Narrowband-biphoton generation due to long-lived coherent population oscillations

A. V. Sharypov and A. D. Wilson-Gordon
Department of Chemistry, Bar-Ilan University, Ramat Gan 52900, Israel

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We study the generation of paired photons due to the effect of four-wave mixing in an ensemble of pumped two-level systems that decay via an intermediate metastable state. The slow population relaxation of the metastable state creates long-lived coherent population oscillations, leading to a narrowband nonlinear response of the medium which determines the narrow spectral width and long coherence time of the biphotons.

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I. INTRODUCTION

Traditionally, paired photons are produced from spontaneous parametric down conversion in nonlinear crystals. The bandwidth of such biphotons is very broad and typically in the terahertz range [1], which makes them useless for some applications in quantum information science that require a strong interaction between photons and atomic systems. This problem can be overcome by generating biphotons in cold atomic systems which have a narrowband nonlinear response. For example, biphotons can be produced when a double-Λ system [2–4] or two-level system (TLS) [5–7] is pumped by two counterpropagating laser fields. Then, phase-matched and energy-time entangled photon pairs are produced due to the effect of four-wave mixing (FWM). Biphotons from such a source have a bandwidth in the megahertz range and a coherence time of hundreds of nanoseconds.

In this paper, we demonstrate that narrowband biphotons can also be produced due to the effect of long-lived coherent population oscillations (CPOs) in a TLS with an intermediate metastable state. In such a system, the width of the nonlinear response is determined by the lifetime of the metastable state, which can vary significantly depending on the nature of the quantum system. For example, in semiconductor quantum wells and dots [8], the CPO lifetime is in the microsecond range, whereas in a ruby crystal [9,10], color centers in a diamond [10–12], or organic film [13], it can be more than a millisecond, leading to a broad range of potential applications of photon pairs based on the CPO effect.

The theoretical model is based on the Heisenberg equations. In order to find the emission spectrum and biphoton wave function, we calculate the first- and second-order correlation functions. Contributions from the correlation of both the boundary operators and the Langevin noise operators have been taken into account, as both of these correlations can contribute to the final result [14].

II. THE MODEL

We consider the interaction of an ensemble of TLSs that decay via a single intermediate metastable state with two counterpropagating pump fields with amplitude E_0 (see Fig. 1). The medium is assumed to be optically thin in the direction of pump propagation and the effect of pump depletion is not taken into account. Due to pumping of the TLS by two counterpropagating laser fields, photon pairs are produced [5–7] where photons from the same pair also counterpropagate [see Fig. 1(a)] so that the phase-matching condition of FWM is satisfied [15] (actually, in such a configuration, biphotons are emitted into the whole 4π space). In order to allow for the spontaneous initiation process, the generated weak fields are described by quantum-mechanical operators $\widehat{E}_{1,2} = E_v \sum_{\mathbf{k}} \widehat{a}_{\mathbf{k}}^{1,2} e^{i\mathbf{k}\cdot\mathbf{r}}$, where the subscript 1 denotes the field at frequency $\omega_1 = \omega_0 + \delta$ propagating along the positive z axis, the subscript 2 denotes the field at frequency $\omega_2 = \omega_0 - \delta$ moving along the negative z axis [see Fig. 1(a)], $E_v = (\hbar\omega_0/2\varepsilon_0 V)^{1/2}$ is the vacuum field, V is the quantization volume, and $\widehat{a}$ is the photon annihilation operator.

To describe the evolution of the atomic ensemble, we begin with the Heisenberg equations of motion in the dipole approximation for the mean values of the atomic operators:

$$
\begin{aligned}
(d/dt + \Gamma_{ba} - i\omega_{ba})\langle\widetilde{\sigma}_{ab}\rangle &= -i\frac{d_{ba}}{\hbar}\widetilde{E}^{(+)}(\langle\widetilde{\sigma}_{bb}\rangle - \langle\widetilde{\sigma}_{aa}\rangle),\\
(d/dt + \Gamma_{ba} + i\omega_{ba})\langle\widetilde{\sigma}_{ba}\rangle &= i\frac{d_{ab}}{\hbar}\widetilde{E}^{(-)}(\langle\widetilde{\sigma}_{bb}\rangle - \langle\widetilde{\sigma}_{aa}\rangle),\\
d\langle\widetilde{\sigma}_{aa}\rangle/dt &= \gamma_{ba}\langle\widetilde{\sigma}_{bb}\rangle + \gamma_{ca}\langle\widetilde{\sigma}_{cc}\rangle\\
&\quad - i\frac{d_{ab}}{\hbar}\widetilde{E}^{(+)}\langle\widetilde{\sigma}_{ba}\rangle + i\frac{d_{ba}}{\hbar}\widetilde{E}^{(-)}\langle\widetilde{\sigma}_{ab}\rangle,\\
(d/dt + \gamma_b)\langle\widetilde{\sigma}_{bb}\rangle &= i\frac{d_{ab}}{\hbar}\widetilde{E}^{(+)}\langle\widetilde{\sigma}_{ba}\rangle - i\frac{d_{ba}}{\hbar}\widetilde{E}^{(-)}\langle\widetilde{\sigma}_{ab}\rangle,\\
(d/dt + \gamma_{ca})\langle\widetilde{\sigma}_{cc}\rangle &= \gamma_{bc}\langle\widetilde{\sigma}_{bb}\rangle,
\end{aligned}
\tag{1}
$$

where $\langle\widetilde{\sigma}_{ij}\rangle = \langle|i\rangle\langle j|\rangle$ is the mean value of the atomic operator, $\widetilde{E}^{(\pm)}$ is the total field operator, d_{ij} is the transition dipole matrix element, Γ_{ba} is the transverse relaxation rate, γ_{ij} is the longitudinal decay rate from the state $|i\rangle$ to the state $|j\rangle$, and $\gamma_b = \gamma_{bc} + \gamma_{ba}$ is the total decay rate from the excited level. We also assume that the system is closed, so that $\langle\widetilde{\sigma}_{aa}\rangle + \langle\widetilde{\sigma}_{bb}\rangle + \langle\widetilde{\sigma}_{cc}\rangle = 1$.

We apply the slowly varying envelope approximation and write the total field operator as $\widetilde{E}^{(+)} = (E_0 + \widehat{E}_1 e^{-i\delta t} + \widehat{E}_2 e^{i\delta t})e^{i\omega_0 t}$ and $\widetilde{E}^{(-)} = (E_0 + \widehat{E}_1^{\dagger} e^{i\delta t} + \widehat{E}_2^{\dagger} e^{-i\delta t})e^{-i\omega_0 t}$. To eliminate the fast oscillating terms in Eqs. (1), we introduce the transformations $\langle\widetilde{\sigma}_{ab,ba}(t)\rangle = \langle\sigma_{ab,ba}(t)\rangle e^{\pm i\omega_0 t}$ and $\langle\widetilde{\sigma}_{jj}(t)\rangle = \langle\sigma_{jj}(t)\rangle|_{j=a,b,c}$. As several fields are applied to the same transition, the Hamiltonian of the system has a periodic time dependence [7,16–20], and the Floquet theory [21] can therefore be applied to $\langle\sigma\rangle$:

$$
\langle\sigma\rangle = \langle\sigma^{(0)}\rangle + \langle\sigma^{(+\delta)}\rangle e^{-i\delta t} + \langle\sigma^{(-\delta)}\rangle e^{i\delta t}. \tag{2}
$$

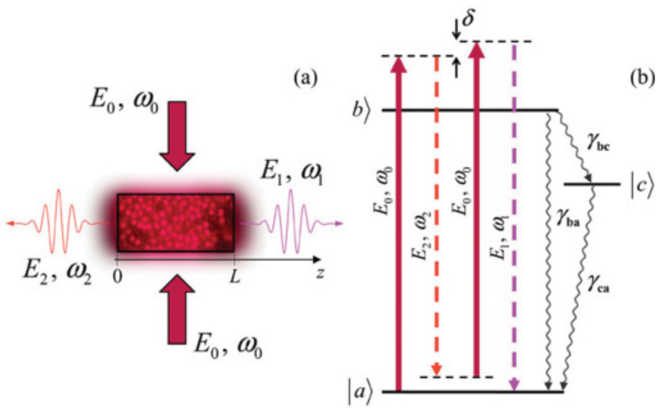

FIG. 1. (Color online) In the presence of the counterpropagating pump fields, phase-matched counterpropagating biphotons are generated inside the medium due to FWM.

The zeroth-order solutions of Eqs. (1) give the response of the medium to the pump field and the population distribution between the quantum states, whereas the first-order solutions determine the medium response to the weak generated fields since they are related to the medium polarizations at frequencies $\omega_{1,2}$ by $P_1 = N d_{ba}\langle\sigma_{ab}^{(+\delta)}\rangle$ and $P_2^* = N d_{ab}\langle\sigma_{ba}^{(+\delta)}\rangle$ [16].

The pump-probe interaction with a TLS is characterized by population beating or coherent population oscillations (CPOs) at δ, which is the frequency difference between pump and probe fields [16]. In an ordinary TLS, the CPOs decay at the same rate as the excited state. However, the situation can be quite different if an intermediate metastable state is included [see Fig. 1(b)]. In this case, long-lived CPOs of the metastable and ground states can be created, which lead to the appearance of a narrow dip in the probe absorption spectrum and a narrow peak in the FWM spectrum [17–19].

Then, making the appropriate approximations for a TLS that decays slowly via a metastable state,

$$\gamma_{ca} \ll \gamma_{ba} \ll \gamma_{bc} \text{ and } V_0 \ll \Gamma_{ba}, \tag{3}$$

where $V_0 = d_{ba}E_0/\hbar$ is the pump Rabi frequency which is assumed to be real, and solving Eqs. (1), we obtain [19,20]

$$\langle\sigma_{ab}^{(+\delta)}(\delta)\rangle = (\alpha_1 \widehat{E}_1 + \beta_1 \widehat{E}_2^\dagger) d_{ab}/\hbar, \tag{4a}$$
$$\langle\sigma_{ba}^{(+\delta)}(\delta)\rangle = (\alpha_2 \widehat{E}_2^\dagger + \beta_2 \widehat{E}_1) d_{ba}/\hbar, \tag{4b}$$

where $\alpha_{1,2}$ are proportional to the effective linear susceptibilities, and $\beta_{1,2}$ are proportional to the effective third-order nonlinear susceptibilities and are responsible for the generation of the paired photons. They are given by

$$\alpha_{1,2} \equiv \pm i(1 + X_{1,2})/[\Gamma_{1,2}(1+\kappa)], \tag{4c}$$
$$\beta_{1,2} \equiv \pm i X_{2,1}/[\Gamma_{1,2}(1+\kappa)], \tag{4d}$$

where

$$X_{1,2} \equiv -\kappa\gamma_{ca}\frac{1 \pm \Omega\delta/\Gamma\Gamma_{1,2}}{W - i\delta} \tag{5a}$$

is the coherent field interaction term;

$$W = (1+\kappa)\gamma_{ca} \tag{5b}$$

determines the characteristic width of the window in which coherent interaction between the fields occurs;

$$\kappa \equiv 2V_0^2/\left[\gamma_{ca}\Gamma_{ba}\left(1 + \Omega^2/\Gamma_{ba}^2\right)\right] \tag{5c}$$

is the saturation parameter;

$$\Gamma_{1,2} \equiv \Gamma_{ba} \pm i(\Omega \mp \delta); \tag{5d}$$

and $\Omega = \omega_0 - \omega_{ba}$ is the detuning of the pump from resonance.

The evolution of the fields generated in the medium is described by the coupled propagation equations [4]

$$\left(\frac{\partial}{\partial ct} + \frac{\partial}{\partial z}\right)\widehat{E}_1(z,t) = i\epsilon_1\sigma_{ab}^{(+\delta)}(z,t), \tag{6a}$$
$$\left(\frac{\partial}{\partial ct} - \frac{\partial}{\partial z}\right)\widehat{E}_2^\dagger(z,t) = -i\epsilon_2\sigma_{ba}^{(+\delta)}(z,t), \tag{6b}$$

where $\epsilon_1 \approx \epsilon_2 = \epsilon = N d_{ba}\omega_0/2c\varepsilon_0$ are the propagation constants, and

$$\sigma_{ab,ba}^{(+\delta)}(\delta) = \langle\sigma_{ab,ba}^{(+\delta)}(\delta)\rangle + F_{ab,ba}^{(+\delta)}(\delta) \tag{7}$$

is the atomic operator that can be split into the sum of its average value [determined by Eqs. (4a) and (4b)] and the deviation from the average, which is the Langevin noise [28], which should be taken into account in order to describe the spontaneous emission correctly.

In order to solve these coupled equations, we make a Fourier transformation, neglecting the term $i\omega/c$ as it does not affect the final result [4], substitute Eq. (7) into Eqs. (6a) and (6b), and write the system of differential equations in the matrix form

$$\partial E/\partial\widetilde{z} = ME + F, \tag{8}$$

where we introduce the field vectors $E \equiv (\widehat{E}_1, \widehat{E}_2^\dagger)^T$, the noise term vector $F \equiv (F_{ab}^{(+\delta)}, F_{ba}^{(+\delta)})^T$, $\widetilde{z} = z d_{ba}\epsilon/\hbar$, and

$$M \equiv i\begin{pmatrix}\alpha_1 & \beta_1\\ \beta_2 & \alpha_2\end{pmatrix}. \tag{9}$$

The formal solution of Eq. (8) can be written in the form

$$E(\widetilde{L}) = e^{M\widetilde{L}}E(0) + \int_0^{\widetilde{L}} d\widetilde{z}\, e^{M(\widetilde{L}-\widetilde{z})}F(\widetilde{z}), \tag{10}$$

where $\widetilde{L} = L d_{ba}\epsilon/\hbar$. The generated biphotons counterpropagate so that photon "1" leaves the medium at the point $z = L$ and photon "2" leaves at $z = 0$. The boundary conditions derive from the vacuum field fluctuations at $z = 0$ for photon 1 and at $z = L$ for photon 2 [see Fig. 1(a)], so that Eq. (10) can be rewritten as

$$\begin{pmatrix}\widehat{E}_1(\widetilde{L})\\ \widehat{E}_2^\dagger(0)\end{pmatrix} = \begin{pmatrix}A_1 & B_1\\ B_2 & A_2\end{pmatrix}\begin{pmatrix}\widehat{E}_1(0)\\ \widehat{E}_2^\dagger(\widetilde{L})\end{pmatrix} + \int_0^{\widetilde{L}} d\widetilde{z}\begin{pmatrix}P_{11} & P_{12}\\ P_{21} & P_{22}\end{pmatrix}F(\widetilde{z}), \tag{11}$$

where the connection between coefficients $A_{1,2}, B_{1,2}, P_{ij}$, and matrix $\begin{pmatrix}\tilde{A}_1 & \tilde{B}_1\\ \tilde{B}_2 & \tilde{A}_2\end{pmatrix} = e^{M\widetilde{L}}$ is as follows:

$$\begin{pmatrix} A_1 & B_1 \\ B_2 & A_2 \end{pmatrix} = \begin{pmatrix} \tilde{A}_1 - \frac{\tilde{B}_1\tilde{B}_2}{\tilde{A}_2} & \frac{\tilde{B}_1}{\tilde{A}_2} \\ -\frac{\tilde{B}_2}{\tilde{A}_2} & \frac{1}{\tilde{A}_2} \end{pmatrix}, \tag{12}$$

$$\begin{pmatrix} P_{11} & P_{12} \\ P_{21} & P_{22} \end{pmatrix} = \begin{pmatrix} 1 & -\frac{\tilde{B}_1}{\tilde{A}_2} \\ 0 & -\frac{1}{\tilde{A}_2} \end{pmatrix} e^{M(\widetilde{L}-\widetilde{z})}. \tag{13}$$

III. THE CORRELATION PROPERTIES OF THE EMITTED PHOTONS

The spectrum of the emitted photons is described by the Fourier transform of the first-order correlation function $G^{(1)}_{1,2}(\tau)$ [22,23]:

$$R_{1,2}(\delta) = \int e^{i\delta\tau} G^{(1)}_{1,2}(\tau) d\tau = \frac{c}{LE_v} \int e^{i\delta\tau} \langle \widehat{E}^{\dagger}_{1,2}(t-\tau)\widehat{E}_{1,2}(t)\rangle d\tau. \tag{14}$$

By taking into account that the commutation relation for the input field operators can be written as $[\widehat{E}(\omega),\widehat{E}^{\dagger}(-\omega')]/E_v^2 = \delta(\omega+\omega')$ [23], and using the solution of Eq. (11), we obtain

$$R_{1,2}(\delta) = |B_{1,2}|^2 + D_{11,22}, \tag{15}$$

where the functions D_{ij} are the corresponding moments of the noise operators derived in the Appendix [see Eqs. (A1)–(A3)].

The correlation between the photons emitted to the left and right is described by the second-order Glauber correlation function $G^{(2)}_{21}$ [22],

$$G^{(2)}_{21}(\tau) = \langle \widehat{E}^{\dagger}_1(L,t)\widehat{E}^{\dagger}_2(0,t+\tau)\widehat{E}_2(0,t+\tau)\widehat{E}_1(L,t)\rangle. \tag{16}$$

As the field emitted by many statistically independent atoms behaves as a Gaussian random variable, we can use the Gaussian moment theorem [4,22] and rewrite Eq. (16) in the form

$$G^{(2)}_{21} = G^{(1)}_1(t)G^{(1)}_2(t+\tau) + |\Phi_{21}(\tau)|^2, \tag{17a}$$

where the first term describe the appearance of uncorrelated photons and can be found from Eq. (15), and the second term

$$\Phi_{21}(\tau) = \langle \widehat{E}_2(0,t+\tau)\widehat{E}_1(L,t)\rangle \tag{17b}$$

describes the appearance of entangled photon pairs and corresponds to the biphoton wave function [4,22,24]. Using the solution of Eq. (11), we find that

$$\Phi_{21}(\tau) = \int e^{-i\delta\tau}(A_2^* B_1 + D_{21}) d\delta, \tag{18}$$

where D_{21} is proportional to the Langevin noise terms and is given by Eq. (A3).

Harris and coworkers [2] have pointed out that a long coherence time for the biphotons can be obtained due to the slow light effect [25] experienced by one of the photons of the entangled pair but not by the other. Here we demonstrate that a long coherence time can be obtained even in an optically thin medium, where

$$\alpha_{1,2}\widetilde{L}, \beta_{1,2}\widetilde{L} \ll 1. \tag{19}$$

Under these conditions, Eqs. (12) and (13) simplify to

$$A_{1,2} = 1, \quad B_{1,2} = \pm i\beta_{1,2}\widetilde{L}, \tag{20a}$$

$$P_{11} = -P_{22} = 1, \quad P_{12} = -i\beta_1\widetilde{z}, \quad P_{21} = -i\beta_2(\widetilde{L}-\widetilde{z}), \tag{20b}$$

and it becomes possible to obtain analytical solutions for the spectrum and second-order correlation function of the biphotons.

The spectrum of the emitted photons is determined by the functions $R_{1,2}(\delta)$ [Eq. (15)], which consist of two terms, $|B_{1,2}|^2$ and $D_{11,22}$. The first term takes into account the correlation of the boundary operators due to the effect of FWM and, under the conditions of Eq. (19), can be written as

$$|B_{1,2}|^2 = \left(\frac{\widetilde{L}\gamma_{ca}\kappa}{\Gamma_{ba}(1+\kappa)}\right)^2 \frac{1}{1+\Omega^2/\Gamma_{ba}^2} \times \left|\frac{1}{W-i\delta} - \frac{1}{2}\left(\frac{1-i\Omega/\Gamma_{ba}}{\Gamma_1} + \frac{1+i\Omega/\Gamma_{ba}}{\Gamma_2}\right)\right|^2. \tag{21}$$

The second term is associated with the Langevin force and, under the conditions given by Eqs. (3) and (19), can be approximated as

$$D_{11,22} \approx \frac{\widetilde{L}\gamma_{ca}\kappa^2}{2\Gamma_{ba}(1+\kappa)^2} \mathrm{Re}\left\{\frac{1}{W-i\delta} - \frac{\gamma_{ca}/\Gamma_{ba}}{2(1+\Omega^2/\Gamma_{ba}^2)} \times \left[\frac{(1-i\Omega/\Gamma_{ba})^2}{\Gamma_1} + \frac{(1+i\Omega/\Gamma_{ba})^2}{\Gamma_2}\right]\right\}. \tag{22}$$

The spectra of the functions $|B_{1,2}|^2$ and $D_{11,22}$ have similar shapes: a narrow CPO peak of width W centered at $\delta = 0$, imposed on a weak naturally broadened pedestal, in the case of zero pump detuning. If the detuning is nonzero, this pedestal splits into two sidebands with the same width, located at the points $\delta = \pm\Omega$. However, as a result of Eqs. (3) and (19), the functions $D_{11,22}$ are much more intense than the functions $|B_{1,2}|^2$. Thus, the main contribution to the total spectrum under these conditions derives from the Langevin noise term.

It should be noted that similar results for the emission spectrum of the system with a shelving state were obtained in Refs. [26,27], in the context of quantum jumps.

We now turn to the calculation of the biphoton wave function [Eq. (17b)]. For an optically thin medium [Eq. (19)], Eq. (A3) simplifies to

$$D_{21} \approx \frac{\widetilde{L}\gamma_{ca}}{2\Gamma_{ba}}\frac{\kappa}{1+\kappa}\left\{\frac{\Gamma_{ba}-i\Omega}{\Gamma_{ba}+i\Omega} \times \left(\frac{\kappa}{1+\kappa}\frac{1}{W+i\delta} - \frac{1}{\Gamma_{ba}+i(\Omega+\delta)}\right)\right\}. \tag{23}$$

Then, substituting Eqs. (20a) and (23) into Eq. (18), and integrating over δ, we obtain

$$\Phi_{21}(\tau) = \eta\frac{\kappa}{1+\kappa}(B+N), \tag{24}$$

where

$$B \equiv i\frac{e^{-W|\tau|} - e^{-\Gamma_{ba}|\tau|}\left(\cos\Omega|\tau| - \frac{\Omega}{\Gamma_{ba}}\sin\Omega|\tau|\right)}{1+i\frac{\Omega}{\Gamma_{ba}}} \tag{25}$$

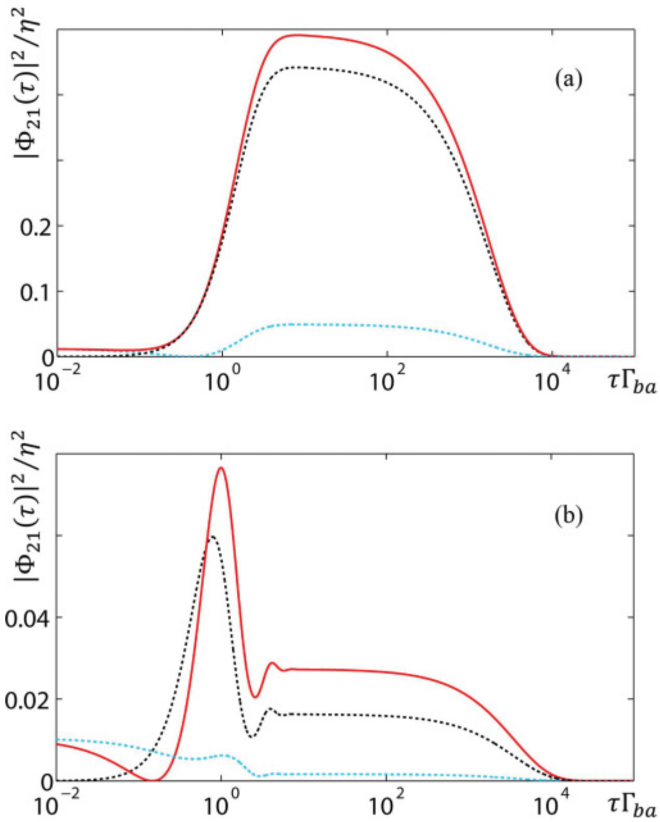

FIG. 2. (Color online) Biphoton wave function for $V_0/\Gamma_{ba} = 0.01$ and $\gamma_{ca}/\Gamma_{ba} = 10^{-4}$ with (a) zero pump detuning ($\kappa = 2$) and (b) $\Omega/\Gamma_{ba} = -2$ ($\kappa = 0.4$); total wave function (red solid line), contribution from B (black upper dashed line), and contribution from N (cyan lower dashed line).

takes into account the correlation between the boundary operators;

$$N \equiv \frac{1}{2}\frac{\left(1 - i\frac{\Omega}{\Gamma_{ba}}\right)^2}{1 + \left(\frac{\Omega}{\Gamma_{ba}}\right)^2}\left(e^{-W|\tau|}\frac{\kappa}{1+\kappa} - e^{-i\Omega|\tau|}e^{-\Gamma_{ba}|\tau|}\right) \tag{26}$$

is due to Langevin noise; and $\eta = 2\pi\widetilde{L}\gamma_{ca}$. The general dynamics of the B and N terms are the same (see Fig. 2): they are characterized by a long coherence time due to the term $e^{-W|\tau|}$, and an antibunching dip at $\tau = 0$ (in the case $\kappa \gg 1$), which arises from destructive interference between the terms $e^{-W|\tau|}$ and $e^{-\Gamma_{ba}|\tau|}$. In addition, during the initial period $1/\Gamma_{ba}$, both terms display beating at a frequency proportional to the pump detuning Ω. However, there are also some differences between B and N: the amplitudes of these two terms can be different, depending on the values of the pump detuning and saturation parameter κ. In addition, the term N does not go exactly to zero at the point $\tau = 0$, and the precise form of the beating for each of these terms will be different.In Fig. 2, we present a numerical calculation of Eq. (24), and also compare the contributions from the terms B and N. It can be seen that for the specific parameters used in the calculation, the Langevin noise term has little influence on the total spectrum, but for another set of parameters, it could play a considerable role, especially beyond the optically thin medium approximation of Eq. (19).

One of the possible ways of realizing the narrowband biphotons experimentally is to use a ruby crystal which fits our theoretical model. Pumping the crystal with an Ar^+ laser, at the wavelength $\lambda = 514$ nm and intensity $I \approx 1\mathrm{kW/cm}^2$, would generate biphotons with approximate coherence time 1.5 ms and bandwidth 600 Hz.

IV. CONCLUSION

In summary, we have demonstrated that the combined effects of FWM and long-lived CPOs in a TLS with an intermediate metastable state are able to produce narrowband biphotons with a long coherence time whose maximum value is equal to the lifetime of the metastable state. The biphotons' wave form and bandwidth can be controlled by the pump intensity and detuning. During the time $1/\Gamma_{ba}$, the biphoton wave function shows antibunching behavior. If the pump field is detuned, damped oscillations are observed during this period.

The theoretical model presented here takes into account the contributions from the correlation of the boundary operators and the Langevin noise operators. For an optically thin medium, it was shown that the emission spectrum is determined only by the Langevin noise term, as the contribution from the boundary operators is negligible, but the contributions to the biphoton wave function from both of these terms are comparable.

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APPENDIX: NOISE CORRELATION

The substitution of Eq. (11) into Eqs. (14) and (17b) gives the noise-correlation terms

$$D_{11} = \int_0^{\widetilde{L}} d\widetilde{z}\big[K_{11}^{11}\langle F_{ba}^{(+\delta)}F_{ab}^{(0)}\rangle + K_{22}^{11}\langle F_{ab}^{(+\delta)}F_{ba}^{(0)}\rangle + K_{12}^{11}\langle F_{ba}^{(+\delta)}F_{ba}^{(0)}\rangle + \big(K_{12}^{11}\big)^*\langle F_{ab}^{(+\delta)}F_{ab}^{(0)}\rangle\big], \tag{A1}$$

$$D_{22} = \int_0^{\widetilde{L}} d\widetilde{z}\big[K_{11}^{22}\langle F_{ab}^{(-\delta)}F_{ba}^{(0)}\rangle + K_{22}^{22}\langle F_{ba}^{(-\delta)}F_{ab}^{(0)}\rangle + K_{12}^{22}\langle F_{ab}^{(-\delta)}F_{ab}^{(0)}\rangle + \big(K_{12}^{22}\big)^*\langle F_{ba}^{(-\delta)}F_{ba}^{(0)}\rangle\big], \tag{A2}$$

$$D_{21} = \int_0^{\widetilde{L}} d\widetilde{z}\big[K_{11}^{21}\langle F_{ba}^{(-\delta)}F_{ab}^{(0)}\rangle + K_{22}^{21}\langle F_{ab}^{(-\delta)}F_{ba}^{(0)}\rangle + K_{12}^{21}\langle F_{ba}^{(-\delta)}F_{ba}^{(0)}\rangle + K_{21}^{21}\langle F_{ab}^{(-\delta)}F_{ab}^{(0)}\rangle\big], \tag{A3}$$

where we took into account that $(F_{ab,ba}^{(\pm\delta)})^\dagger = F_{ba,ab}^{(\mp\delta)}$, and introduced the following functions:

$$K_{11}^{11} = |P_{11}|^2, \quad K_{22}^{11} = |P_{12}|^2, \quad K_{12}^{11} = P_{11}^*P_{12}, \tag{A4}$$

$$K_{11}^{22} = |P_{21}|^2, \quad K_{22}^{22} = |P_{22}|^2, \quad K_{12}^{22} = P_{21}P_{22}^*, \tag{A5}$$

$$K_{11}^{21} = P_{21}^*P_{11}, \quad K_{22}^{21} = P_{22}^*P_{12}, \quad K_{12}^{21} = P_{21}^*P_{12}, \quad K_{21}^{21} = P_{22}^*P_{11}. \tag{A6}$$

The expression for the atomic operator σ can be split into the sum of its average value and the deviation from the average [see Eq. (7)]. By taking into account that $\langle F\rangle = 0$, we are able to express the noise-correlation terms in the following way: $\langle F_i(\tau)F_j\rangle = \langle\sigma_i(\tau)\sigma_j\rangle - \langle\sigma_i(\tau)\rangle\langle\sigma_j\rangle$. According to the quantum regression theorem, the function $\langle\sigma_i(\tau)\sigma_j\rangle$ can be determined from the same equation of motion as the function $\langle\sigma_i(\tau)\rangle$ [28,29]. By using the Laplace transform, we can write correlation functions in terms of the steady-state solution $\langle\sigma_j\rangle$ of Eq. (1) and the U matrix [26,27,30–32], where

$$\langle\sigma^{(0)}\rangle = \left(\langle\sigma_{ab}^{(0)}\rangle,\langle\sigma_{ba}^{(0)}\rangle,\langle\sigma_{aa}^{(0)}\rangle,\langle\sigma_{bb}^{(0)}\rangle\right)^T, \tag{A7}$$

$$B = (0,0,\gamma_{ca},0)^T, \tag{A8}$$

$$M = \begin{pmatrix} -(\Gamma_{ba}+i\Omega) & 0 & iV & -iV \\ 0 & -(\Gamma_{ba}-i\Omega) & -iV & iV \\ iV & -iV & -\gamma_{ca} & \gamma_{ba}-\gamma_{ca} \\ -iV & iV & 0 & -\gamma_b \end{pmatrix}, \tag{A9}$$

$U^{(\pm\delta)} = \frac{1}{\mp i\delta I - M}$, $\langle\sigma^{(0)}\rangle = -M^{-1}B$, and I is the identity matrix.

The correlation functions, after some algebra, take the following form:

$$\langle F_{ab}^{(\pm\delta)}F_{ab}^{(0)}\rangle = \mathrm{Re}\sum_{j=1}^{4} U_{1j}^{(\pm\delta)}\Phi_{1j}, \tag{A10}$$

$$\langle F_{ab}^{(\pm\delta)}F_{ba}^{(0)}\rangle = \mathrm{Re}\sum_{j=1}^{4} U_{1j}^{(\pm\delta)}\Phi_{2j}, \tag{A11}$$

$$\langle F_{ba}^{(\pm\delta)}F_{ab}^{(0)}\rangle = \mathrm{Re}\sum_{j=1}^{4} U_{2j}^{(\pm\delta)}\Phi_{1j}, \tag{A12}$$

$$\langle F_{ba}^{(\pm\delta)}F_{ba}^{(0)}\rangle = \mathrm{Re}\sum_{j=1}^{4} U_{2j}^{(\pm\delta)}\Phi_{2j}, \tag{A13}$$

where

$$\Phi_{1j} = \begin{pmatrix} -\langle\sigma_{ab}^{(0)}\rangle^2 \\ \langle\sigma_{bb}^{(0)}\rangle - |\langle\sigma_{ab}^{(0)}\rangle|^2 \\ \langle\sigma_{ab}^{(0)}\rangle(1-\langle\sigma_{aa}^{(0)}\rangle) \\ -\langle\sigma_{ab}^{(0)}\rangle\langle\sigma_{bb}^{(0)}\rangle \end{pmatrix}, \tag{A14}$$

$$\Phi_{2j} = \begin{pmatrix} \langle\sigma_{aa}^{(0)}\rangle - |\langle\sigma_{ba}^{(0)}\rangle|^2 \\ -\langle\sigma_{ba}^{(0)}\rangle^2 \\ -\langle\sigma_{ba}^{(0)}\rangle\langle\sigma_{aa}^{(0)}\rangle \\ \langle\sigma_{ba}^{(0)}\rangle(1-\langle\sigma_{bb}^{(0)}\rangle) \end{pmatrix}. \tag{A15}$$

Under the conditions of Eq. (3), functions $\langle\sigma_{ij}^{(0)}\rangle$ and $U_{1j,2j}$ can be approximated as follows: $\langle\sigma_{ab,ba}^{(0)}\rangle \approx \pm i\frac{V}{(\Gamma_{ba}\pm i\Omega)(1+\kappa)}$, $\langle\sigma_{aa}^{(0)}\rangle \approx \frac{1}{1+\kappa}(1+\kappa\gamma_{ca}/\gamma_b)$, $\langle\sigma_{bb}^{(0)}\rangle \approx \frac{\kappa}{1+\kappa}\gamma_{ca}/\gamma_b$,

$$U_{1j}^{(\pm\delta)} = \frac{1}{D^{(\pm\delta)}}\begin{pmatrix} \Gamma_2^{(\pm\delta)}\Gamma_{ca}^{(\pm\delta)}\Gamma_b^{(\pm\delta)} + V^2\Gamma_T^{(\pm\delta)} \\ V^2\Gamma_T^{(\pm\delta)} \\ iV\Gamma_2^{(\pm\delta)}\Gamma_b^{(\pm\delta)} \\ -iV\Gamma_2^{(\pm\delta)}\left(\Gamma_{ca}^{(\pm\delta)}+\gamma_{ca}-\gamma_{ba}\right) \end{pmatrix}, \tag{A16}$$

$$U_{2j}^{(\pm\delta)} = \frac{1}{D^{(\pm\delta)}}\begin{pmatrix} V^2\Gamma_T^{(\pm\delta)} \\ \Gamma_1^{(\pm\delta)}\Gamma_{ca}^{(\pm\delta)}\Gamma_b^{(\pm\delta)} + V^2\Gamma_T^{(\pm\delta)} \\ -iV\Gamma_1^{(\pm\delta)}\Gamma_b^{(\pm\delta)} \\ iV\Gamma_1^{(\pm\delta)}\left(\Gamma_{ca}^{(\pm\delta)}+\gamma_{ca}-\gamma_{ba}\right) \end{pmatrix}, \tag{A17}$$

where $\Gamma_{ca}^{(\pm\delta)} = \gamma_{ca} \mp i\delta$, $\Gamma_b^{(\pm\delta)} = \gamma_b \mp i\delta$, $\Gamma_T^{(\pm\delta)} = \Gamma_b + \Gamma_{ca} + \gamma_{ca} - \gamma_{ba}$, and $D^{(\pm\delta)} = \Gamma_1^{(\pm\delta)}\Gamma_2^{(\pm\delta)}[\Gamma_{ca}^{(\pm\delta)}\Gamma_b^{(\pm\delta)} + V_0^2\Gamma_T^{(\pm\delta)}(\frac{1}{\Gamma_1^{(\pm\delta)}} + \frac{1}{\Gamma_2^{(\pm\delta)}})]$.

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