Generation of time-bin entangled photon pairs using a quantum-dot cavity system

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We present a scheme to realize a deterministic solid state source of time-bin entangled photon pairs using cavity-assisted stimulated Raman adiabatic passage (STIRAP) in a single quantum dot. The quantum dot is embedded inside a semiconductor cavity, and the interaction of a coherent superposition of two temporally separated input pulses and the cavity mode leads to a two-photon Raman transition, which produces a time-bin entangled photon pair through the biexciton-exciton cascade. We show that the entanglement of the generated state can be measured using triple coincidence detection, and the degree of entanglement is quantified as the visibility of the interference. We also discuss the effect of pure dephasing on entanglement of the generated photon pair. Pronounced interference visibility values of greater than $1/\sqrt{2}$ are demonstrated in triple coincidence measurement using experimentally achievable parameters, thus demonstrating that the generated photons are suitable for applications with Bell’s inequality violation and quantum cryptography.

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I. INTRODUCTION

A source of entangled photon pairs is an essential building block for various quantum information processing protocols, such as quantum cryptography and quantum teleportation. Generally, the employed entangled state of photons in these experiments are entangled in both the energy and the polarization degrees of freedom. However, because of unavoidable polarization dispersion in optical fibers, the polarization entangled photons are not suitable for distribution over large distances. In related experiments, entangled states of photons in energy and time degrees of freedom, using discrete time intervals (time-bin) for photon emission, have been demonstrated and the entanglement between these photons has been successfully distributed over distance of 50 km. In these experiments, the time-bin entangled photons were generated through the parametric down converter (PDC) using the pump as a superposition of two time separated pulses. A PDC is a heralded source of entangled photons where the number of generated photon pairs are probabilistic. At low pump intensity, when the probability of generating more than one photon pair remains small, the efficiency of the source remains very low (less than 20%). For quantum information processing applications, one requires a scalable source which generates precisely a single photon pair on demand. In the last few years, there has been considerable progress for developing on demand single photon and entangled photon sources using single quantum dots (QDs)\cite{13,14}, where the QDs provide the potential advantages of integrability and scalability in such experiments. In semiconductor QDs, polarization entangled photons have been successfully generated in the biexciton-exciton cascade decay\cite{15}.

In 2005, Simon and Poizat\cite{15} proposed an on-demand generation of time-bin entangled photons through the biexciton-exciton cascade in idealized QDs, where the biexciton state is created by pumping through two pulses interacting at two distinct times. The state of the time-bin entangled photon pair is given by

$$|\psi\rangle = \sqrt{p_1}|\text{early}\rangle_1|\text{early}\rangle_2 + e^{i\phi}\sqrt{p_2(1-p_1)}|\text{late}\rangle_1|\text{late}\rangle_2,$$

where \text{early} and \text{late} are two time bins and $p_1$ is the probability of generating a photon pair in the early time bin (from the first pulse) and $p_2$ is probability of generating a photon pair in the late time bin (from the second pulse); the total probability is then $p_1 + p_2 = 1$. For generating maximally entangled state $|\psi\rangle$, one requires $p_1 = p_2 = 1/2$. Therefore, a precisely regulated population transfer between the QD energy levels is essential. Moreover, pure dephasing processes present in semiconductor produce detrimental effects on the entanglement of the generated state. In quantum information protocols, such as entanglement swapping, it is essential that the photons should not have any other correlation except the time-bin entanglement. However, in the biexciton-exciton cascade, emitted photons also have time correlations. These undesirable temporal correlations can be minimized by manipulating emission rates of photons using resonant cavities\cite{15}.

In this work, we propose to generate an efficient time-bin entangled photon pair using stimulated Raman adiabatic passage (STIRAP). The coherent excitation in the system of QDs embedded in a semiconductor cavity have been an active area of research\cite{17,18}. We consider the initial QD state is in a metastable state, and there have been several methods for achieving this using electrical control of QD-cavity mode resonance\cite{19,20}. We demonstrate that the STIRAP process then provides an efficient regulated way for population transfer. We also investigate how the cavity enhanced decay rates suppress the detrimental effects of pure dephasing.

Our paper is organized as follows. In Sec. II, we present a formal theory of generation of time-bin entangled photon pair from a single QD coupled to a semiconductor cavity. In Sec. III, we investigate the measure of photon entanglement by a triple coincidence detection and also study the effects of dephasing. In section IV, we present our conclusions.
II. GENERATING TIME-BIN ENTANGLLED PHOTOON PAIRS USING STIRAP

We consider a QD embedded in a semiconductor microcavity, where the energy level diagram of the system is shown in Fig. 1. The dipole transitions from the biexciton state \(|u\rangle\) to the exciton state \(|y\rangle\), and from the exciton state \(|y\rangle\) to the ground state \(|g\rangle\), are coupled through a \(y\)-polarized single mode of the semiconductor cavity, with coupling constants \(g_1\) and \(g_2\), respectively. Because of the large biexciton binding energy of semiconductor QDs, it is not possible to couple the biexciton and exciton transitions from the same cavity mode, and thus manipulation of the biexciton binding energy becomes essential in these systems.\(^21,22\) Usually the binding energy of the charge-neutral biexciton has a negative value, however by changing the confinement size or by changing the strain, it has been found that the biexciton binding energy can be tuned to zero or a positive value. Very recently, manipulation of the binding energy of the biexciton has also been reported by applying lateral electric fields.\(^22\) Moreover, construction of an electrode for applying a lateral electric field in the vicinity of a QD within a photonic crystal cavity has also been reported.\(^22\) Therefore, it is now possible to manipulate the binding energy of biexcitons inside semiconductor cavities. The other advantage of lateral electric fields is that they can be used to create a voltage-tunable metastable exciton state \(|m\rangle\).

In III-V QDs, bright neutral excitons are formed when an electron from the \(s\)-shell or the \(p\)-shell of the valence band is excited to the \(s\)-shell or \(p\)-shell of the conduction band, respectively; the transitions from \(s\)-shell to \(p\)-shell and vice versa are essentially symmetrically forbidden. However, in the presence of an applied lateral voltage, the charge carrier symmetry can be suitably broken so that a bright exciton is formed, e.g., from the \(s\)-shell conduction band to \(p\)-shell conduction band.\(^22\) Consequently, by applying a lateral voltage larger than the values required to break the symmetry, and then using a \(\pi\) pulse excitation to this state, a symmetrically forbidden exciton can be created which behaves as the metastable state \(|m\rangle\), after lowering the applied voltage again. In what follows below, we will assume that this initial state can be created and focus on its evolution after applying STIRAP pulses.

Initially, the QD is prepared in the metastable state \(|m\rangle\) and the cavity mode in the vacuum state. An \(x\)-polarized pump field with a Rabi frequency \(\Omega_p(t)\) is applied between the metastable state \(|m\rangle\) and the biexciton state \(|u\rangle\). The Hamiltonian of the system, in the rotating frame at the pump frequency (interaction picture), can be written as

\[
H = \hbar \Delta_p |m\rangle \langle m| + \hbar \Delta_1 |y\rangle \langle y| + \hbar (\Delta_1 + \Delta_2) |g\rangle \langle g| \\
+ \hbar \Omega_p(t) |u\rangle \langle m| + g_1 |u\rangle \langle y| |a| + g_2 |y\rangle \langle g| |a + H.c.|, \tag{1}
\]

where \(\Delta_p\), and \(\Delta_1\) (\(\Delta_2\)) are the detunings of the pump field, and the cavity mode with the biexciton (exciton) transition frequencies, respectively; \(H.c\) refers to Hermitian conjugate. For simulating the dynamics of the system, we perform quantum master equation calculations in the density matrix representation. The evolution of the QD-cavity system is given by

\[
\dot{\rho} = -\frac{i}{\hbar} [H, \rho] - \frac{1}{2} \sum_{\mu} [L_H^\dagger L_H \rho - 2 L_H \rho L_H^\dagger + \rho L_H^\dagger L_H], \tag{2}
\]

where \(L_\mu\) are the Lindblad operators, with terms \(\sqrt{\gamma_1} |m\rangle \langle u|\), \(\sqrt{\gamma_1} |y\rangle \langle u|\), and \(\sqrt{2\gamma_2} |g\rangle \langle y|\) corresponding to the spontaneous decays, and \(\sqrt{2\gamma_1} |u\rangle \langle u|\), \(\sqrt{\gamma_1} |m\rangle \langle m|\), and \(\sqrt{\gamma_2} |y\rangle \langle y|\), corresponding to pure dephasing of the biexciton and exciton states. The emission of the photons from the cavity mode is given by the Lindblad operator \(\sqrt{\kappa}|a\rangle \langle a|\), where \(2\kappa\) is the decay rate of the leaky cavity. We safely neglect the spontaneous decay of the metastable state \(|m\rangle\) during the evolution, as the lifetime of the metastable state is, by definition, very large.

We numerically solve the optical Bloch equations using Eq. \((2)\), for density matrix elements \(\rho_{ij} = \langle i | \rho | j \rangle\). To simplify the notation, we use the definitions \(|Y\rangle = |y, 1\rangle\), \(|G\rangle = |g, 1\rangle\), and \(|G'\rangle = |g, 2\rangle\), where the alpha-numeric notation corresponds to the energy state of QD and the number corresponds to the cavity photon. The complete dynamics of the system is expressed by the following
two-photon Raman resonance condition can be satisfied
Stark shifts in energy states remains constant and the
also remember that the Stark shifts of energy levels also
the evolution of the population in the QD energy lev-
of the pump field is given by \( \Omega \)

\[
\rho_{mm} = -i\Omega_p(t)\rho_{am} + i\Omega_p(t)\rho_{mv} + \gamma_1\rho_{uu}, \quad (3)
\]

\[
\rho_{uu} = i\Omega_p(t)\rho_{um} - i\Omega_p(t)\rho_{mu} + ig_1\rho_{ay} - ig_1\rho_{yu} \quad (4)
\]

\[
-2\gamma_1\rho_{uu},
\]

\[
\rho_{yy} = -ig_1\rho_{yy} + ig_1\rho_{yu} - ig_2\sqrt{2}\rho_{yG} + ig_2\sqrt{2}\rho_{yG'} - (\kappa + \gamma_2)\rho_{PY}, \quad (5)
\]

\[
\rho_{GG'} = ig_2\sqrt{2}\rho_{G'Y} - ig_2\sqrt{2}\rho_{G'Y'} - 2\kappa\rho_{GG'}, \quad (6)
\]

\[
\rho_{yy} = -ig_2(\rho_{G'G} - \rho_{GG}) + \kappa\rho_{YY} + \gamma_1\rho_{uu} - \gamma_2\rho_{yy}, \quad (8)
\]

\[
\rho_{gg} = \kappa\rho_{GG} + \gamma_3\rho_{yy}, \quad (9)
\]

\[
\rho_{um} = -i\Delta_p\rho_{um} - i\Omega_p\rho_{mm} - ig_1\rho_{ym} + i\Omega_p\rho_{uu} \quad (10)
\]

\[
-\gamma_1 + \frac{3\gamma_d}{2}, \quad (11)
\]

\[
\rho_{uu} = -i\Delta_1\rho_{uu} - i\Omega_p\rho_{uu} - i\Omega_p\rho_{yy} + ig_1\rho_{uu} \quad + ig_2\sqrt{2}\rho_{uu} - (\gamma_1 + \kappa + 2\gamma_d, \quad (12)
\]

\[
-2\rho_{uu},
\]

\[
\rho_{YY} = -i\Delta_2\rho_{YY} - i\Omega_p\rho_{YY} - ig_1\rho_{YY} + i\Omega_p\rho_{uu} \quad + ig_2\sqrt{2}\rho_{YY} - (\gamma_1 + \kappa + 2\gamma_d)\rho_{YY,} \quad (13)
\]

\[
\rho_{mY} = -i(\Delta_1 - \Delta_2)\rho_{mY} - i\Omega_p(t)\rho_{mY} - ig_1\rho_{mY} \quad + ig_2\sqrt{2}\rho_{mY} - (\gamma_d + \kappa + 2\gamma_d)\rho_{mY}, \quad (14)
\]

\[
\rho_{mG} = -i(\Delta_1 - \Delta_2)\rho_{mG} - i\Omega_p(t)\rho_{mG} - ig_1\rho_{mG} \quad + ig_2\sqrt{2}\rho_{mG} - (\gamma_d + \kappa + 2\gamma_d)\rho_{mG}, \quad (15)
\]

\[
\rho_{GG} = -i\Delta_2\rho_{GG} - i\Omega_p\rho_{GG} - ig_1\rho_{GG} \quad + ig_2\sqrt{2}\rho_{GG} - (\gamma_d + \kappa + 2\gamma_d)\rho_{GG}, \quad (16)
\]

\[
\rho_{Gy} = -i\Delta_2\rho_{Gy} - i\Omega_p\rho_{Gy} + ig_2\rho_{yy} + \frac{\kappa + \gamma_d}{2}\rho_{Gy}, \quad (17)
\]

When the pump field and the cavity mode satisfy the
two-photon Raman resonance condition \( \Delta_p \approx \Delta_1 + \Delta_2 \),
the evolution of the population in the QD energy levels
follows the cavity-assisted STIRAP. Here, one must
also remember that the Stark shifts of energy levels also
play an important role in two-photon Raman resonance
condition. However, for constant cavity couplings,
the Stark shifts in energy states remains constant and
the two-photon Raman resonance condition can be satisfied
easily by changing the detuning of the pump field \( \Delta_p \) only.

In Fig. 2 we show the numerical simulations after solving
Eqs. (13)-(17). The pump field is chosen to be a coherent
superposition of two time-separated Gaussian pulses of the same width, but different amplitudes, which can be
generated by passing a Gaussian pulse through an unbalanced
two arm interferometer. The Rabi frequency of the pump field is given by \( \Omega_p(t) = \Omega_1(t) + \Omega_2(t - T) \),
where \( \Omega_1(t) \) is the Rabi frequency of each pulse and \( T \)
is the time gap between the pulses. We select a typical
value of \( \Omega_1(t) \) such that the population of the state \( |m\rangle \)
is pumped to the state \( |y, 1\rangle \) in STIRAP with probability

\[ p_1 = 1/2. \]

Due to the nature of the leaky cavity mode, the photon is emitted from the final state \( |y, 1\rangle \) and the
system is evolved into the state \( |y, 0\rangle \) state. The population
in state \( |y, 0\rangle \) is transferred to the state \( |g, 1\rangle \) through the
cavity mode. After emitting another photon from the state \( |g, 1\rangle \), the system finally reaches the ground state
\( |g, 0\rangle \). Thus a photon pair in the early time bin is emitted
during the interaction of the first pulse \( \Omega_1(t) \). The
remaining population in state \( |m\rangle \) is similarly pumped
by another pulse \( \Omega_2(t - T) \) and a photon pair is generated
with probability \( p_2 = 1 - p_1 \) in the late time bin. The
state of the generated photon pair is thus maximally
time-bin entangled state. For a QD embedded in a micro-
cavity, the off-resonant exciton has a spontaneous decay
rate of the order of 0.1 – 1 meV \( (10^{-3} \text{ meV}) \) for \( g = 0.1 \text{ meV} \),
and the cavity decay condition \( \kappa >> g \) can be achieved
easily. We stress that all of these parameters correspond
closely to those in present day experiments.

During the time interaction with the pump pulses, the
population in the upper state \( |u\rangle \) remains always less that
0.1 and the population in \( |g, 2\rangle \) remains negligible. The state of emitted photon pair from the cavity mode can therefore be written as

\[ \rho(t) = \left[ a_1(t)a_2^\dagger(t) + a_1^\dagger(t - T)a_2(t - T) \right] |0\rangle \rangle, \]

where \( |0\rangle \rangle \) is vacuum field, \( \langle a_1(t) \rangle = (\sigma_{yy}(t), \) and
\( \langle a_2(t) \rangle = (\sigma_{GG}(t), \) for \( \sigma_{ij} = |i\rangle \rangle \langle j| \). In quantum information
protocols, such as entanglement swapping, it is essential that the photons in the mode \( a_1 \) and \( a_2 \) should not have any other correlation except the time-bin entanglement. However, in the biexciton-exciton cascade, the \( a_1 \) mode photon is always generated after the emission
of the \( a_2 \) mode photon. Thus the \( a_1 \) and \( a_2 \) modes

\[ P_{22}(t) = P_{22}(0) \exp(-t/\tau_{22}), \]

where \( \tau_{22} \) is the coherence time of the cavity field and
\( P_{22}(t) \) is the population in state \( |g, 2\rangle \).
remain time correlated. This undesirable temporal correlation becomes negligible for $\Gamma_1/\Gamma_2 >> 1.2$, where $\Gamma_i$ is the emission rate of the photon in $a_i$ mode. In our scheme above, the first photon in mode $a_1$ is generated in resonant Raman process, which is emitted with the cavity mode decay rate $\kappa$, and the second photon is generated through cavity enhanced spontaneous emission. The condition $\Gamma_1/\Gamma_2 >> 1$ can therefore be easily satisfied by choosing a suitably large value of the detuning $\Delta_2$ such that $g_2^2 \kappa/(\kappa^2 + \Delta_2^2) << \kappa$.

III. TRIPLE COINCIDENT DETECTION OF THE PHOTON ENTANGLEMENT, AND THE INFLUENCE OF PURE DEPHASING

Next, we discuss how to measure the entanglement of the generated state of the photons. The Concurrence of the state (18) is directly related to the coherence of the state (18). For measuring the degree of entanglement, photons from each mode are passed through an unbalanced two path interferometer; the time difference between two arms is $T$, with phase difference $\phi$, and $T$ is similar to the time difference between the two pulses in the pump fields. After passing through the interferometers, the field operators at the output of the interferometers can be expressed as

$$a_3(t) = a_1(t) + e^{i\phi}a_1(t-T), \quad (19)$$
$$a_4(t) = a_2(t) + e^{i\phi}a_2(t-T). \quad (20)$$

The post-selection, for detecting both photon simultaneously after passing through the interferometers, projects the state (18) into the state

$$|\psi_c(t)\rangle = \left[ a_1^\dagger(t)a_2^\dagger(t) + (1 + e^{2i\phi})a_1(t-T)a_2^\dagger(t-T) 
\quad + a_1^\dagger(t-2T)a_2^\dagger(t-2T) \right] |\{0\}\rangle. \quad (21)$$

Clearly, the state (21) has three terms which are distinguishable in time. The middle term, appearing at $t = T$, provides the information about the entanglement of state (18). For separating different terms in state (21), the time of detection of photons is measured with reference to the pump photons using a triple coincidence detection. The probability of triple coincidence detection of one photon at the output of each interferometer, and one from the input pulse $\Omega_1$, is given by

$$G^{(3)}(\tau) = \int_0^\infty dt' \int_{-T_{bin}}^{T_{bin}} dr' \langle \Omega_1(t') \rangle^2 \times \langle a_3^\dagger(t' + \tau)a_4^\dagger(t' + \tau + \tau')a_3(t' + \tau + \tau') \rangle \quad (22)$$

where $T_{bin}$ is the width of the time-bins; which is chosen larger than the biexciton-exciton cascade decay and smaller than $T$. We can simplify the above expression for $G^{(3)}(\tau)$ using the property of field operators, $a_1(t)a_2(t-T)|\{0\}\rangle = 0$, as both photons are generated almost together in the cascade decay. Subsequently, we can simplify the correlation function in (22) as

$$\langle a_3^\dagger(t' + \tau)a_4^\dagger(t' + \tau + \tau')a_3(t' + \tau + \tau') \rangle = \langle a_1^\dagger(t' + \tau)a_2^\dagger(t' + \tau + \tau')a_2(t' + \tau + \tau')a_1(t' + \tau + \tau') \rangle$$
$$\quad + \langle a_1^\dagger(t' - T + \tau)a_2^\dagger(t' - T + \tau + \tau')a_2(t' - T + \tau + \tau')a_1(t' - T + \tau + \tau') \rangle$$
$$\quad + 2 \cos 2\phi \langle a_1^\dagger(t' + \tau)a_2^\dagger(t' + \tau + \tau')a_2(t' - T + \tau + \tau')a_1(t' - T + \tau + \tau') \rangle. \quad (23)$$

which is evaluated for state (18) by applying quantum regression formula.\textsuperscript{25} We relegate the details of the $G^{(3)}(\tau)$ calculation to the Appendix.

In Fig. 3 we plot $G^{(3)}(\tau)$ for the same parameters used in Fig. 2 where the time-dependent populations were shown. The computed value of $G^{(3)}(\tau)$ has three peaks centered at $\tau = 0$, $T$, and $2T$. The first peak at $\tau = 0$ correspond to the photons generated in the early time-bin that have passed through the short arms of the interferometers. Similarly the peak centered at $\tau = 2T$ corresponds to the photon pair generated in the late time-bin that have passed through the long arms of the interferometers. The central peak at $\tau = T$ corresponds to the overlap of the photons generated in early time bin and passed through the longer arms in the interferometers and the photons generated in the late time-bin and passed through the short arms. Thus only the central peak contains the information about the entanglement and can be easily selected by choosing a narrow time window around $\tau = T$. We have also found that the required value of $T$ is slightly less than the actual time between the pump pulses, which shows that in STIRAP, the photons are actually generated before the pump pulse reaches its maximum. For the parameters used in Fig. 2, the time between pump pulses is $15\pi/g$, but the central peak in Fig. 3 is a maximum for $T = 14\pi/g$.

The coherence in the generated state (18) can be measured by varying the phase $\phi$ between the overlapping amplitudes corresponding to the early and the late time bins along the central peak. In Fig 4, we plot the interference pattern produced in the measurement of $P_c = \int_{T-2T_{bin}}^{T+2T_{bin}} G^{(3)}(\tau) d\tau$. The visibility of the interference pattern, defined as $V=\text{maximum of } P_c - \text{minimum of } P_c)/\text{maximum of } P_c + \text{minimum of } P_c$, gives the
FIG. 3: The triple coincidence correlation of detecting one photon at output of each interferometer and one from the input pulse $\Omega_1$ for $T = 14\pi/g$. The other parameters are the same as in Fig. 2

FIG. 4: The integrated value of the triple coincidence correlation $G^{(3)}(\tau)$ along the central peak at $\tau = T$. The interference pattern appears on changing the phase $\phi$ produced by the interferometers.

FIG. 5: The dependence of the visibility, i.e. entanglement of the generated time-bin entangled state of the photon on dephasing rate $\gamma_d$.

IV. CONCLUSIONS

We have presented a cavity-QED STIRAP scheme for generating a scalable source of time-bin entangled photon pairs, and we also investigated the role of pure dephasing on entanglement. The generated state of the photons can be detected by measuring the correlations between the pump and the generated photons. We found that for small values of pure dephasings, it is possible to achieve larger values of entanglement using current working technologies.

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Appendix: Calculation of multi-time correlations

Here we briefly discuss the method for calculating two-time correlation and four-time correlation used in Sec. III. We follow the approach discussed by Gardiner and Zoller25 for evaluating multi-time correlations. The required two time correlation can be expressed as

$$
\langle a_1^\dagger(t) a_2^\dagger(t + \tau) a_2(t + \tau) a_1(t) \rangle = Tr \{ a_2(t + \tau) a_1(t) \rho(0) a_1^\dagger(t) a_2^\dagger(t + \tau) \} \quad (A.1)
$$

$$
= Tr \{ a_2(t + \tau) \rho'(t) a_1^\dagger(t) a_2^\dagger(t + \tau) \}, \quad (A.2)
$$

$$
= Tr \{ a_2 \rho'(t + \tau) a_1^\dagger(t) a_2^\dagger(t + \tau) \} \quad (A.3)
$$

where $Tr$ stands for trace and operators $a_i$ appearing without time parenthesis are in Schrodinger picture.
\[ \rho'(t) = a_1(t)\rho(0)a_1^\dagger(t) = a_1\rho(t)a_1^\dagger \] is calculated after evolving the initial state \( \rho(0) = \rho_{mm}|m\rangle\langle m| \) for time \( t \) using Eqs. (3)-(17) and then operating by \( a_1 \) and \( a_1^\dagger \) from left and right, respectively. Clearly, \( \rho' \) also follows the same equations of motions (3)-(17). Now, using initial value \( \rho'(t) = a_1\rho(t)a_1^\dagger \) at time \( t \), and evolving for time \( \tau \), \( \rho'(t+\tau) \) is calculated. The value of the required correlation is calculated using Eq. (A.3). A similar straightforward approach, considering the times appearing in the \( a \) operators in ascending order, is applied in evaluating the four time correlations as follows:

\[
\langle a_1^\dagger(t)a_2^\dagger(t+\tau)a_2(t-T)\rangle = Tr \left\{ a_2(t-T)\rho(0)a_1^\dagger(t)\right\} 
\]

(A.4)

\[
= Tr \left\{ a_2(t-T+\tau)\rho_1(t-T)a_1^\dagger(t)a_2^\dagger(t+\tau)\right\} , 
\]

(A.5)

\[
= Tr \left\{ \rho_2(t-T+\tau)a_1^\dagger(t)a_2^\dagger(t+\tau)\right\} , 
\]

(A.6)

\[
= Tr \left\{ \rho_3(t)a_2^\dagger(t+\tau)\right\} , 
\]

(A.7)

\[
= Tr \left\{ \rho_3(t+\tau)a_1^\dagger\right\} , 
\]

(A.8)

where \( \rho(0) = \rho_{mm}|m\rangle\langle m| \), \( \rho_1(t-T) = a_1(t-T)\rho(0) = a_1\rho(t-T) \), \( \rho_2(t-T+\tau) = a_2(t-T+\tau)\rho_1(t-T) = a_2\rho_1(t-T+\tau) \), and \( \rho_3(t) = \rho_2(t-T+\tau)a_1^\dagger(t) \equiv \rho_2(t)a_1^\dagger \).

Thus the density matrices \( \rho, \rho_1, \rho_2, \) and \( \rho_3 \) are evolved for times, \( 0 \) to \( t-T \), \( t-T \) to \( t-T+\tau \), \( t-T+\tau \) to \( t \), and \( t \) to \( t+\tau \) respectively. The evolution of \( \rho(0) \) is given by Eqs. (3)-(17), while the evolutions of density matrices \( \rho_i \) for \( i=1,2,3 \), follow the similar equations, written for \( \rho \), as

\[
\dot{\rho}_{Gy} = -ig_1\rho_{uy} - ig_2\sqrt{2}\rho_{Gy} + ig_2\rho_{YG}, 
\]

\[
-(\gamma_2 + \kappa/2 + \gamma_d)\rho_{gy}, 
\]

(A.9)

\[
\dot{\rho}_{Gg} = -ig_2\rho_{yg} - \frac{1}{2}i\rho_{Gg}, 
\]

\[
(A.10)
\]

\[
\dot{\rho}_{uy} = -i\Delta_1\rho_{uy} - i\Omega_p(t)\rho_{my} - ig_1\rho_{uy} + ig_2\rho_{ug}, 
\]

\[
-(\gamma_1 + \gamma_2/2 + 3\gamma_d/2)\rho_{uy}, 
\]

(A.12)

\[
\dot{\rho}_{Gy} = i\Delta_2\rho_{Gy} - ig_2\sqrt{2}\rho_{Gy} + ig_2\rho_{Gg}, 
\]

\[
-(\kappa + \gamma_2/2 + \gamma_d/2)\rho_{Gy}, 
\]

(A.13)

\[
\dot{\rho}_{YG} = -i\Delta_2\rho_{YG} - ig_1\rho_{UG} - ig_2\sqrt{2}\rho_{YG} + ig_2\rho_{yG}, 
\]

\[
-(\kappa + \gamma_2/2 + \gamma_d/2)\rho_{YG}, 
\]

(A.14)

\[
\dot{\rho}_{yg} = -i\Delta_2\rho_{yg} - ig_2\rho_{Gg} - \frac{1}{2}(\gamma_2 + \gamma_d)\rho_{yg}, 
\]

(A.15)

\[
\dot{\rho}_{my} = -i(\Delta_1 - \Delta_p)\rho_{my} - i\Omega_p^\ast(t)\rho_{uy} + ig_2\rho_{mg}, 
\]

\[
-(\gamma_2/2 + \gamma_d)\rho_{my}, 
\]

(A.16)

\[
\dot{\rho}_{ug} = -i(\Delta_1 + \Delta_2)\rho_{ug} - i\Omega_p\rho_{mg} - ig_1\rho_{ug}, 
\]

\[
-(\kappa/2 + \gamma_1 + \gamma_d)\rho_{ug}, 
\]

(A.17)

\[
\dot{\rho}_{mg} = -i(\Delta_1 + \Delta_2 - \Delta_p)\rho_{mg} - i\Omega_p^\ast(t)\rho_{ug} + ig_2\rho_{my}, 
\]

\[
-\frac{1}{2}(\kappa + \gamma_d)\rho_{mg}, 
\]

(A.18)

\[
\dot{\rho}_{Gg} = -ig_2\sqrt{2}\rho_{Gg} - \kappa\rho_{Gg}, 
\]

(A.19)

\[
\dot{\rho}_{YG} = -i\Delta_2\rho_{YG} - ig_2\sqrt{2}\rho_{Gg} - ig_1\rho_{ug}, 
\]

\[
\frac{1}{2}(\kappa + \gamma_2 + \gamma_d)\rho_{YG}, 
\]

(A.20)

\[
\dot{\rho}_{ug} = -i(\Delta_1 + \Delta_2)\rho_{ug} - i\Omega_p\rho_{mg} - ig_1\rho_{Yg} 
\]

\[
-\gamma_2 + \gamma_d\right), 
\]

(A.21)

Finally, the value of four-times correlations used in Sec.III are found using Eq. (A.3).
