quadrature components, VLF inequalities can be given by

\begin{align}
V_{ab} &= \delta^2(X_{a} - X_{b}) + \delta^2(P_{a} + P_{b} + g_{a}P_{a} + g_{a}P_{b}) < 2,
V_{bc} &= \delta^2(X_{b} - X_{c}) + \delta^2(P_{b} + P_{c} + g_{b}P_{b} + g_{b}P_{c}) < 2,
V_{bd} &= \delta^2(X_{b} - X_{d}) + \delta^2(P_{b} + P_{d} + g_{b}P_{b} + g_{b}P_{d}) < 2,
V_{cd} &= \delta^2(X_{c} - X_{d}) + \delta^2(P_{c} + P_{d} + g_{c}P_{c} + g_{c}P_{d}) < 2,
\end{align}

where \(\delta^2(X) = \langle X^2 \rangle - \langle X \rangle^2\) and the parameters \(g_{a}, g_{b}, g_{c}\) 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

\begin{align}
V_{ab} < 2, \quad V_{bc} < 2, \quad V_{cd} < 2.
\end{align}

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 \(V_{ab}\), these average values \(\langle \hat{a}^2 \rangle, \langle \hat{a}^2 \rangle, \langle \hat{b}^2 \rangle, \langle \hat{b}^2 \rangle, \langle \hat{a}^2 \rangle, \langle \hat{b}^2 \rangle\), and \(\langle \hat{a}, \rangle, \langle \hat{b}, \rangle, \langle \hat{b} \rangle\) are needed. For this system, at the initial time \(t = 0\), all the four modes are in vacuum; thus we have \(\langle \hat{a}(0) \rangle = \langle \hat{a}(0) \rangle = \langle \hat{b}(0) \rangle = \langle \hat{b}(0) \rangle = 0\), which leads to the average values of the amplitude all being zero, i.e., \(X_{a}(0) = X_{b}(0) = P_{a}(0) = P_{b}(0) = 0\). Due to the bosonic communication relations, not all the moments vanish, such as \(\langle X_{a}(0)X_{b}(0) \rangle = \langle P_{a}(0)P_{b}(0) \rangle = \delta_{ab}/2\), and \(\langle X_{a}(0)X_{b}(0) \rangle = \langle P_{a}(0)P_{b}(0) \rangle = \delta_{ab}/2\). The variances at time \(t\) can be obtained based on Eqs. (31) and (32),

\begin{align}
\delta^2X_a &= \delta^2X_b = \delta^2P_a = \delta^2P_b = \frac{1}{4\xi} \left[ \xi + \cos(2\eta_{+}t) + \xi - \cos(2\eta_{-}t) \right],
\delta^2X_c &= \delta^2X_d = \delta^2P_c = \delta^2P_d = \frac{1}{4\xi} \left[ \xi - \cos(2\eta_{+}t) + \xi + \cos(2\eta_{-}t) \right],
\end{align}

and

\begin{align}
\langle X_{a}X_{b} \rangle &= -\langle P_{a}P_{b} \rangle = \frac{1}{4\xi} \left[ \xi + \sin(2\eta_{+}t) + \xi - \sin(2\eta_{-}t) \right],
\langle X_{c}X_{d} \rangle &= -\langle P_{c}P_{d} \rangle = \frac{1}{4\xi} \left[ \xi - \sin(2\eta_{+}t) + \xi + \sin(2\eta_{-}t) \right].
\end{align}
From the optimization process, one can find the required variance can be rewritten as, taking $V_{ab}$ as an example,

$$V_{ab} = 4\delta_{x}^{2}x_{a} + (g_{g}^{2} + g_{d}^{2})\delta_{x}^{2}p_{e} - (X_{a}X_{b}) + 2g_{g}(P_{a}P_{e} + P_{a}P_{c}) + 2g_{d}g_{d}P_{a}(P_{c}P_{d}).$$

Following Ref. [33], a simple minimization of the right-hand sides of Eq. (41) with respect to the $g_{c}$ and $g_{d}$ gives

$$g_{c} = g_{d} = -\frac{(P_{c}P_{e} + P_{a}P_{d})}{(P_{e}P_{d}) + (P_{c}P_{d})},$$

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

$$V_{ab} = 4\delta_{x}^{2}x_{a} + 2g_{g}^{2}\delta_{x}^{2}p_{e} - (X_{a}X_{b}) + 4g_{g}(P_{a}P_{e} + P_{a}P_{c}) + 2g_{d}g_{d}(P_{e}P_{d}).$$

Similarly, one can obtain that

$$V_{bc} = (2 + g_{g}^{2})\delta_{x}^{2}x_{b} + (2 + g_{d}^{2})\delta_{x}^{2}x_{c} - (X_{a}X_{b}) + 2g_{g}g_{d}(P_{a}P_{e} + P_{a}P_{c}) + 2g_{d}g_{d}(P_{e}P_{d} + P_{a}P_{c}),$$

$$V_{ca} = 4\delta_{x}^{2}x_{c} + 2g_{g}^{2}\delta_{x}^{2}p_{c} - (X_{a}X_{b}) + 4g_{g}(P_{a}P_{e} + P_{a}P_{c}) + 2g_{d}g_{d}(P_{e}P_{d}),$$

where we have set

$$g_{a} = \frac{1}{2}(P_{a}P_{b} + P_{a}P_{d})\delta_{x}^{2}P_{a}$$

$$-\frac{1}{2}(P_{a}P_{b} + P_{a}P_{d})P_{a}P_{d} - \frac{1}{2}(P_{b}P_{a} + P_{a}P_{d})P_{b}P_{a} - \frac{1}{2}(P_{a}P_{d} + P_{a}P_{c})P_{a}P_{c} - \frac{1}{2}(P_{b}P_{a} + P_{a}P_{d})P_{b}P_{c},$$

and

$$g_{d} = (4g_{g}^{2} + g_{d}^{2})\delta_{x}^{2}p_{e} - (X_{a}X_{b}) + 2g_{g}g_{d}(P_{a}P_{e} + P_{a}P_{c}) + 2g_{d}^{2}(P_{e}P_{d} + P_{a}P_{c}),$$

$$g_{c} = -\frac{1}{2}(P_{a}P_{b} + P_{a}P_{d})\delta_{x}^{2}P_{a}$$

$$-\frac{1}{2}(P_{a}P_{b} + P_{a}P_{d})P_{a}P_{d} - \frac{1}{2}(P_{b}P_{a} + P_{a}P_{d})P_{b}P_{a} - \frac{1}{2}(P_{a}P_{d} + P_{a}P_{c})P_{a}P_{c} - \frac{1}{2}(P_{b}P_{a} + P_{a}P_{d})P_{b}P_{c},$$

$$g_{g} = (4g_{g}^{2} + g_{g}^{2})\delta_{x}^{2}p_{e} - (X_{a}X_{b}) + 2g_{g}g_{g}(P_{a}P_{e} + P_{a}P_{c}) + 2g_{g}^{2}(P_{e}P_{d} + P_{a}P_{c}),$$

$$g_{d} = (4g_{g}^{2} + g_{d}^{2})\delta_{x}^{2}p_{e} - (X_{a}X_{b}) + 2g_{g}g_{d}(P_{a}P_{e} + P_{a}P_{c}) + 2g_{d}^{2}(P_{e}P_{d} + P_{a}P_{c}).$$

V. DISCUSSIONS

First, let us consider the two-mode entanglement case. As mentioned above, we can obtain the two-mode entanglement case by taking $\Omega_{2} = \Omega_{1} = \Omega_{2} = 0$. In this case, we have $\kappa_{b} = 0$, $\xi = \kappa_{b}^{2}$ and $\eta_{+} = \xi$, $\eta_{-} = 0$, and from Eqs. (37), (39), and (43) we can find that $V_{ab} = 2e^{-\alpha_{g}^{2}} < 2$, which demonstrates the genuine bipartite entanglement, as expected [12]. In Fig. 2, for the two-mode case, we plot the graph of squeezing parameter $\kappa_{g}^{2}$ as the function of $\Omega_{1}$ and $\Delta$ for several given values of $\delta$: $\Omega_{1} = \sqrt{\Delta} = \sqrt{\Delta}_{2} = -250$ 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 $\kappa_{g}^{2}$ increases with the decreasing value of $\delta$ and the increasing absolute value of $\Delta$ (especially for $\Delta > 0$), respectively. In addition, for the case of $\Delta > 0$, $\kappa_{g}^{2}$ becomes bigger as the increasing value $\Omega_{1}$. This case is not true for the case of $\Delta < 0$. It is interesting to notice that the effect of $\Omega_{1}$ on $\kappa_{g}^{2}$ is unsymmetrical, i.e., one can obtain a bigger value of $\kappa_{g}^{2}$ with the increasing value of $\delta > 0$ than that with the increasing absolute value of $\Delta < 0$ for a given value of $\delta$. Especially, $\kappa_{g}^{2}$ 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 $\Delta$: (a) $\Delta = 34.78 \gamma$, (b) $\Delta = 104.35 \gamma$, (c) $\Delta = 173.91 \gamma$, (d) $\Delta = 347.83 \gamma$, with $\delta = 0.52 \times 10^{3} \gamma$, $\Omega_{1} = 104.35 \gamma$ ($\Omega_{2} = 0.56 \Omega_{1}$), and $G = -43.48 \gamma$. These parameters are based on the Rb atoms [34–36]. The black line is for $V_{bc}$, red line is for $V_{ab}$, and blue line is for $V_{bc}$ (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 quadrupartite entanglement. Furthermore, the correlation $V_{bc}$ between $\beta$ and $\beta'$ modes seems stronger than those involved in the other two modes. The interaction time range, within which quadrupartite entanglement is present, decreases with the increasing detune $\Delta$. In order to observe the quadrupartite entanglement with other parameters fixed, small
$\Delta$ corresponds to a long interaction time (or long vapor cell), and large $\Delta$ corresponds to a short interaction time (or short vapor cell).

In Fig. 4, we plot the VLF correlations for different combination of $\delta$, $\Delta$, 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 $\delta/\gamma$. Therefore, the interaction time to observe the entanglement is different for different $\delta$. The bigger $\delta/\gamma$ 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 $-43.86\gamma$ to $-76.75\gamma$, which tells us that the quadripartite entanglement can exist over a long
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
theoretical scheme to generate the entanglement of four light beams in a three-level double-$A$ 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.

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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 $\hat{\Omega}_2 = 0$ ($\hat{\Omega}_1 = \Omega$) in Eq. (23), i.e.,

$$H_{et} = \kappa_1 a^\dagger b + \kappa_2 a b,$$

(A1)

where $\kappa_1 = \kappa \Omega^2 d_1^a d_2^a$ and $\kappa$ is defined in Eq. (25) with $\hat{\Omega}_2 = 0$.

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

$$\dot{Q}_{31} = i\Gamma_p Q_{31} + i(\Omega^* + d_1^e e^{i\beta^*} b^*)(Q_{11} - Q_{33}) + i(\Omega^* + d_2^e e^{i\beta^*} a^*) Q_{21} + F_{31},$$

$$\dot{Q}_{23} = i\Gamma_p Q_{23} - i(\Omega + d_3^e e^{-i\beta} b) Q_{23} - i\Omega(a^\dagger + d_3^e e^{-i\beta} a) Q_{23} + F_{23},$$

(A2)

$$\dot{Q}_{21} = i\Gamma_1 Q_{21} + i(\Omega + d_3^e e^{i\beta} a^*) Q_{11} - i(\Omega^* + d_2^e e^{i\beta} b^*) Q_{23},$$

$$\dot{Q}_{13} = -i\Gamma_p Q_{13} - i(\Omega + d_3^e e^{-i\beta} b^*) (Q_{11} - Q_{33}) - i(\Omega + d_2^e e^{i\beta} a^*) Q_{13} + F_{13},$$

$$\dot{Q}_{32} = -i\Gamma_a Q_{32} + i(\Omega^* + d_1^e e^{i\beta} b) Q_{12} + i(\Omega^* + d_2^e e^{-i\beta a^*}) Q_{22} - Q_{33} + F_{32},$$

(A3)

$$\dot{Q}_{12} = -i\Gamma_1 Q_{12} - i(\Omega^* + d_2^e e^{-i\beta a^*}) Q_{12} + i(\Omega + d_3^e e^{-i\beta} b) Q_{32} + F_{12},$$

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

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

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 effective Hamiltonian, so that here we only consider the Langevin noise effect to the field. The degree 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.

$$0 = i\Gamma_p Q_{31} + i\Omega^* (Q_{11} - Q_{33}) + i\Omega^* Q_{21} + F_{31},$$

$$0 = -i\Gamma_a Q_{32} + i\Omega^* (Q_{22} - Q_{33}) + i\Omega^* Q_{12} + F_{32},$$

(A4)

$$0 = i\Gamma_1 Q_{21} - i\Omega^* Q_{23} + i\Omega Q_{31} + F_{21},$$

with

$$i F_{31} = \Gamma_p Q_{31} + \Omega^* Q_{21},$$

$$i F_{23} = \Gamma_a Q_{32} - \Omega Q_{21},$$

$$i F_{21} = \Gamma_1 Q_{21} - \Omega^* Q_{23} + \Omega Q_{31},$$

(A5)

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

$$Q_{31} \rightarrow F_a \equiv \alpha_3 Q_{31} + \alpha_2 Q_{23} + \alpha_1 F_{21}, \quad \text{for } Q_{31},$$

$$Q_{23} \rightarrow F_b \equiv \beta_3 Q_{23} + \beta_2 Q_{12} + \beta_1 F_{12}, \quad \text{for } Q_{23},$$

(A6)

where we have set

$$\alpha_3 = \frac{i}{T}(\Omega^*)^2, \quad \alpha_2 = \frac{i}{T}(i\gamma + \delta + \Delta)\Omega^*,$$

(A7)

and

$$\beta_3 = \frac{i}{T} \Omega^2, \quad \beta_2 = \frac{i}{T}(-i\gamma + \Delta)\Omega,$$

(A8)

with the definition of the denominator as

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

(A9)

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

$$\dot{H}_{et} = \kappa_1 a^+ b + \kappa_2 a b + \{d_3^e e^{-i\beta b^*} F_a + d_3^e e^{i\beta a^*} F_b + \text{h.c.}\},$$

(A10)

and the Heisenberg motion equations for optical fields are given by

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

(A11)

where we have set $\tilde{F}_b = d_3^e e^{-i\beta} F_b$, $\tilde{F}_a = d_3^e e^{i\beta} F_a$.

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

$$\frac{d}{dt} X_a = \kappa_1 X_a + i (\tilde{F}_a)^+ - \tilde{F}_b,$$

$$\frac{d}{dt} X_b = \kappa_1 X_a + i (\tilde{F}_a)^+ + \tilde{F}_a,$$

(A12)
Consequently, we have
\[
\begin{align*}
\frac{d}{dt} P_a &= -\kappa_1 P_b - \frac{(\tilde{F}_b)^+ + \tilde{F}_b}{\sqrt{2}}, \\
\frac{d}{dt} P_b &= -\kappa_1 P_a + \frac{(\tilde{F}_a)^+ + \tilde{F}_a}{\sqrt{2t}}.
\end{align*}
\] (A13)

In order to solve Eqs. (A12) and (A13), we rewrite Eq. (A12) as
\[
\frac{d}{dt} u(t) = J u(t) + F(t),
\] (A14)

where
\[
\begin{align*}
u(t) &= \begin{bmatrix} X_a(t) \\ X_b(t) \end{bmatrix}, \\
J &= \begin{bmatrix} 0 & \kappa_1 \\ -\kappa_1 & 0 \end{bmatrix}, \\
F(t) &= \frac{1}{\sqrt{2}} \begin{bmatrix} i(\tilde{F}_a)^+ - i \tilde{F}_b \\ \tilde{F}_b + (\tilde{F}_a)^+ \end{bmatrix} = \begin{bmatrix} f_a(t) \\ f_b(t) \end{bmatrix},
\end{align*}
\] (A15)

The solution of Eq. (A15) is
\[
u(t) = e^{Jt} \left[ u(0) + \int_0^t e^{-Jt'} F(t') dt' \right].
\] (A16)

In order to solve Eq. (A14), we can diagonalize the matrix \( J \) as
\[
J = U J_d U^{-1},
\] (A17)

with
\[
\begin{align*}
J_d &= \begin{bmatrix} -\kappa_1 & 0 \\ 0 & \kappa_1 \end{bmatrix}, \\
U &= \begin{bmatrix} -1 & 1 \\ 1 & 1 \end{bmatrix}, \\
U^{-1} &= \frac{1}{2} \begin{bmatrix} -1 & 1 \\ 1 & 1 \end{bmatrix}.
\end{align*}
\] (A18)

Consequently, we have
\[
\begin{align*}
X_a(t) &= \cosh[\kappa_1 t] X_{a0} + \sinh[\kappa_1 t] X_{b0} \\
&+ \int_0^t [\cosh[\kappa_1(t - t')] f_a(t')] dt', \\
&+ \sinh[\kappa_1(t - t')] f_b(t') dt',
\end{align*}
\] where we have set \( X_{a0} \) and \( X_{b0} \) are the initial quadrature amplitude. From Eqs. (A19) and (A20), we obtain
\[
\begin{align*}
X_a(t) - X_b(t) &= (X_{a0} - X_{b0}) e^{-\kappa_1 t} \\
&+ \int_0^t [f_a(t') - f_b(t')] e^{-\kappa_1 (t - t')} dt'.
\end{align*}
\] (A21)

In a similar way, we obtain
\[
P_b(t) = \cosh[\kappa_1 t] P_{a0} - \sinh[\kappa_1 t] P_{b0} \\
+ \int_0^t [\cosh[\kappa_1(t - t')] f'_a(t')] dt',
\] (A22)
denoting $d_{32}^* \beta_{ij} \to \beta_{ij}$, $d_{31} \alpha_{ij} \to \alpha_{ij}$, we obtain

\[
\langle f_a(t') f_a(t'') \rangle = \frac{1}{2} \left[ e^{2\delta t} D_{b^+, b^+} - D_{b^+, b^+} - D_{b^+, b^+} + e^{-2\delta t} D_{b, b} \right] \delta(t' - t''), \tag{A29}
\]

\[
\langle f_a(t') f_b(t'') \rangle = \frac{1}{2} \left[ e^{2\delta t} D_{b^+, a^+} + D_{b^+, a^+} - D_{b^+, a^+} - e^{-2\delta t} D_{b, a} \right] \delta(t' - t''),
\]

\[
\langle f_b(t') f_a(t'') \rangle = \frac{1}{2} \left[ e^{2\delta t} D_{a^+, a^+} + D_{a^+, a^+} - D_{a^+, a^+} + e^{-2\delta t} D_{a, a} \right] \delta(t' - t''),
\]

and

\[
\langle f_a(t') f_b(t'') \rangle = \frac{1}{2} \left[ e^{2\delta t} D_{a^+, a^+} + D_{a^+, a^+} + D_{b^+, b^+} + e^{-2\delta t} D_{b, b} \right] \delta(t' - t''),
\]

where $D_{oo}$ is defined by $D_{oo} = \langle F_0 F_0 \rangle$, $D_{oo'} = \langle (F_0^+) F_0 \rangle$, and $D_{o'o'} = \langle F_0 (F_0')^+ \rangle$, whose expressions will be determined later.

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

\[
I = e^{-2\delta t} + \frac{1}{4\kappa_1} \left( D_{b^+, b^+} + D_{b, b^+} \right) + i (D_{a^+, b^+} - D_{b^+, a^+} + D_{b, a^+} - D_{a^+, b^+}), \tag{A32}
\]

where $D_{b, b} \delta(t - t') = \langle [F_b(t)]^+ F_b(t') \rangle$, $D_{a^+, b^+} \delta(t - t') = \langle [F_b(t)]^+ F_b(t') \rangle$, $D_{b^+, a^+} \delta(t - t') = \langle [F_b(t)] F_b(t') \rangle$.

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 $F_{13}$, $F_{32}$, $F_{12}$.

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

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

\[
D_{b^+, b} = \gamma |d_{32}|^2 \left[ 2 |\beta_{31}|^2 \langle Q_{11} \rangle + (|\beta_{21}|^2 + |\beta_{31}|^2) \right],
\]

\[
D_{b, b^+} = \gamma |d_{32}|^2 \left[ 2 |\beta_{23}|^2 \langle Q_{22} \rangle + (|\beta_{21}|^2 + |\beta_{23}|^2) \right],
\]

\[
D_{a^+, a} = \gamma d_{31}^* d_{32} \left[ 2 |\beta_{31}|^2 \langle Q_{11} \rangle + (|\beta_{21}|^2 + |\beta_{31}|^2) \right],
\]

\[
D_{b^+, a^+} = \gamma d_{31}^* d_{32} \left[ 2 |\beta_{31}|^2 \langle Q_{11} \rangle + (|\beta_{21}|^2 + |\beta_{31}|^2) \right],
\]

\[
D_{b, a^+} = \gamma d_{31}^* d_{32} \left[ 2 |\beta_{23}|^2 \langle Q_{22} \rangle + (|\beta_{21}|^2 + |\beta_{23}|^2) \right].
\]
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

$$1 = Q_{11}^{(0)} + Q_{22}^{(0)} + Q_{33}^{(0)},$$

$$0 = \gamma_1 Q_{33}^{(0)} - i \Omega^* Q_{13}^{(0)} + i \Omega Q_{31}^{(0)},$$

$$0 = \gamma_2 Q_{33}^{(0)} - i \Omega^* Q_{23}^{(0)} + i \Omega Q_{32}^{(0)},$$

$$0 = \Gamma_p Q_{33}^{(0)} + \Omega^* (Q_{11}^{(0)} - Q_{33}^{(0)}) + \Omega^* Q_{21}^{(0)},$$

$$0 = \Gamma_\alpha Q_{32}^{(0)} - \Omega^* (Q_{22}^{(0)} - Q_{33}^{(0)}) - \Omega^* Q_{12}^{(0)},$$

$$0 = \Gamma_{21} Q_{23}^{(0)} - \Omega^* Q_{23}^{(0)} + \Omega Q_{31}^{(0)},$$

$$0 = \Gamma_{13} Q_{13}^{(0)} + \Omega (Q_{11}^{(0)} - Q_{33}^{(0)}) + \Omega Q_{12}^{(0)},$$

$$0 = \Gamma_{23} Q_{23}^{(0)} - \Omega (Q_{22}^{(0)} - Q_{33}^{(0)}) - \Omega Q_{21}^{(0)},$$

$$0 = \Gamma_{21} Q_{12}^{(0)} - \Omega Q_{32}^{(0)} + \Omega^* Q_{13}^{(0)}.$$

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

$$\left( \begin{array}{cccccc} 1 & 1 & 1 & 0 & 0 & 0 \\ 0 & \gamma_1 & i \Omega & -i \Omega^* & 0 & 0 \\ 0 & 0 & \gamma_2 & i \Omega & -i \Omega^* & 0 \\ -\Omega^* & 0 & \Omega^* & -\Gamma_p & 0 & 0 \\ 0 & -\Omega^* & \Omega^* & 0 & 0 & \Gamma_\alpha \\ 0 & 0 & 0 & -\Omega & 0 & 0 \end{array} \right) \left( \begin{array}{c} Q_{11}^{(0)} \\ Q_{22}^{(0)} \\ Q_{33}^{(0)} \\ Q_{31}^{(0)} \\ Q_{32}^{(0)} \\ Q_{33}^{(0)} \end{array} \right) = \left( \begin{array}{c} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{array} \right).$$

Taking $\gamma_1 = \gamma_2 = \gamma_3 = \gamma$ 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

$$0 = Q_{11}^{(1)} + Q_{22}^{(1)} + Q_{33}^{(1)},$$

$$m_1 = i \gamma Q_{33}^{(1)} + \Omega^* Q_{13}^{(1)} - \Omega Q_{31}^{(1)},$$

$$m_2 = i \gamma Q_{33}^{(1)} + \Omega^* Q_{23}^{(1)} - \Omega Q_{32}^{(1)},$$

$$m_3 = \Gamma_p Q_{31}^{(1)} + \Omega^* (Q_{11}^{(1)} - Q_{33}^{(1)}) + \Omega^* Q_{21}^{(1)},$$

$$m_4 = \Gamma_\alpha Q_{32}^{(1)} - \Omega^* Q_{12}^{(1)} - \Omega^* (Q_{22}^{(1)} - Q_{33}^{(1)}),$$

$$m_5 = \Gamma_{21} Q_{23}^{(1)} - \Omega^* Q_{23}^{(1)} + \Omega Q_{31}^{(1)},$$

where we have set

$$m_1 = e^{-i \beta t} B Q_{31}^{(0)} - e^{i \beta t} B^+ Q_{13}^{(0)},$$

$$m_2 = e^{i \beta t} A Q_{32}^{(0)} - e^{-i \beta t} A^+ Q_{23}^{(0)},$$

$$m_3 = -e^{-i \beta t} A^+ Q_{21}^{(0)} - e^{i \beta t} B^+ (Q_{11}^{(0)} - Q_{33}^{(0)}),$$

$$m_4 = e^{i \beta t} B^+ Q_{12}^{(0)} + e^{-i \beta t} A^+ (Q_{22}^{(0)} - Q_{33}^{(0)}),$$

$$m_5 = e^{i \beta t} B^+ Q_{23}^{(0)} - e^{-i \beta t} A Q_{31}^{(0)}.$$
The solutions of $Q_{jk}^{(i)}$ ($j, k = 1, 2, 3$) are given by
\[
\begin{pmatrix}
Q_{11}^{(i)} \\
Q_{21}^{(i)} \\
Q_{31}^{(i)} \\
Q_{12}^{(i)} \\
Q_{22}^{(i)} \\
Q_{32}^{(i)} \\
Q_{13}^{(i)} \\
Q_{23}^{(i)} \\
Q_{33}^{(i)}
\end{pmatrix} = U_1^{-1}
\begin{pmatrix}
0 \\
m_1 \\
m_2 \\
m_3 \\
m_4 \\
m_5 \\
m_6 \\
m_7 \\
m_8
\end{pmatrix},
\tag{C3}
\]
where
\[
U_1 = \begin{pmatrix}
1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & \Omega^* & -\Omega & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & 0 & 0 & \Omega^* & -\Omega & 0 & 0 \\
\Omega^* & 0 & -\Omega^* & 0 & (-\Delta + i\gamma) & 0 & 0 & -\Omega^* & 0 \\
0 & -\Omega^* & \Omega^* & 0 & 0 & (\delta + \Delta - i\gamma) & 0 & 0 & 0 \\
0 & 0 & 0 & \Omega & -\Omega^* & 0 & 0 & \delta & 0 \\
0 & -\Omega & 0 & (-\Delta - i\gamma) & 0 & 0 & 0 & 0 & \Omega \\
0 & -\Omega & 0 & 0 & (\delta + \Delta + i\gamma) & 0 & -\Omega & 0 & 0 \\
0 & 0 & 0 & \Omega^* & 0 & 0 & -\Omega & 0 & \delta
\end{pmatrix}
\tag{C4}
\]
and
\[
N = \det U_1 = 2\gamma^2|\Omega|^2[(2\Delta^2 + (\delta + 2\Delta)\delta + 2\gamma^2)|\Omega|^2 + 8|\Omega|^4].
\tag{C5}
\]
We write $Q_{31}^{(i)}$ and $Q_{23}^{(i)}$ as
\[
Q_{31}^{(i)} = \frac{D_1}{N}, \quad Q_{23}^{(i)} = \frac{E_1}{N},
\tag{C6}
\]
where
\[
D_1 = \det \begin{pmatrix}
1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & \Omega^* & m_1 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & 0 & m_2 & \Omega^* & -\Omega & 0 & 0 \\
\Omega^* & 0 & -\Omega^* & 0 & m_3 & 0 & 0 & \Omega^* & 0 \\
0 & -\Omega^* & \Omega^* & 0 & m_4 & 0 & (\delta + \Delta - i\gamma) & 0 & -\Omega^* \\
0 & 0 & 0 & m_5 & -\Omega^* & 0 & 0 & \delta & 0 \\
\Omega & 0 & -\Omega & (-\Delta - i\gamma) & m_6 & 0 & 0 & 0 & \Omega \\
0 & -\Omega & \Omega & 0 & m^*_4 & (\delta + \Delta + i\gamma) & 0 & -\Omega & 0 \\
0 & 0 & 0 & \Omega^* & m^*_5 & 0 & -\Omega & 0 & \delta
\end{pmatrix}
\tag{C7}
\]
and
\[
E_1 = \det \begin{pmatrix}
1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & \Omega^* & -\Omega & m_1 & 0 & 0 & 0 \\
0 & 0 & i\gamma & 0 & m_2 & -\Omega & 0 & 0 & 0 \\
\Omega^* & 0 & -\Omega^* & 0 & (-\Delta + i\gamma) & m_3 & 0 & \Omega^* & 0 \\
0 & -\Omega^* & \Omega^* & 0 & m_4 & (\delta + \Delta - i\gamma) & 0 & -\Omega^* & 0 \\
0 & 0 & 0 & m_5 & \delta & 0 & 0 & 0 & 0 \\
\Omega & 0 & -\Omega & (-\Delta - i\gamma) & 0 & m^*_3 & 0 & \Omega & 0 \\
0 & -\Omega & \Omega & 0 & 0 & m^*_4 & 0 & -\Omega & 0 \\
0 & 0 & 0 & \Omega^* & 0 & m^*_5 & -\Omega & 0 & \delta
\end{pmatrix}
\tag{C8}
\]
Keeping the terms including only single operator and satisfying the phase-matching condition, we write
\[
D_1 \rightarrow \gamma A e^{i\delta \mu_n}, \quad E_1 \rightarrow -\Omega^2 \gamma B^* e^{i\delta \mu_n},
\tag{C9}
\]
where

$$\mu_a = -i Q_{12}^{(0)}[\delta^2(\Delta^2 + \gamma^2 + (\delta + 2\Delta)\delta) + 2\delta(\delta - 2\Delta)\Omega]\Omega^2 + 4|\Omega|^4] + (Q_{13}^{(0)} - Q_{33}^{(0)}[i\delta^2(\Delta^2 + \gamma^2) - (\gamma - i\Delta)\delta^3 + 2\delta(2\gamma + i\delta))\Omega^2 - 4i|\Omega|^4]\Omega^2 = (Q_{23}^{(0)}[\delta^2(\gamma - i\Delta)(\Delta^2 + 2\Delta\delta + \gamma^2 + \delta^2) + (2i\Delta^2\delta - 2\Delta\gamma\delta + i\Delta\delta^2 - 3\gamma\delta^2 - i\delta^3)\Omega^2 + 4(\gamma + 2i\delta))\Omega^4]} - \Omega Q_{23}^{(0)}[2i\Delta^2\delta + 2\Delta\gamma\delta + 3i\Delta\delta^2 + 4i\gamma^2\delta - \gamma^2\delta^2 + i\delta^3 + 4(-i\Delta + 2\gamma + i\delta))\Omega^2]. \tag{C10}$$

and

$$\mu_b = (Q_{11}^{(0)} - Q_{22}^{(0)}[\delta^2[i(\Delta^2 + \gamma^2) - (\gamma - i\Delta)\delta]) + 2\delta(2\gamma + i\delta))\Omega^2 - 4i|\Omega|^4) - i Q_{12}^{(0)}[(\Delta^2 + \gamma^2)\delta^2 + 2(3\delta + 2\Delta)\delta\Omega^2 + 4|\Omega|^4] - \frac{Q_{13}^{(0)}}{\Omega}[\delta^2(\Delta^2 + \gamma^2)i(\gamma + i\Delta + i\delta)] + \delta(2i\Delta^2\delta + 2\Delta\gamma\delta + 3i\Delta\delta^2 - 3\gamma\delta^2 - i\delta^3)\Omega^2 + 4(\gamma + 2i\delta))\Omega^4]} + Q_{23}^{(0)}[\delta(2i\Delta^2 - 2\Delta\gamma + i\Delta\delta + 4i\gamma^2 - 3\gamma\delta) + 4(2\gamma + 2i\delta + i\Delta))\Omega^2]. \tag{C11}$$

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

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

$$\mu_b = \frac{2\gamma\delta^2}{G_2}[\delta^2(\Delta^2 + \gamma^2)(i\gamma - \Delta - \delta) + \delta(4\gamma^2 - 2i\gamma\delta - 4i\Delta\gamma - \delta^2 - 4i\Delta\delta)\Omega^2 - 2(2i\gamma - 6\Delta - 5\delta)\Omega^4]. \tag{C12}$$

where

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

Finally, Eq. (C9) becomes

$$D_1 \rightarrow \gamma A e^{i\delta}(\Omega)^2 \mu_a = -A e^{i\delta}(\Omega)^2 \frac{2\gamma\delta^2}{G_2}[\delta^2(-\Delta - i\gamma)(\Delta^2 + 2\Delta\delta + \gamma^2 + \delta^2)] - \delta(4\gamma^2 - 2i\gamma\delta - 4i\Delta\gamma + 3\delta^2 + 4i\Delta\delta)\Omega^2 + 2(2i\gamma + 6\Delta + \delta)\Omega^4]. \tag{C14}$$

$$E_1 \rightarrow -\Omega^2 B^+ e^{i\delta}\mu_b = -B^+ e^{i\delta}\Omega^2 \frac{2\gamma\delta^2}{G_2}[\delta^2(\Delta^2 + \gamma^2)(-\Delta + i\gamma - \delta)] + \delta(4\gamma^2 - 2i\gamma\delta + 4i\Delta\gamma - \delta^2 - 4i\Delta\delta)\Omega^2 - 2(2i\gamma - 6\Delta - 5\delta)\Omega^4]. \tag{C15}$$

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

$$Be^{-i\delta}Q_{31}^{(1)} + A^+ e^{-i\delta}Q_{23}^{(1)} + H.c. = B e^{-i\delta} D_1 \frac{A}{N} + A^+ e^{-i\delta} E_1 \frac{B}{N} + H.c. \rightarrow \Lambda_a AB + \Lambda_b A^+ B^+ + H.c.$$

$$= (\Lambda_a + \Lambda_b^+)AB + (\Lambda_a^+ + \Lambda_b)A^+ B^+ = \Lambda AB + \Lambda^+ A^+ B^+. \tag{C16}$$

where we have set

$$\Lambda_a = \frac{2\delta^2\gamma^2}{N G_2}(\Omega^2)[\delta^2(-\Delta - i\gamma)(\Delta^2 + 2\Delta\delta + \gamma^2 + \delta^2) - \delta(4\gamma^2 - 2i\gamma\delta - 4i\Delta\gamma + 3\delta^2 + 4i\Delta\delta)\Omega^2 + 2(2i\gamma + 6\Delta + \delta)\Omega^4],$$

$$\Lambda_b = \frac{2\delta^2\gamma^2}{N G_2}(\Omega^2)[\delta^2(\Delta^2 + \gamma^2)(-\Delta + i\gamma - \delta) + \delta(4\gamma^2 - 2i\gamma\delta + 4i\Delta\gamma - \delta^2 - 4i\Delta\delta)\Omega^2 - 2(2i\gamma - 6\Delta - 5\delta)\Omega^4]. \tag{C17}$$

$$\Lambda = \Lambda_a + \Lambda_b^* = \frac{2\delta^2\gamma^2}{N G_2}(\Omega^2)[\delta^2(2\gamma^2 - i\gamma\delta + (\delta + 2\Delta)(\delta + \Delta))(\gamma - i\Delta) + 4\delta^2(\delta + 2\Delta)\Omega^2 - 4(2i\gamma + 6\Delta + 3\delta)\Omega^4]. \tag{C18}$$

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

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

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

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

(C20)

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

\[
\begin{align*}
n_1 &= e^{-i\delta t} B Q_{31}^{(1)} - e^{i\delta t} B^+ Q_{13}^{(1)}, \\
n_2 &= e^{i\delta t} A Q_{32}^{(1)} - e^{-i\delta t} A^+ Q_{23}^{(1)}, \\
n_3 &= -e^{-i\delta t} A^+ Q_{11}^{(1)} - e^{i\delta t} B^+ (Q_{11}^{(1)} - Q_{33}^{(1)}), \\
n_4 &= e^{i\delta t} B^+ Q_{22}^{(1)} + e^{-i\delta t} A^+ (Q_{22}^{(1)} - Q_{33}^{(1)}), \\
n_5 &= e^{i\delta t} B^+ Q_{23}^{(1)} - e^{i\delta t} A Q_{31}^{(1)}.
\end{align*}
\]

(C21)

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

\[
Q_{31}^{(2)} = H_{31}^{(2)}N, \quad Q_{23}^{(2)} = H_{23}^{(2)}N,
\]

(C22)

where

\[
H_{31}^{(2)} = \begin{vmatrix}
1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & \Omega^* & n_1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & 0 & n_2 & \Omega^* & -\Omega & 0 & 0 & 0 \\
\Omega^* & 0 & -\Omega^* & 0 & n_3 & 0 & 0 & \Omega^* & 0 & 0 \\
0 & -\Omega^* & \Omega^* & 0 & n_4 & 0 & (\delta + \Delta - i\gamma) & 0 & -\Omega^* & 0 \\
0 & 0 & 0 & 0 & n_5 & -\Omega^* & 0 & \delta & 0 & 0 \\
\Omega & 0 & -\Omega & (-\Delta - i\gamma) & n_3^* & 0 & 0 & 0 & \Omega & 0 \\
0 & -\Omega & \Omega & 0 & n_4^* & (\delta + \Delta + i\gamma) & 0 & -\Omega & 0 & 0 \\
0 & 0 & 0 & \Omega^* & n_5^* & 0 & -\Omega & 0 & \delta & 0
\end{vmatrix},
\]

(C23)

\[
H_{23}^{(2)} = \begin{vmatrix}
1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & \Omega^* & -\Omega & n_1 & 0 & 0 & 0 & 0 \\
0 & 0 & i\gamma & 0 & 0 & n_2 & -\Omega & 0 & 0 & 0 \\
\Omega^* & 0 & -\Omega^* & 0 & (-\Delta + i\gamma) & n_3 & 0 & \Omega^* & 0 & 0 \\
0 & -\Omega^* & \Omega^* & 0 & 0 & n_4 & (\delta + \Delta - i\gamma) & 0 & -\Omega^* & 0 \\
0 & 0 & 0 & 0 & \Omega & n_5 & 0 & \delta & 0 & 0 \\
\Omega & 0 & -\Omega & (-\Delta - i\gamma) & 0 & n_3^* & 0 & 0 & \Omega & 0 \\
0 & -\Omega & \Omega & 0 & 0 & n_4^* & 0 & -\Omega & 0 & 0 \\
0 & 0 & 0 & \Omega^* & 0 & n_5^* & -\Omega & 0 & \delta & 0
\end{vmatrix}.
\]

(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

$$\Omega Q_{31}^{(2)} = \Omega \frac{H_{31}^{(2)}}{N} \rightarrow \mu_1 AB + \mu_2 A^+ B^+, \quad \text{(C25)}$$

$$\Omega^* Q_{31}^{(2)} = \Omega^* \frac{H_{31}^{(2)}}{N} \rightarrow \mu_3 AB + \mu_4 A^+ B^+, \quad \text{(C26)}$$

where we have set

$$\mu_1 = -8\gamma^6\delta^4|\Omega|^2 \frac{\delta^5[\gamma + i(\delta + \Delta)]^2(\gamma^2 + \Delta^2)}{N^3}$$

$$\mu_2 = -8\gamma^6\delta^2|\Omega|^2 \frac{(\delta^5[\gamma^2 + (\delta + \Delta)]^2(\gamma^2 + \Delta^2)}{N^3}$$

$$\mu_3 = -8\gamma^6\delta^4|\Omega|^2 \frac{(\delta^5[\gamma^2 + (\delta + \Delta)]^2(\gamma^2 + \Delta^2)}{N^3}$$

$$\mu_4 = -8\gamma^6\delta^4|\Omega|^2 \frac{(\delta^5[\gamma^2 + (\delta + \Delta)]^2(\gamma^2 + \Delta^2)}{N^3}$$

Thus substituting Eqs. (C25) and (C26) into Eq. (15), we have

$$H_V \rightarrow \text{Be}^{-i\theta} Q_{31}^{(1)} + \Omega Q_{31}^{(1)} + A^+ B^+ + \text{Be}^{-i\theta} Q_{32}^{(1)} + \Omega^* Q_{32}^{(1)} + \text{H.c.}$$

$$= \Lambda AB + \Lambda^* A^+ B^+ + [\mu_1 AB + \mu_2 A^+ B^+] + \text{H.c.}$$

$$= (\Lambda + \mu_1 + \mu_3 + \mu_4) AB + \text{H.c.} = \lambda_z AB + \lambda_z^* A^+ B^+, \quad \text{(C30)}$$

where

$$\lambda_z = \Lambda + \mu_1 + \mu_3 + \mu_4 + \mu_4^*$$

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

$$- \frac{16\delta^2(\delta + 2\Delta)}{G_2^3} \left\{ (\gamma^2 + (\delta + \Delta)^2)[\gamma^2 + \Delta^2 |\Omega|^2 \Omega^*] + (\delta^2 - 2\gamma^2 + 6\delta \Delta + 6\Delta^2)(\Omega^*)^2 \right\}.$$

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

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

$$G_2 \approx \delta^2(\delta^2 + 2\Delta \delta + 2\Delta^2 + 2\gamma^2). \quad \text{(C33)}$$
We only examine the phase-matching items, and notice $$\lfloor \Omega \rceil e^{i\bar{\chi}_{\ell,p}\tau} + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor$$

$$\lfloor \Omega \rceil e^{i\bar{\chi}_{\ell,p}\tau} + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor = \lfloor \Omega \rceil e^{i\bar{\chi}_{\ell,p}\tau} + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor$$

\(d_{31}^{*}d_{31}^{*} + d_{31}^{*}d_{31}^{*} + d_{31}^{*}d_{31}^{*} + d_{31}^{*}d_{31}^{*} + d_{31}^{*}d_{31}^{*} + d_{31}^{*}d_{31}^{*} + d_{31}^{*}d_{31}^{*} + d_{31}^{*}d_{31}^{*}
\)

\[D1\]

\(|\Omega \rceil e^{i\bar{\chi}_{\ell,p}\tau} + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor^2 = |\Omega \rceil^2 + |\Omega_{2}\rceil^2 + \Omega_{2} e^{i\bar{\chi}_{\ell,p}\tau}\rfloor + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor^2.
\]

\(D2\)

We only examine the phase-matching items, and notice $$\lfloor \Omega \rceil e^{i\bar{\chi}_{\ell,p}\tau} + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor$$

\(\Omega_{1} e^{i\bar{\chi}_{\ell,p}\tau} + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor^2 = |\Omega_{1}\rceil^2 + |\Omega_{2}\rceil^2.
\]

\(D4\)

\(\Omega_{1} e^{i\bar{\chi}_{\ell,p}\tau} + \Omega_{2} e^{-i\bar{\chi}_{\ell,p}\tau}\rfloor^2 = |\Omega_{1}\rceil^2 + |\Omega_{2}\rceil^2.
\]

\(D5\)

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

$$H_{\text{eff}} = \kappa_{1}(a^{+}b^{+} + c^{+}d^{+} + 2\sqrt{\kappa_{1}k_{2}}(a^{+}b^{+} + b^{+}c^{+}) + H.c.,
\]

\(D6\)

where $$\kappa_{1}$$ and $$\kappa_{2}$$ 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.