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ABSTRACT

We study entanglement created between two isolated qubits by interaction with entangled-photon pairs obtained by parametric down-conversion of a laser pump field. The induced entanglement is quantified using the mixed state Concurrence proposed by Wootters *et al.* [Phys. Rev. Lett. **78**, 5022 (1997)]. A universal value of qubit-entanglement, which is independent on the photon-pair wavefunction is identified to leading order in the qubit–field interaction and the pump field amplitude. The qubit entanglement decreases at higher laser pump intensities due to interference between the entangled photon pairs, which creates excitations in the qubit system. Maximal Concurrence is produced by only generating coherences between the ground and the highest excited qubit states.

I. INTRODUCTION

Entangled-photon pairs, produced by an optically nonlinear parametric down-conversion (PDC) crystal, are widely used in quantum optics[1–5] and provide a valuable tool in quantum-light optical spectroscopy.[6–10] The entanglement of quantum states of light is often described in terms of the wavefunction of entangled photon pairs.[11,12] However, photon-entanglement that can be used as a resource in quantum computation and quantum thermodynamics is yet to be developed.[13] We propose to quantify the entanglement of photon quantum light by its capacity to create entanglement in a two-qubit material system. Entanglement transfer from two-mode squeezed light to matter has been studied earlier[14–16] using a quantum master equation formulation. It has been shown that the optimal entanglement can be obtained by manipulating the matter with laser light. Reference 17 explores the transferred entanglement to qubits from various types of quantum light states. Different protocols that utilize a single-photon field to create entanglement between qubits have been proposed. These include the detection of single-photon emitted from qubits[18] and qubits–light interaction in a single-photon cavity.[19]

Here, we study two qubits interacting with entangled-photon pairs created by a classical laser source by means of an optically pumped nonlinear (PDC) crystal. These photon pairs are frequency-entangled and the amount of entanglement depends on the full two-photon wavefunction which, in turn, is related to the non-linear PDC crystal and the pump field properties, and has found numerous applications in quantum spectroscopy.[8,9] This is important as the same degree of entanglement may be achieved in different wavefunctions. This wavefunction-dependent information is not present in the phenomenological models used in the previous studies.[14–16] We employ a realistic model of squeezed light and show that for a weak classical pump, where the density of entangled photon pairs is low and their mutual interaction in the PDC crystal is negligible, the generated entanglement in the qubit system does not depend on the details of the entangled-photon pair wavefunction. The induced entanglement in the qubit system per photon pair by a weak field can thus be viewed as a standard unit of entanglement carried by the entangled-photon pair. As the pump intensity is increased, the induced entanglement is reduced and starts to depend on the details of the photon pair wavefunction. This can be attributed to the negative contributions to the generated entanglement by two photons that belong to different entangled-pairs. The decreased entanglement is accompanied by the generation of excited state population in the qubit system which, in turn, produces a classical-like "longitudinal" correlation between the two qubits.

A. Two-qubit *X*-state and entanglement

The entanglement of pure or mixed quantum states of a two-qubit system is well understood.[13,20,21] A reasonable measure of entanglement should be local unitary (LU) invariant, i.e., any two states that differ by an action of a direct product of two unitary transformations, referred to as LU transformations, applied to the two qubits should have the same degree of entanglement. The most detailed multi-parameter measure of entanglement is obtained by specifying an equivalence class of the system states with respect to LU transformations, or, equivalently, an orbit of the group $U(2) \times U(2)$ of LU transformations acting in the space of states. Hereafter, we choose the Concurrence as a single-parameter measure of the entanglement strength. This choice is rationalized by the Hill–Wootters theorem[22,23] that establishes a direct relation between the Concurrence $C(\rho)$ of a mixed state ρ of two qubits and its entanglement of formation $E(\rho)$, the latter defined in terms of an apparently complex optimization problem, and represents the cost of creating a mixed state from a Bell counterpart, and is closely related to the purification cost.[20] A time-reversal symmetry interpretation, which immediately reveals the LU nature of the Concurrence function, is discussed in Appendix A.

The density matrix ρ of two qubits in a mixed-state is completely determined by the set of one- and two-point correlation functions, $\boldsymbol{s} = \mathrm{Tr}(\hat{\boldsymbol{s}}\rho)$, $\boldsymbol{s}' = \mathrm{Tr}(\hat{\boldsymbol{s}}'\rho)$, and $c_{j\alpha} = \mathrm{Tr}(\hat{s}_j \hat{s}'_\alpha \rho)$, where s and s' denote observables for each qubit in its ground and excited states, for example "spin" values, and has overall 15 parameters. In this paper, we focus on an 11-dimensional subclass of mixed states known as X-states[24] characterized by vanishing mixed correlations. Obviously, due to an implied parity argument in the absence of permanent dipoles, and because of vanishing odd-point correlation functions of the quantum field, a two-qubit system initially in a separable state excited by entangled light, remains within the X-state class at all times. The X-states have been studied in the quantum information field widely[25,26] and show up naturally in variety of entangled-photon spectroscopy setups.[9] They can therefore serve as an important link between quantum information and quantum spectroscopy applications.

A convenient representation for the density matrix can be obtained by choosing the basis sets in the Hilbert spaces of individual qubits to be the eigenstates of $\boldsymbol{s} \cdot \boldsymbol{\sigma}$ and $\boldsymbol{s}' \cdot \boldsymbol{\sigma}$ (denoted by $|0\rangle$ and $|1\rangle$), so that the density-matrix assumes the following form:

$$\rho = n_{00}|00\rangle\langle 00| + n_{11}|11\rangle\langle 11| + n_{10}|10\rangle\langle 10| + n_{01}|01\rangle\langle 01| + \kappa|11\rangle\langle 00| + \kappa^*|00\rangle\langle 11| + \zeta|10\rangle\langle 01| + \zeta^*|01\rangle\langle 10|, \tag{1}$$

where $|ij\rangle, i, j = 0, 1$, is the standard product state, with the parameters satisfying the conditions $\sum_{ij} n_{ij} = 1$, $n_{ij} \geq 0$, and $|\kappa| \leq \sqrt{n_{00} n_{11}}$, $|\zeta| \leq \sqrt{n_{01} n_{10}}$, and the Concurrence becomes (details are given in Section 1 of Appendix A)

$$C(\rho) = \max\left(2\left(|\kappa| - \sqrt{n_{01} n_{10}}\right), 2\left(|\zeta| - \sqrt{n_{00} n_{11}}\right), 0\right). \tag{2}$$

For given populations, the Concurrence can assume values in the range $0 \leq C \leq |\sqrt{n_{00} n_{11}} - \sqrt{n_{10} n_{01}}|$.

The density matrix of a two-qubit system driven by the entangled-photon quantum field can be computed from first principles. To develop a perturbative expansion that properly takes into account the quantum nature of light, we split the system into the two-qubit system of interest with the Hamiltonian H_s, and the (auxiliary) optical counterpart that consists of the quantum electromagnetic field, and a classical optical source, described by the time-dependent polarization $\boldsymbol{P}$ that represents the pump laser, the PDC crystal, and the linear optical devices, e.g., mirrors, lenses, splitters, etc., with the quantum electromagnetic field coupled to the classical source and optical setting. The total Hamiltonian is given by $H_T(t) = H_s + H_a(t) + H_{int}$ with $H_{int} = -\int d\boldsymbol{r} \boldsymbol{P} \cdot \boldsymbol{E}$, representing the interaction between the qubit system and the quantum electromagnetic field; the latter is a part of the auxiliary system.

II. MODEL AND RESULTS

To illustrate the entanglement of two externally driven qubits, we consider a specific quantum system consisting of two non-interacting two-level atoms. Initially, both atoms are in their ground states described by a product state $|00\rangle \equiv |0\rangle \otimes |0\rangle$, while $|10\rangle, |01\rangle$, and $|11\rangle$, respectively, represent the states when only the first atom, only the second atom, and both atoms are in their excited states. This system is irradiated by light. The radiation-matter dipole interaction Hamiltonian is, $H_{int} = (\hat{d} + \hat{d}').\hat{E}^\dagger + h.c.$, where $\hat{d}$ and $\hat{d}'$ denote dipole operators for the "left" and the "right" atoms with excitation frequencies Ω_0 and Ω_0', respectively, and $\hat{E}^\dagger(\hat{E})$ is the field operator.

The reduced density matrix of the atomic system is obtained by tracing out the field degrees-of-freedom, $\rho(t) = \mathrm{Tr}_F\{\tilde{U}(t,0)\tilde{\rho}_T(t)\}$, where $\rho_T(t)$ is the full (atom + field) density matrix and the time dependence is with respect to the non-interacting Hamiltonian, $H_s + H_a(t)$, and $\tilde{U}(t) = T\exp[-(i/\hbar)\int_0^t d\tau \tilde{H}_{int-}(\tau)]$ is the time-ordered Liouville superoperator.[29,30]

We start by a time-dependent perturbative expansion of the two-qubit reduced density matrix in powers of H_{int}. The resulting expression contains convolutions of the multi-point Liouville space correlation functions of the qubit polarization operators, having time evolution with H_s, and multi-point Liouville space correlation functions of the electric field operators, having time evolution with $H_a(t)$. The latter can be computed using the effective action approach, based on integrating out the matter variables, associated with the optical subsystem. This leads to an effective field Hamiltonian[27] $H_a \equiv H_{eff} = \mathscr{E}_p \chi^{(2)} \int d\omega \int d\omega' \Phi(\omega, \omega') \hat{E}_1^\dagger(\omega) \hat{E}_2^\dagger(\omega') + H.C.$, where $\hat{E}_1^\dagger$ and $\hat{E}_2^\dagger$ are the creation operators for the two-entangled field modes, $\mathscr{E}_p$ and $\chi^{(2)}$ denote the pump field amplitude and second-order nonlinear response of the PDC crystal, and $\Phi(\omega, \omega')$ is the corresponding field amplitude which contains information on the entanglement between the two modes as discussed in Appendix B. We consider the case where one photon (denoted by subscript "1") from the entangled-photon pair interacts with the left atom and the other photon (denoted by subscript "2") interacts with the right atom. The interaction Hamiltonian is then $H_{int} = \hat{d}^\dagger \cdot \hat{E}_1 + \hat{d}'^\dagger \cdot \hat{E}_2 + h.c.$

By computing multi-point entangled-photon field correlation functions and applying Wick's theorem, we obtain a perturbative diagrammatic expansion in the field-matter interactions (equivalently, in the qubit transition dipoles). A regular expansion in powers of the classical driving field $\mathscr{E}_p$ can be obtained by further expanding the two-point field correlation functions in powers of $\mathscr{E}_p$. We consider perturbative results to second-order in $\mathscr{E}_p$. All results presented below are thus perturbative in the pump-field and also in the interaction between the entangled-photons and qubits. Examples of the diagrams contributing to the time-evolution of the qubit state are given in the figure in Section 1 Appendix B, where thick solid lines represent the two-point entangled photon correlation functions, computed using linear response from optical components. The perturbative approach is applicable to most experimental situations: the PDC process is, in general, weak and the generated entangled-photon field does not show strong (non-linear) dependence on the pump amplitude. The universal entanglement transfer proposed below can be directly tested experimentally.

The only surviving coherence in this setup is between states $|00\rangle$ and $|11\rangle$, denoted as κ. The populations of the atomic states and coherence to leading order in the pump field amplitude and in the entangled-field-qubit interaction are given by

$$n_{00} = 1 - \frac{|\chi^{(2)}\mathscr{E}_p|^2}{2\hbar^4}\left(|d'|^2\mathscr{P}_1 + |d|^2\mathscr{P}_2\right), \tag{3}$$

$$n_{01} = \frac{|\chi^{(2)}\mathscr{E}_p|^2}{2\hbar^4}|d'|^2\mathscr{P}_1, \tag{4}$$

$$n_{10} = \frac{|\chi^{(2)}\mathscr{E}_p|^2}{2\hbar^4}|d|^2\mathscr{P}_2, \tag{5}$$

$$\kappa(t) = -\chi^{(2)}(\mathscr{E}_p)^* e^{-i(\Omega_0+\Omega_0')t}\frac{dd'}{2\hbar^3}, \tag{6}$$

where $\mathscr{P}_i = \sqrt{(\pi\sigma_p^2/1 + 2\gamma\sigma_p^2 T_i^2)}$, σ_p is the spectral width of the pump, T_1 and T_2 are the time-delays of the two entangled photon wavepackets with respect to the peak of the pump pulse at the output of the PDC crystal, and $(\mathscr{E}_p)^*$ denotes complex conjugate of the pulse amplitude. The population $n_{11} = 0$. In deriving Eqs. (3)–(6), we have approximated the pump pulse spectral envelop by a Gaussian of width σ_p and that the phase of the induced polarization in the PDC crystal when averaged over the crystal volume can be represented by a Gaussian. This allows us to expand the field propagators analytically in Hermite polynomials as discussed in Appendix B. However, such approximations are not needed to obtain the perturbative results, Eqs. (3)–(6). A microscopic derivation of the above equations without invoking these approximations will be presented elsewhere.

It should be emphasized that the coherence κ is proportional to $(\mathscr{E}_p)^*$ while all other elements of the density matrix are proportional to $|\mathscr{E}_p|^2$. In the following, we take T_2 as unit of time and define the dimensionless amplitude $\mathscr{E} = \chi^{(2)}\mathscr{E}_p/\hbar T_2$, all quantities, such as σ_p, T_1, $\mathscr{P}_i$, etc., are expressed in units of T_2. Thus, for an arbitrary weak pump when $|\mathscr{E}| \gg |\mathscr{E}|^2$, the interaction with the entangled photon pair only generates the coherence κ without affecting the ground state population of the two qubits. Equations (3)–(6) hold in the perturbative limit when $\lambda = \mathscr{E}\sqrt{(\mathscr{P}_1\mathscr{P}_2/\pi)} \ll 1$.

An explicit expression for the Concurrence can be obtained in this case and is given in Eqs. (2). It is interesting to observe that the reduced state of the two atoms depends on the phase of the pump field through the coherence κ. However, the Concurrence is independent on this phase as it only depends on the magnitude of the coherences.

For a small pump amplitude when the quadratic terms $\mathscr{E}^2$ can be ignored in comparison with the linear terms, the atomic system can be approximated to remain in the ground state, that is, $n_{00} \approx 1$, and the Concurrence, as determined by Eq. (2), is equal to twice the absolute value of the coherence κ. In fact, any (small) population created in the singly excited state tends to decrease the Concurrence. Thus, higher order ($\mathscr{E}^2$) terms lead to the creation of population which, in turn, reduces the Concurrence and hence the entanglement between the two atoms.

To leading order in $\mathscr{E}$, the Concurrence is given by

$$C = \frac{|\mathscr{E}|}{\hbar^2}|dd'|\tilde{C} \tag{7}$$

with $\tilde{C} = 1$. Note that the induced entanglement is independent on the entangled-photon pair wavefunction and is proportional to the pump-field amplitude. The factor $|dd'|$ determines how effectively the photon-entanglement is transferred to create entanglement between the two atoms. Since the photon pair entanglement is determined by the wavefunction Φ and is independent on the pump amplitude, Eq. (7) thus gives a measure of the transferrable entanglement, which is independent on the entanglement in the photon pair. It does not mean that the transferred entanglement is non-zero even if the two photons are not entangled, in which case the coherence $\kappa = 0$ and, from the definition of the Concurrence, the transferred entanglement also vanishes. Thus, any amount of entanglement in the field obtained from a PDC process gives rise to the same entanglement in the qubit system for weak field and qubit–field interaction.

To second-order in the pump amplitude, the induced entanglement in the qubit system is obtained in terms of reduced photon-entanglement,

$$\tilde{C} = 1 - \mathscr{E}\sqrt{\mathscr{P}_1\mathscr{P}_2}, \tag{8}$$

which now depends on the pump pulse shape and the entangled-photon pair wavefunction $\mathscr{P}_1$ and $\mathscr{P}_2$.

In Eqs. (7) and (8), the net entanglement created in the qubits is expressed as the product of two terms. One term depends on the qubit properties alone while the other term is dependent only on the entangled-field. Interestingly, to lowest (leading) order, the field term is unity, which is suppressed as the pump intensity is increased as shown in Eq. (8). This field-dependent term can be therefore interpreted as the useful entanglement ($\tilde{C}$) contained in the photon pair, which is available to be utilized using the qubits. Equation (7) thus provides a working definition of the maximum entanglement $\tilde{C} = 1$ contained in a photon pair, which is transferrable to a qubit-system under the weak field and qubit interaction limit. Although we have considered a qubit system, the result is valid for any material system as long as we remain in the perturbative regime under which Eqs. (3)–(6) hold.

Figure 1 shows the variation of the Concurrence ($\tilde{C}$) with the pump field amplitude for different values of the spectral-width. The dashed curves use a non-perturbative (in pump amplitude) density matrix obtained by using Eqs. (B2)–(B7) in Eqs. (B12)–(B14), and the solid lines are perturbative results, Eq. (8). The Concurrence decreases with increasing pump amplitude. This decrease depends on the spectral width, higher values of σ_p leads to faster initial decay, which is well captured by the perturbative expression in Eq. (8). As σ_p becomes large such that $2\gamma\sigma_p^2 T_i^2 \gg 1$, Eq. (8) gives $\tilde{C} \approx 1 - \mathscr{E}\sqrt{\pi/(2\gamma T_1)}$. On the other hand, for small σ_p such that $2\gamma\sigma_p^2 T_i^2 \ll 1$, $\tilde{C} \approx 1 - \mathscr{E}\sqrt{\pi}\sigma_p$. The variation in Concurrence shows a scaling behavior with respect to $\mathscr{E}\sigma_p$ for small σ_p, which remains valid even for higher values of the pump amplitudes as shown in Fig. 2.

To conclude, the entanglement in a two-level two-atom (or two-qubit) system induced by the interaction with an entangled-photon pair is discussed using a perturbative approach. Although the standard measure of photon pair entanglement obtained by the PDC process is

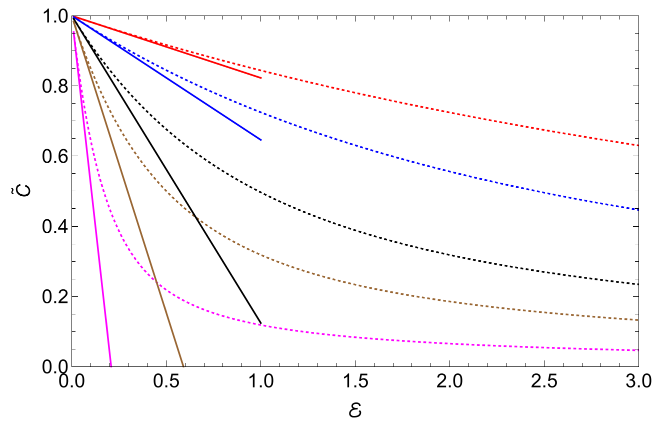

FIG. 1. Variation of the Concurrence with the pump pulse amplitude for spectral widths, $\sigma_p = 0.1$ (red), 0.2 (blue), 0.5 (black), 1.0 (brown), and 5.0 (magenta). Dashed curves are obtained from Eq. (2) using non-perturbative density-matrix elements from Eqs. (B12)–(B14), solid lines are the analytic result, Eq. (8), obtained using the perturbative results, Eqs. (3)–(6).

defined in terms of its wavefunction and is independent of the pump-field amplitude, we show that this does not provide a working definition of the photon-entanglement. Equations (7) and (8) clearly demonstrate that for a single entangled-photon pair, the entanglement transferred to the two-qubits is independent of the photon pair wavefunction. For higher pump intensity, when several photon pairs can interact with the qubit, the induced entanglement depends on the shape of the photon pair wavefunction and is decreased because the excess photons are used in generating excitations in the qubit system. This can be interpreted as reduction of the photon pair entanglement. We provide a working definition of the entanglement contained in a photon field, which can be measured in experiments that involve the interaction of photons with matter.

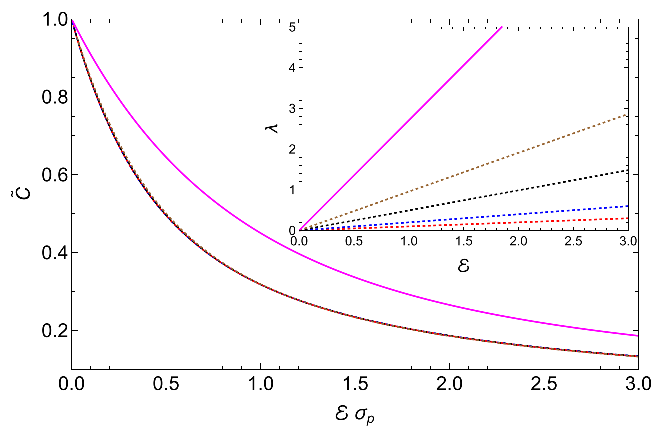

FIG. 2. Variations of the Concurrence with the scaled pump pulse amplitude $\mathcal{E}\sigma_p$ for the same set of spectral widths as in Fig. 1. All curves for smaller $\sigma_p < 5.0$ (magenta) exhibit the scaling predicted by perturbative Eq. (8), even beyond the range of $\mathcal{E}$ where $\lambda > 1$ and the perturbative expression is not valid, as shown in the Inset.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Upendra Harbola: Conceptualization (equal); Formal analysis (equal); Methodology (equal); Writing – original draft (equal); Writing – review & editing (equal). **Luca Candelori:** Formal analysis (equal); Methodology (equal); Writing – review & editing (equal). **John R. Klein:** Formal analysis (equal); Methodology (equal); Writing – review & editing (equal). **Vladimir Y. Chernyak:** Conceptualization (equal); Formal analysis (equal); Methodology (equal); Writing – review & editing (equal). **Shaul Mukamel:** Conceptualization (equal); Formal analysis (equal); Methodology (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its supplementary material.

APPENDIX A: CONCURRENCE OF A MIXED STATE OF TWO QUBITS

The entanglement of formation E has been introduced in Ref. 20 as a solution of an apparently complicated optimization problem; therefore, computation of E by applying the definition directly is not straightforward. To address this issue, Hill and Wootters introduced in Ref. 22 the quantity $C(\rho)$, referred to as the Concurrence of a mixed state ρ, i.e., a function

$$C : \mathscr{L} \to [0, 1], \tag{A1}$$

and showed that $E(\rho)$ is expressed in terms of $C(\rho)$ via the following Shannon entropy when ρ has at least two non-zero eigenvalues:

$$E = -\sum_{n=0,1} \frac{1 + (-1)^n\sqrt{1 - C^2}}{2} \log_2 \frac{1 + (-1)^n\sqrt{1 - C^2}}{2}. \tag{A2}$$

They further conjectured the validity of Eq. (A2) for an arbitrary mixed state. Later, Wooters proved the conjecture and established the validity of Eq. (A2) for an arbitrary mixed state.[23] The definition involves associating with a density matrix ρ a counterpart ρ^* using an operation of so-called "spin flip." One further introduces an operator $R(\rho)$

$$R(\rho) = \sqrt{\sqrt{\rho}\rho^*\sqrt{\rho}}, \tag{A3}$$

and defines Concurrence $C(\rho)$ in terms of $R(\rho)$

$$C(R) = \max(2\lambda_{\max}(R) - \mathrm{Tr}(R), 0) = \max(\bar{C}(R), 0) \tag{A4}$$

with $\lambda_{\max}(R)$ denoting the maximal eigenvalue of R. To make sense of the definition in Eqs. (A3) and (A4), we recall that a square root of a diagonalizable operator with real non-negative eigenvalues is well defined in an obvious way, so that $\sqrt{\rho}$ is well defined. To make sense of the outer square root we note that the operator ρ^* is Hermitian and has the same eigenvalues as ρ (this will be demonstrated later), which allows to recast $\sqrt{\rho}\rho^*\sqrt{\rho} = (\sqrt{\rho^*}\sqrt{\rho})^\dagger(\sqrt{\rho^*}\sqrt{\rho})$, so that the operator under the outer square root in Eq. (A3) is a product of an operator with its Hermitian conjugate, and is therefore Hermitian with non-negative eigenvalues. Equivalently, to simplify computations, the operator $R(\rho)$ in Eq. (A3) can be replaced with $\tilde{R}(\rho) = \rho\rho^*$. Indeed, if u is an eigenvector of $\sqrt{\rho}\rho^*\sqrt{\rho}$ with an eigenvalue λ, i.e., $\sqrt{\rho}\rho^*\sqrt{\rho}u = \lambda u$, applying the operator $\sqrt{\rho}$ to both sides results in $\rho\rho^*\sqrt{\rho}u = \lambda\sqrt{\rho}u$, i.e., $\sqrt{\rho}u$ is the eigenvector of $\rho\rho^*$ with the same eigenvalue λ. Note that the operator $\rho\rho^*$ is Hermitian with respect to the modified scalar product defined by $\langle\Psi'|\Psi\rangle_\rho = \langle\Psi'|\Psi|\Psi\rangle$.

The operation $\rho \mapsto \rho^*$ has a very nice coordinate free interpretation in terms of time reversal symmetry, which immediately shows that the Concurrence is LU invariant. Interpreting a qubit as a $1/2$ spin we consider a real structure on its space of states as an antilinear map $J: \mathbb{C}^2 \to \mathbb{C}^2$ that commutes with the action of SU(2). Such a map does exist, is defined up to a unimodular factor, and is responsible for time reversal symmetry. Naturally $J \otimes J$ represents the time reversal operator on the Hilbert space $\mathscr{H} = \mathbb{C}^2 \otimes \mathbb{C}^2$ of two qubits. Extending the time reversal symmetry operator to mixed states in a natural way $\rho \mapsto (J\otimes J)\rho(J\otimes J) = \rho^*$ we obtain an antilinear operator that is defined uniquely, commutes with the LU transformations, and represents time reversal symmetry. The compatibility of the time reversal operator with the LU transformations immediately implies the LU invariant nature of Concurrence, as well as provides a simple expression in terms of the correlation functions

$$(\rho(s, s', c))^* = \rho(-s, -s', c), \tag{A5}$$

which rationalizes why sometimes the $\rho \mapsto \rho^*$ is referred to as a spin flip operation.

1. Concurrence of a mixed *X* state of two qubits

We present the closed expression for the Concurrence $C(\rho)$ of a mixed X-state ρ, using various parameterizations of the state. The presented expressions are used to analyze the Concurrence created in a two-qubit system, excited by a beam of entangled photon pairs.

The density matrix of an X-state with pure correlations can be represented in the following standard form:

$$\begin{aligned}\rho = \frac{1}{4}\sigma_0\otimes\sigma_0 + \frac{1}{2}s\sigma_z\otimes\sigma_0 + \frac{1}{2}s'\sigma_0\otimes\sigma_z + \mu_z\sigma_z\otimes\sigma_z \\ + \mu\sigma_+\otimes\sigma_- + \mu^*\sigma_-\otimes\sigma_+ + \eta\sigma_+\otimes\sigma_+ \\ + \eta^*\sigma_-\otimes\sigma_-.\end{aligned} \tag{A6}$$

In the basis set $(|00\rangle, |11\rangle, |01\rangle, |10\rangle)$ the density matrix in Eq. (A6) becomes block-diagonal $\rho = \rho^{(0)} \oplus \rho^{(1)}$ with

$$\begin{aligned}\rho^{(0)} &= \begin{pmatrix} \frac{1}{4} + \mu_z + \frac{s+s'}{2} & \eta \\ \eta^* & \frac{1}{4} + \mu_z - \frac{s+s'}{2} \end{pmatrix}, \\ \rho^{(1)} &= \begin{pmatrix} \frac{1}{4} - \mu_z + \frac{s-s'}{2} & \mu \\ \mu^* & \frac{1}{4} - \mu_z - \frac{s-s'}{2} \end{pmatrix},\end{aligned} \tag{A7}$$

resulting in

$$\begin{aligned}(\rho^{(0)})^* &= \begin{pmatrix} \frac{1}{4} + \mu_z - \frac{s+s'}{2} & \eta \\ \eta^* & \frac{1}{4} + \mu_z + \frac{s+s'}{2} \end{pmatrix}, \\ (\rho^{(1)})^* &= \begin{pmatrix} \frac{1}{4} - \mu_z - \frac{s-s'}{2} & \mu \\ \mu^* & \frac{1}{4} - \mu_z + \frac{s-s'}{2} \end{pmatrix}.\end{aligned} \tag{A8}$$

Therefore, the matrix $\tilde{R} = \rho\rho^*$ is also block-diagonal, i.e., $\tilde{R} = \tilde{R}^{(0)} \oplus \tilde{R}^{(1)}$, with

$$\tilde{R}^{(0)} = \begin{pmatrix} \left(\frac{1}{4}+\mu_z\right)^2 - \left(\frac{s+s'}{2}\right)^2 + \eta^*\eta & 2\eta\left(\frac{1}{4}+\mu_z+\frac{s+s'}{2}\right) \\ 2\eta^*\left(\frac{1}{4}+\mu_z-\frac{s+s'}{2}\right) & \left(\frac{1}{4}+\mu_z\right)^2 - \left(\frac{s+s'}{2}\right)^2 + \eta^*\eta \end{pmatrix} \tag{A9}$$

and

$$\tilde{R}^{(1)} = \begin{pmatrix} \left(\frac{1}{4}-\mu_z\right)^2 - \left(\frac{s-s'}{2}\right)^2 + \eta^*\eta & 2\eta\left(\frac{1}{4}-\mu_z+\frac{s-s'}{2}\right) \\ 2\eta^*\left(\frac{1}{4}-\mu_z-\frac{s-s'}{2}\right) & \left(\frac{1}{4}-\mu_z\right)^2 - \left(\frac{s-s'}{2}\right)^2 + \eta^*\eta \end{pmatrix}. \tag{A10}$$

We have for the eigenvalues

$$\begin{aligned}\lambda_\pm(\rho^{(0)}) &= \frac{1}{4} + \mu_z \pm \sqrt{((s+s')/2)^2 + |\eta|^2}, \\ \lambda_\pm(\rho^{(1)}) &= \frac{1}{4} - \mu_z \pm \sqrt{((s-s')/2)^2 + |\eta|^2},\end{aligned} \tag{A11}$$

and

$$\begin{aligned}\lambda_\pm(\tilde{R}^{(0)}) &= \left(\sqrt{(1/4+\mu_z)^2 - ((s+s')/2)^2} \pm |\eta|\right)^2, \\ \lambda_\pm(\tilde{R}^{(1)}) &= \left(\sqrt{(1/4-\mu_z)^2 - ((s-s')/2)^2} \pm |\mu|\right)^2.\end{aligned} \tag{A12}$$

Based on Eq. (A11), the requirement of the non-negativity of the density matrix (i.e., all eigenvalues are non-negative) leads to $-(1/4) \le \mu_z \le (1/4)$, $-(1/2) - 2\mu_z \le s + s' \le (1/2) + 2\mu_z$, $-(1/2) + 2\mu_z \le s - s' \le (1/2) - 2\mu_z$, $|\eta| \le \sqrt{(1/4+\mu_z)^2 - ((s+s')/2)^2}$, and $|\mu| \le \sqrt{(1/4-\mu_z)^2 - ((s-s')/2)^2}$, while Eq. (A12) implies for the naive Concurrence

$$\begin{aligned}
\bar{C} &= 2\left(|\eta| - \sqrt{(1/4-\mu_z)^2 - ((s-s')/2)^2}\right) \\
&\quad \text{for } \sqrt{(1/4+\mu_z)^2 - ((s+s')/2)^2} + |\eta| \\
&\quad \geq \sqrt{(1/4-\mu_z)^2 - ((s-s')/2)^2} + |\mu|, \\
\bar{C} &= 2\left(|\mu| - \sqrt{(1/4+\mu_z)^2 - ((s+s')/2)^2}\right) \\
&\quad \text{for } \sqrt{(1/4+\mu_z)^2 - ((s+s')/2)^2} + |\eta| \\
&\quad \leq \sqrt{(1/4-\mu_z)^2 - ((s-s')/2)^2} + |\mu|
\end{aligned} \tag{A13}$$

so that Eq. (A13) provides explicit expressions for the Concurrence in the case of no mixed correlations.

Using the population-coherence variables the density matrix can be represented in a following standard form:

$$\begin{aligned}
\rho = n_{00}|00\rangle\langle 00| + n_{11}|11\rangle\langle 11| + n_{10}|10\rangle\langle 10| + n_{01}|01\rangle\langle 01| \\
+ \kappa|11\rangle\langle 00| + \kappa^*|00\rangle\langle 11| + \zeta|10\rangle\langle 01| + \zeta^*|01\rangle\langle 10|, \\
n_{00} + n_{11} + n_{10} + n_{01} = 1,
\end{aligned} \tag{A14}$$

and we immediately observe that the density matrix in Eq. (A14) has a form of the density matrix in Eq. (A6), with

$$\begin{aligned}
s &= \frac{1}{2}(-n_{00} + n_{11} + n_{10} - n_{01}), \\
s' &= \frac{1}{2}(-n_{00} + n_{11} - n_{10} + n_{01}), \\
\mu_z &= \frac{1}{4}(n_{00} + n_{11} - n_{10} - n_{01}), \\
\eta &= \kappa, \quad \mu = \zeta,
\end{aligned} \tag{A15}$$

resulting in

$$\begin{aligned}
(1/4+\mu_z)^2 - ((s+s')/2)^2 &= n_{00}n_{11}, \\
(1/4-\mu_z)^2 - ((s-s')/2)^2 &= n_{01}n_{10}.
\end{aligned} \tag{A16}$$

Upon substitution of Eqs. (A15) and (A16) into Eq. (A13), we arrive at the following explicit expressions for the Concurrence:

$$\begin{aligned}
\bar{C} &= 2\left(|\kappa| - \sqrt{n_{01}n_{10}}\right) \text{ for } |\kappa| + \sqrt{n_{00}n_{11}} \geq |\zeta| + \sqrt{n_{01}n_{10}}, \\
\bar{C} &= 2\left(|\zeta| - \sqrt{n_{00}n_{11}}\right) \text{ for } |\kappa| + \sqrt{n_{00}n_{11}} \leq |\zeta| + \sqrt{n_{01}n_{10}}
\end{aligned} \tag{A17}$$

with the explicit set of constraints for the parameters

$$\begin{aligned}
n_{00} + n_{11} + n_{01} + n_{10} = 1, \quad n_{jk} \geq 0 \quad \text{for } j,k = 0,1, \\
|\kappa| \leq \sqrt{n_{00}n_{11}}, \quad |\zeta| \leq \sqrt{n_{01}n_{10}}.
\end{aligned} \tag{A18}$$

In summary, Eq. (A17) provide explicit expressions for the Concurrence in terms of the matrix elements (populations and coherences) of the two-qubit density matrix in the case entangled photons driving, so that Concurrence can be easily analyzed by looking at the aforementioned populations and coherences.

APPENDIX B: PERTURBATIVE DIAGRAMMATIC EXPANSION OF THE QUBIT DENSITY MATRIX

Here, we present some details of the perturbation theory for computing the reduced density matrix of a two-qubit system, excited from its ground separable state by a beam of entangled photon pairs, created in an optically nonlinear PDC crystal via the parametric down-conversion process. In all the considered cases, we assume the field-qubit interaction to be in the perturbative regime, so the lowest-order effects are considered. The main focus is on the weak-pump regime that provides the most important qualitative results, presented in this paper, and does not require any further approximations, thus allowing to obtain the main results in a clean analytic way.

We consider field of entangled photon pairs produced by parametric down-conversion using a classical pump field and a birefringent crystal. The field is fully characterized by various correlations of the field operators that are encoded in the effective action S_{eff} of the field. Assuming a harmonic approximation for S_{eff}, all higher-order correlations can be expressed in terms of the two-point counterparts, also known as one-particle Greens' functions or propagators. There are two distinct one-particle propagators: the standard $\langle \hat{E}_1^\dagger(t_1)\hat{E}_2(t_2)\rangle$ and the anomalous $\langle \hat{E}_i^\dagger(t_1)\hat{E}_i^\dagger(t_2)\rangle, i = 1, 2$. In Eq. (5) of the main text, the amplitude of the two entangled-photon state is determined by the amplitude of the pump-field and $\chi^{(2)}$ nonlinearity of the crystal. Quantification of the entanglement between photon pairs generated in PDC process is readily done by introducing decomposition of the two-photon amplitude in terms of Schmidt basis defined in the individual photon spaces. If ψ_{ik} is the Schmidt basis in the ith photon space with r_k weight, the entanglement between the photons in a pair is defined in terms of the von Neumann entropy $E_{photon} = -\sum_k \lambda_k^2 \log(\lambda_k^2)$, where $\lambda_k = r_k/\sqrt{\sum_k r_k^2}$, and the field propagators can be recast as

$$\langle E_i^\dagger(\omega_1)E_i(\omega_2)\rangle = g_i(\omega_1,\omega_2), \tag{B1}$$

$$\langle E_1^\dagger(\omega_1)E_2^\dagger(\omega_2)\rangle = e^{-i\phi}h_{12}(\omega_1,\omega_2), \tag{B2}$$

where ϕ is the phase of the pump field and

$$g_i(\omega_1,\omega_2) = \sum_k \sinh^2(r_k)\psi_{ik}(\omega_1)\psi_{ik}^*(\omega_2), \tag{B3}$$

$$h_{12}(\omega_1,\omega_2) = \sum_k \sinh(r_k)\cosh(r_k)\psi_{1k}(\omega_1)\psi_{2k}(\omega_2). \tag{B4}$$

Note that the two-photon amplitude depends linearly on the pump field amplitude $\mathscr{E}_p = |\mathscr{E}_p|e^{i\phi}$ and, as a result, r_k depends linearly on $|\mathscr{E}_p|$.

The higher-order correlations are obtained using Wick's factorization. For example,

$$\begin{aligned}
&\langle E_1^\dagger(\omega_1)E_2^\dagger(\omega_2)E_1(\omega_3)E_2(\omega_4)\rangle \\
&\quad = g_1(\omega_1,\omega_3)g_2(\omega_2,\omega_4) + h_{12}(\omega_1,\omega_2)h_{12}^*(\omega_1,\omega_2),
\end{aligned} \tag{B5}$$

$$\begin{aligned}
&\langle E_1^\dagger(\omega_1)E_2^\dagger(\omega_2)E_2^\dagger(\omega_3)E_2(\omega_4)\rangle \\
&\quad = e^{-i\phi}[h_{12}(\omega_1,\omega_2)g_2(\omega_3,\omega_4) + h_{12}(\omega_1,\omega_3)g_2(\omega_2,\omega_4)],
\end{aligned} \tag{B6}$$

$$\begin{aligned}
&\langle E_i^\dagger(\omega_1)E_i^\dagger(\omega_2)E_i(\omega_3)E_i(\omega_4)\rangle \\
&\quad = g_i(\omega_1,\omega_3)g_i(\omega_2,\omega_4) + g_i(\omega_1,\omega_4)g_i(\omega_2,\omega_3).
\end{aligned} \tag{B7}$$

Note that propagators of the type $\langle E_1^\dagger(\omega_1)E_2(\omega_2)\rangle = \langle E_i^\dagger(\omega_1)E_i^\dagger(\omega_2)\rangle = 0$.

It is difficult to find analytic form of the Schmidt basis for a general pump field. However, for a Gaussian pump, $\mathscr{E}_p(\omega) = \mathscr{E}_p e^{-(\omega-\omega_p)^2/(2\sigma_p^2)}$, Schmidt modes can be approximated using

Hermite functions.[28] This allows to obtain analytic expressions for the field propagators.[28,29] Important parameters that determine the field propagators are the pump-field spectral width, σ_p, the maximal time-delays T_1 and T_2 of the two entangled photons with respect to the pump pulse maximum, and the central frequencies (ω_0 and $\bar{\omega}_0$) of the two entangled photon wavepackets. The Schmidt modes are obtained in terms of Hermite functions, $\psi_{1k}(\omega) = H_k[k_{a_1}(\omega - \Omega_0)]$ and $\psi_{2k}(\omega) = H_k[k_{a_2}(\omega - \Omega'_0)]$ where $H_k(k_{a_i}x) = \sqrt{(k_{a_i}/2^k k!\sqrt{\pi})}e^{-k_{a_i}^2 x^2/2 - i(3\pi/8)} \times h_k(k_{a_i}x)$ with $h_n(k_{a_i}x)$ being the Hermite polynomial of order k and

$$k_{a_i} = \sqrt{2a_i\frac{1-\mu^2}{1+\mu^2}}, \quad a_i = \gamma T_i^2 + \frac{1}{2\sigma_p^2}, \quad c = \gamma T_1 T_2 + \frac{1}{2\sigma_p^2}, \tag{B8}$$

$$\mu = \frac{1}{c}\left(\sqrt{a_1 a_2 - c^2} - \sqrt{a_1 a_2}\right), \tag{B9}$$

and $\gamma = 0.048\,22$. Note that $-1 < \mu < 0$, $r_k = (\chi^{(2)}\mathscr{E}_p/\hbar)\mu^k\sqrt{(\pi/2)(1+\mu^2)/\sqrt{a_1 a_2}}$, and thus $\lambda_k = \mu^k\sqrt{1-\mu^2}$.

1. Perturbative calculation for the reduced atomic state

The combined light–matter system evolves in time according to the Schrodinger equation. The state of the atomic system is determined by the reduced density-matrix obtained upon tracing out the field degrees of freedom and is formally represented as a vector $|\rho(t)\rangle\rangle$ in Liouville space whose evolution in the interaction picture is given by,[29,30]

$$\begin{aligned} |\rho(t)\rangle\rangle &= \mathrm{Tr}_F \mathscr{T} e^{-\frac{i}{\hbar}\int_{t_0}^t d\tau \hat{H}'_-(\tau)}|\rho(t_0)\rangle\rangle \\ &= \mathrm{Tr}_F \sum_{n=0}^{\infty}\left(\frac{-i}{\hbar}\right)^n \int_{t_0}^{t} d\tau_n \cdots \int_{t_0}^{\tau_2} d\tau_1 \\ &\quad \times \hat{\mathscr{T}}\hat{H}'_-(\tau_n)\cdots\hat{H}'_-(\tau_1)|\rho(t_0)\rangle\rangle, \end{aligned} \tag{B10}$$

where $\mathscr{T}$ is time-ordering superoperator which re-arranges a product of time-dependent superoperators so that time increases from right to left, and Tr_F represents a trace over the field degrees of freedom and the time-dependence of operators is in the interaction picture. The superoperator $\hat{H}'_- = \hat{H}_L - \hat{H}_R$ is defined in terms of the left and the right superoperators, acting on the ket and bra of the density matrix. Equation (B10) allows us to compute the state of the atomic system perturbatively in the interaction with light.

Elements of the density-matrix in the product basis of the two qubits are denoted as $n_{xy;x'y'} = \langle\langle xy; x'y'|\rho(t)\rangle\rangle \equiv \langle xy|\rho(t)|x'y'\rangle$, where $x, y, x', y' = 0, 1$. These elements can be obtained from Eq. (B10). For example, to leading order in the interaction between the field and qubits, the element $n_{0000}(t)$ is given by

$$\begin{aligned} n_{0000}(t) = 1 &- \frac{1}{\hbar^2}\int d\tau_1 d\tau_2 \sum_{\alpha=L,R}(-1)^{N_R}\langle\langle 11;11|\mathscr{T}\hat{d}_\alpha(\tau_1)\hat{d}^\dagger_\alpha(\tau_2)|\rho(0)\rangle\rangle \\ &\times\langle\langle\hat{I}|\mathscr{T}\hat{E}^\dagger_\alpha(\tau_1)\hat{E}_\alpha(\tau_2)|\rho_F\rangle\rangle + \frac{1}{\hbar^4}\int d\tau_1 d\tau_2 d\tau_3 d\tau_4 \\ &\times\sum_{\alpha\beta}(-1)^{N_R}\langle\langle 11;11|\mathscr{T}\hat{d}^\dagger_\alpha(\tau_1)\hat{d}_\alpha(\tau_2)\hat{d}^\dagger_\beta(\tau_3)\hat{d}_\beta(\tau_4)|\rho(0)\rangle\rangle \\ &\times\langle\langle\hat{I}|\mathscr{T}\hat{E}_\alpha(\tau_1)\hat{E}^\dagger_\alpha(\tau_2)\hat{E}_\beta(\tau_3)\hat{E}^\dagger_\beta(\tau_4)|\rho_F\rangle\rangle, \end{aligned} \tag{B11}$$

where N_R is the total number of right (R) superoperators appearing in the field propagator. Equation (B10) denoted qubit state to fourth-order in the interaction with the entangled field. Note that the field propagators of the type $\langle\hat{E}^\dagger_i(t_1)\hat{E}^\dagger_i(t_2)\rangle$ and $\langle\hat{E}^\dagger_i(t_1)\hat{E}_j(t_2)\rangle$, where $i \neq j$, do not contribute as they are identically zero. An explicit expression for the density matrix elements in terms of field correlations is given below. To simplify notations, in the following, we denote population of the state $|00\rangle$ by $n_{0000} \equiv n_{00}$, and similarly for other states by n_{01}, n_{10}, n_{11},

$$\begin{aligned} n_{00} = 1 &- \frac{|d|^2}{2\hbar^2}\langle\hat{E}^\dagger_1(\Omega_0)\hat{E}_1(\Omega_0)\rangle \\ &+ \frac{|d|^4}{2\hbar^4}\langle\hat{E}^\dagger_1(\Omega_0)\hat{E}^\dagger_1(\Omega_0)\hat{E}_1(\Omega_0)\hat{E}_1(\Omega_0)\rangle \\ &+ \frac{3}{2}\frac{|d|^2|d'|^2}{\hbar^4}\langle\hat{E}^\dagger_2(\Omega'_0)\hat{E}^\dagger_1(\Omega_0)\hat{E}_2(\Omega'_0)\hat{E}_1(\Omega_0)\rangle \\ &+ (d, \Omega_0 \Longleftrightarrow d', \Omega'_0). \end{aligned} \tag{B12}$$

In deriving the above expression, we have used $\hat{E}(\tau) = \int d\omega e^{-i\omega\tau}\hat{E}(\omega)$ and ignored the principal part contribution in the identity, $(1/\omega + i\eta) = \mathscr{P}(1/\omega) - i\pi\delta(\omega)$ with $\eta \to 0^+$. The other diagonal matrix elements are similarly obtained as

$$\begin{aligned} n_{01} &= \frac{|d'|^2}{2\hbar^2}\langle\hat{E}^\dagger_2(\Omega'_0)\hat{E}_2(\Omega'_0)\rangle \\ &\quad - \frac{3}{2}\frac{|d|^2|d'|^2}{\hbar^4}\langle\hat{E}^\dagger_2(\Omega'_0)\hat{E}^\dagger_1(\Omega_0)\hat{E}_2(\Omega'_0)\hat{E}_1(\Omega_0)\rangle \\ &\quad - \frac{|d'|^4}{2\hbar^4}\langle\hat{E}^\dagger_2(\Omega'_0)\hat{E}_2{}^\dagger(\Omega'_0)\hat{E}_2(\Omega'_0)\hat{E}_2(\Omega'_0)\rangle, \\ n_{11} &= \frac{3}{2}\frac{|d|^2|d'|^2}{\hbar^4}\langle\hat{E}^\dagger_2(\Omega'_0)\hat{E}^\dagger_1(\Omega_0)\hat{E}_2(\Omega'_0)\hat{E}_1(\Omega_0)\rangle, \end{aligned} \tag{B13}$$

and n_{10} is obtained by interchanging $(d, \Omega) \Longleftrightarrow (d', \bar{\Omega}_{12})$ in the expression for n_{01}.

The only surviving coherences are between states $|a\rangle$ and $|d\rangle$ given by

$$\begin{aligned} \kappa(t) &= \frac{3}{4}\frac{dd'}{\hbar^4}e^{i(\Omega_0+\Omega'_0)t}\Big(|d|^2\langle\hat{E}^\dagger_2(\Omega'_0)\hat{E}^\dagger_1(\Omega_0)\hat{E}^\dagger_1(\Omega_0)\hat{E}_1(\Omega_0)\rangle \\ &\quad + |d'|^2\langle\hat{E}^\dagger_1(\Omega_0)\hat{E}^\dagger_2(\Omega'_0)\hat{E}^\dagger_2(\Omega'_0)\hat{E}_2(\Omega'_0)\rangle\Big) \\ &\quad - \frac{dd'}{4\hbar^2}e^{i(\Omega_0+\Omega'_0)t}\Big(\langle\hat{E}^\dagger_1(\Omega_0)\hat{E}^\dagger_2(\Omega'_0)\rangle + \langle\hat{E}^\dagger_2(\Omega'_0)\hat{E}^\dagger_1(\Omega_0)\rangle\Big). \end{aligned} \tag{B14}$$

Note that only element κ involves photon correlations of unequal number of field creation and annihilation operators.

We compute the qubit state perturbatively to lowest-order in the pump-field and the interaction of entangled light-field with the qubits. This requires expanding the field propagators in the pump-field. Since fourth- and higher-order field propagators can be evaluated in terms of the (second-order) basic propagators using Wick's factorization, it, in principle, allows us to generate a diagrammatic perturbation expansion in terms of the basic propagators. This amounts to expanding S_{eff} around $S^{(0)}_{eff}$ and results in the perturbative diagrams in the pump field amplitude. All the physical processes that contribute to the evolution of the qubit state at the

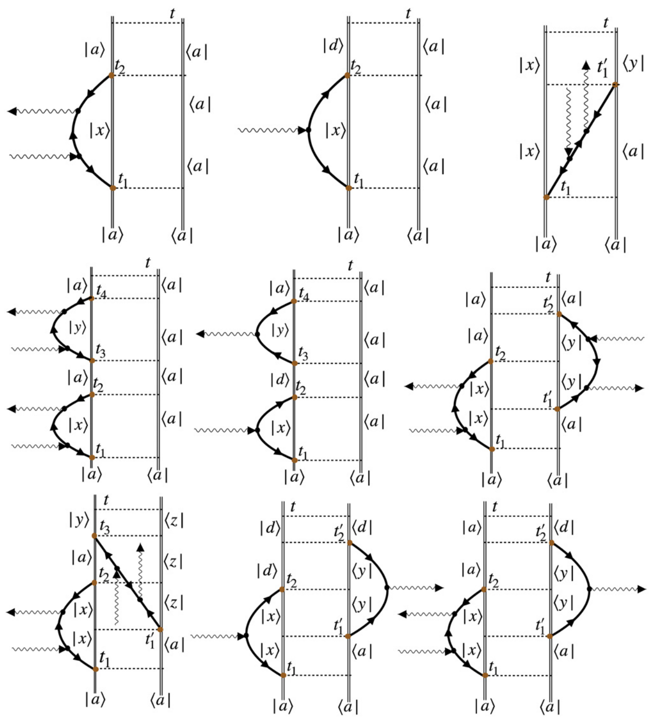

Fig. 3. Representative diagrams in the perturbation theory in the pump-field and (qubit) system-entangled field interactions. Top panel: second-order and lower panels: fourth-order interaction diagrams that contribute in time-evolution of the reduced system density matrix. States $|a\rangle$ and $|d\rangle$ denote the ground and doubly excited states $|00\rangle$ and $|11\rangle$, respectively, and $|x\rangle, |y\rangle, |z\rangle$ denote single-excited states $|01\rangle$ and $|10\rangle$. Thick black lines are propagators for the entangled field where an arrow pointing toward (away) the vertex (brown dots) denotes field annihilation (creation) operator $\hat{E}(\hat{E}^\dagger)$. The field propagator is computed with respect to the affective action S^0_{eff} (see the text). The whole set of diagrams can be obtained by considering propagators connecting to vertices not connected in the above diagrams.

lowest-order are shown diagrammatically in Fig. 3. To lowest order in pump-field, the basic propagators for the entangled field are obtained by replacing $g_1 \approx |\chi^{(2)}\mathscr{E}_p/\hbar|^2\sqrt{\pi/2a_2}$, $g_2 \approx \sqrt{(a_2/a_1)}g_1$, and $h_{12} \approx |(\chi^{(2)}\mathscr{E}_p/\hbar)|$ in in Eqs. (B1) and (B2). This provides the results used in Eqs. (6)–(9) of the main text.

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