Bell inequalities and density matrix for polarization-entangled photons out of a two-photon cascade in a single quantum dot

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We theoretically investigate the joint photodetection probabilities of the biexciton-exciton cascade in single semiconductor quantum dots and analytically derive the density matrix and the Bell's inequalities of the entangled state. Our model includes different mechanisms that may spoil or even destroy entanglement such as dephasing, energy splitting of the relay excitonic states, and incoherent population exchange between these relay levels. We explicitly relate the fidelity of entanglement to the dynamics of these processes and derive a threshold for violation of Bell's inequalities. Applied to standard InAs/GaAs self-assembled quantum dots, our model indicates that spontaneous emission enhancement of the excitonic states by cavity effects increases the fidelity of entanglement to a value allowing for violation of Bell's inequalities.

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I. INTRODUCTION

Entangled photon pairs are an essential tool for quantum information science, ranging from quantum cryptography [1], to the realization of quantum relays [2,3] or quantum information processing [4,5]. Quantum relays are probably one of the most advanced applications using entanglement and have been implemented in real world quantum teleportation setups [6,7] or entanglement swapping demonstrations [8,9]. In these experiments, entangled photons were obtained by parametric down conversion, but other sources based on four-wave mixing are also investigated. Such nonlinear sources of entanglement can combine narrow spectral bandwidths with a maximal generation rate [9–11]. However, although these sources may be very useful and easy to implement, they always suffer from the Poissonian statistics of the emitted photons pairs leading to multipair emission, which decreases the visibility of entanglement [12]. The need to minimize the likelihood of producing multiple photon pairs forces these sources to be operated at low rates of photon pair generation per coherence length or excitation pulse (usually lower than 0.1). On the other hand, a deterministic source of entangled photons would make it possible to suppress these multipair events and to create light pulses with increased probability of containing a single photon pair, hence rendering all the above mentioned protocols much more efficient. From this point of view, sources based on the cascade emission from a single dipole (such as a single atom or a single quantum dot) may be a good candidate. In such a system, the single dipole can be described as a four-level system emitting a single pair of photons upon each excitation cycle. For example, in self-assembled quantum dots, this cascade emission involves a biexciton, which consists of twoelectron-hole pairs trapped in the dot with opposite angular momentum and which decays radiatively through two relay bright exciton [13,14]. This decay may release time-binentangled photons [15], or polarization-entangled photons [13]. Time-bin-entangled photons can also be obtained from two successive indistinguishable single photons [16]. The origin of polarization entanglement here resides in the existence of two radiative decay paths with different polarizations which are otherwise indistinguishable. However, in such solid-state single emitters, polarization entanglement is spoiled by the anisotropic exchange interaction caused by in-plane anisotropy of the exciton wave function [17,18]; such electron-hole exchange interaction lifts the excitonic states degeneracy and provides information about which pathways the two photons were released along via the energy of the emitted photons [19]. Reducing the excitonic energy splitting within the radiative linewidth of the excitonic levels (by spectral filtering [20], use of external magnetic [21] or electric [22] field, growth optimization [23],...) can, in principle, allow us to erase the which path information due to the excitonic fine structure and recover entanglement. However, dephasing interactions with the solid-state environment (for example, through collisions with phonons and electrostatic interactions with fluctuating charges located in the dipole vicinity [24]) may also degrade the strong correlations between the polarization of the two photons. Moreover, any incoherent mechanisms inducing a population exchange between the excitonic levels (such as transitions through the dark states or spin-flip processes) may deteriorate the visibility of entanglement.

This paper theoretically investigates the joint photodetection probabilities in the biexciton cascade and analytically derives the density matrix as well as a nonoptimal but nevertheless interesting entanglement witness based on the Clauser-Horne-Shimony-Holt (CHSH) inequalities. Several incoherent process have been taken into account such as exciton energy splitting, incoherent population exchange between the excitonic levels, and cross dephasing between these two relay states. The following part of the paper begins by defining a Hamiltonian of a four-level system interacting with a solid-state environment and subject to incoherent population exchange between the two relay levels. We derive from such Hamiltonian a time evolution equation of the system excited on its upper state and derive the joint photodetection probability. In Sec. III, we quantify the entanglement of the photons produced by deriving an analytical expression of the Clauser-Horne-Shimony-Holt (CHSH) inequality as a function of the different dynamical parameters of the fourlevel system, as well as the density matrix corresponding to



FIG. 1. Schematic description of the two-photon cascade in a typical four-level system with an energy splitting $2\hbar \delta \omega$ of the relay level, yielding two collinearly polarized photons (either *H* or *V*).

the biexciton cascade. We then stress in Sec. IV the necessity to make use of the Purcell effect [25], in order to violate Bell's inequalities from the cascade emission in self-assembled quantum dots.

II. THEORETICAL FRAMEWORK

A. Four-level system

In the cascade emission from a four-level system, the decay paths involve two radiative transitions, one from an upper level $|2\rangle$ to an intermediate state $|1_H\rangle$ or $|1_V\rangle$, and the other from these relay states to the ground state $|0\rangle$ (see Fig. 1). The energies of these levels $|2\rangle$, $|1_H\rangle$, and $|1_V\rangle$ are, respectively, denoted $\hbar(\omega_1 + \omega_2)$, $\hbar(\omega_1 + \delta\omega)$, and $\hbar(\omega_1 - \delta\omega)$. We will further assume that this $\{|2\rangle, |1_H\rangle, |1_V\rangle, |0\rangle$ basis corresponds to the eigenbasis of the quantum dot, with therefore an excitonic energy splitting $2\delta\omega$ but no coherent coupling between the two excitonic eigenstates [19]. Radiative transitions from the biexciton in such basis release collinearly polarized photons with linear polarization denoted *H* and *V* (see Fig. 1). In the ideal case ($\delta\omega$ =0), the four-level system relaxes, generating the maximally entangled two-photon state

$$|\Phi^{+}\rangle = \frac{1}{\sqrt{2}}(|H,\omega_{1}\rangle|H,\omega_{2}\rangle + |V,\omega_{1}\rangle|V,\omega_{2}\rangle), \qquad (1)$$

by cascade emission [13,26]. The phase difference between the two component states $|H, \omega_1\rangle |H, \omega_2\rangle$ and $|V, \omega_1\rangle |V, \omega_2\rangle$ is null, as determined by the angular momenta of the different involved levels and the Clebsch-Gordan coefficients [27,28]. Unfortunately, in realistic two-level systems (such as single quantum dots for example), the relay levels are split ($\delta \omega \neq 0$). Furthermore, relaxation mechanisms between the two relay states $|1_H\rangle$ and $|1_V\rangle$ can occur (for example, from spinflip processes). They will be accounted for by two phenomenological decay rates $\Gamma_{flip} \pm \delta \Gamma_{flip}$, which will be later supposed to be equals (which is a good approximation for a small excitonic energy splitting).

In addition, the relay levels and the upper level may be subject to sudden, brief and random fluctuations of their energies without population exchange (arising, for example, from collisions with thermal phonons). In our model, the ground level $|0\rangle$ is chosen as the reference in energy and phase. Dephasing of the upper level $|2\rangle$ is described by the dephasing rate Γ_2 . On the two relay levels $|1_H\rangle$ and $|1_V\rangle$, we distinguish two dephasing processes without population exchange between these relay levels: (1) dephasing processes that occur simultaneously and attach the same information on the phase and energy of these two levels with a dephasing rate denoted Γ_1 ; and (2) dephasing processes that do not affect identically the two relay levels and whose impact depends on the polarization of the excitonic states. These last processes will be described by polarization-dependent dephasing rates Γ_H and Γ_V . The cross dephasing between the two relay states is therefore $\Gamma = \Gamma_H + \Gamma_V$. This model includes all possible dephasing processes without population modifications that may occur.

B. Dynamics of the four-level system

In order to account for the open nature of the four-level system (resulting from its coupling with the phonon and the photon reservoirs for example), we describe the time evolution of the density operator ρ by means of the following master equation in the Lindblad form [29]:

$$\frac{d\rho}{dt} = -[iH,\rho] + (\mathcal{L}_r + \mathcal{L}_d + \mathcal{L}_{flip})\rho.$$
(2)

In the previously described eigenbasis $\{|2\rangle, |1_H\rangle, |1_V\rangle, |0\rangle\}$ of the four-level system, the Hamiltonian *H* has the form

$$H = (\omega_1 - \delta\omega)|1_V\rangle\langle 1_V| + (\omega_1 + \delta\omega)|1_H\rangle\langle 1_H| + (\omega_1 + \omega_2)|2\rangle\langle 2|.$$
(3)

The Lindblad operators include three contributions. The first one describes the interaction of the emitter with the electromagnetic field by emitting photons, whenever it undergoes a transition from its upper state to the relay levels or from the relay levels to the ground state. This radiative relaxation is accounted for by the following Liouvillian:

J

$$\mathcal{L}_{r} = \sum_{p=H,V} \left\lfloor \frac{\gamma_{1}}{2} \mathcal{L}(|0\rangle\langle 1_{p}|) + \frac{\gamma_{2}}{2} \mathcal{L}(|1_{p}\rangle\langle 2|) \right\rfloor, \tag{4}$$

where γ_1 and γ_2 are, respectively, the radiative decay rates between the relay states and the ground state and between the upper level and the relay levels. We assume that these decay rates do not depend on the decay path the photons were released along. $\mathcal{L}(D)\rho=2D\rho D^{\dagger}-D^{\dagger}D\rho-\rho D^{\dagger}D$ is the Lindblad operator. The second contribution \mathcal{L}_d is related to dephasing processes and reads

$$\mathcal{L}_{d} = \Gamma_{2}\mathcal{L}(|2\rangle\langle 2|) + \sum_{p=H,V} \Gamma_{p}\mathcal{L}(|1_{p}\rangle\langle 1_{p}|) + \Gamma_{1}\mathcal{L}(|1_{H}\rangle\langle 1_{H}| + |1_{V}\rangle\langle 1_{V}|).$$
(5)

This Liouvillian includes, phenomenologically, any dephasing effect (e.g., phonons) occurring on the levels of the dot without population transfers as described previously. The last contribution \mathcal{L}_{flip} accounts for the incoherent coupling between the two relay states

The phenomenological rate Γ_{flip} between the two relay states $|1_{H}\rangle$ and $|1_{V}\rangle$ appears to be twice the sum of the different rates α_i and β_i (i=P,Q) involved in this equation. The rate $\delta\Gamma_{flip}$ is expressed likewise as $2(\alpha_Q - \alpha_P + \beta_Q - \beta_P)$. These rates simulate any unspecified process inducing an incoherent interaction between the two relay levels with population exchange before radiative relaxation. $\delta\Gamma_{flip}$ accounts for asymmetry of these processes. These nonradiative processes may include spin-flip processes and transitions through dark states (assuming that the probability for the source to be in these dark states is small compared to the probabilities related to the optically active states).

In accordance with all these assumptions, the time evolution of the four-level system can be decomposed by use of master equation (2) in a set of differential equations, which reduces for the purpose of this paper to

$$\frac{dV}{dt} = AV. \tag{7}$$

In this equation, V is a vector composed of the following mean values:

$$V = \begin{pmatrix} \operatorname{Tr}[S_{\Delta}\rho] \\ \operatorname{Tr}[S_{Q}\rho] \\ \operatorname{Tr}[S_{P}\rho] \end{pmatrix}.$$
(8)

The operator $S_{\Delta} = |1_H\rangle\langle 1_H| - |1_V\rangle\langle 1_V|$ is related to population difference between the two excitonic relay states. The two other Pauli matrices $S_Q = i|1_V\rangle\langle 1_H| - i|1_H\rangle\langle 1_V|$ and S_P $= |1_H\rangle\langle 1_V| + |1_V\rangle\langle 1_H|$ correspond to the quadratures of the dipole between these two relay states. The A matrix is given by

$$A = \begin{pmatrix} -\gamma_1 - 2\Gamma_{flip} & 0 & 0\\ 0 & -\mu_Q & 2\delta\omega\\ 0 & -2\delta\omega & -\mu_P \end{pmatrix}.$$
 (9)

The decay constants $\mu_{P/Q}$ are equal to $\gamma_1 + \Gamma_{flip} \pm \delta \Gamma_{flip} + \Gamma$.

For further reference, we define the matrix transformation M(U) of V, where U is an arbitrary unitary transformation of the excitonic levels of the source (letting the upper and fundamental states remain unchanged), by

$$M(U)V(t) = \begin{pmatrix} \operatorname{Tr}[US_{\Delta}U^{\dagger}\rho(t)] \\ \operatorname{Tr}[US_{Q}U^{\dagger}\rho(t)] \\ \operatorname{Tr}[US_{P}U^{\dagger}\rho(t)] \end{pmatrix}.$$
 (10)

M(U)V(t) are the mean values (8) measured at time t under the transformed basis $\{|2\rangle, U|1_H\rangle, U|1_V\rangle, |0\rangle\}$.

C. Joint photodetection probability

Violation of Bell inequalities as well as the reconstruction of the density matrix is experimentally obtained by measur-



FIG. 2. Experimental setup for measuring CHSH or reconstructing the density matrix.

ing the joint photon detection probabilities $P_{\pm,\pm}$ on the output of a binary polarization analyzer such as a polarizing beam splitter. Since several points of the Bloch sphere have to be measured [26], a quarter-wave plate followed by a half-wave plate are inserted in the photons path (see Fig. 2). The exciton and biexciton photons are spectrally separated by means of optical filters and send through the optical path denoted *i* (*i*=1 or 2 for the exciton and biexciton, respectively).

The fast axis of the quarter- (respectively, half-) wave plate is rotated by an angle χ_i (respectively, θ_i) with respect to the horizontal polarization direction defined by the optical table. By applying the projection theorem, measuring +1 in the optical setup *i* corresponds to the detection of a photon *i* emitted by the source with the polarization $\Lambda(\theta_i, \chi_i)^{\dagger} | H \rangle$, where $\Lambda(\theta, \chi)$ describes the transformation of the polarization basis $\{H, V\}$ when a photon successively propagates through a quarter- and a half-wave plate rotated by the angles θ and χ .

$$\Lambda(\theta, \chi) = R(\theta)T(\pi)R(\chi - \theta)T(\pi/2)R(-\chi), \qquad (11)$$

where R(x) is the rotation matrix and T(r) is the Jones matrix of a retarder plate.

$$R(x) = \begin{pmatrix} \cos(x) & \sin(x) \\ -\sin(x) & \cos(x) \end{pmatrix}, \quad T(r) = \begin{pmatrix} 1 & 0 \\ 0 & e^{-ir} \end{pmatrix}.$$
 (12)

In the following, for the sake of clarity we will denote $\Lambda(\theta, \chi)^{\dagger}|1_{H}\rangle$ the superposition of the source's states $|1_{H}\rangle$ and $|1_{V}\rangle$, which analytically corresponds to the same transformation $\Lambda(\theta, \chi)^{\dagger}|H\rangle$ of the photonic state $|H\rangle$.

Experimentally one measures the joint photodetection probabilities $P_{\pm,\pm}^{\text{det}}(\theta_2,\chi_2,\theta_1,\chi_1)$ of the first photon and second photon in channels \pm of their respective optical setups with each retarder plate rotated by θ_i and χ_i .

The source is pumped at time t=0 from its ground state to the excited state $|2\rangle$ with a laser pulse shorter than the lifetime $1/(2\gamma_2)$ of the upper state. We will further postselect joint photodetection events corresponding to a sequential detection of the biexcitonic photon and then of the excitonic photon during one excitation cycle. In this context, the probability of joint photodetection $P_{+,+}^{det}(\theta_2, \chi_2, \theta_1, \chi_1)$ is proportional to the emission probability $P_{+,+}(\theta_2, \chi_2, \theta_1, \chi_1)$ of a pair of photons with respective polarization orientation $\Lambda(\theta_i,\chi_i)^{\dagger}|H\rangle$, at respective energies $\hbar\omega_2$ and $\hbar\omega_1$, assuming that the source is in state $|2\rangle$ at time t=0. This radiative transition probability can be regarded as the product of two probabilities: the probability of emission of the first photon with polarization $\Lambda(\theta_2, \chi_2)^{\dagger} | H \rangle$, multiplied by the conditional probability of radiative transition from the relay levels to the ground state with emission of a photon polarized along $\Lambda(\theta_1,\chi_1)^{\dagger}|H\rangle$. This amounts to considering the photon cascade as a two-step process and applying the quantummeasurement projection postulate. First the photon at energy $\hbar \omega_2$ and polarized along $\Lambda(\theta_2, \chi_2)^{\dagger} | H \rangle$ is detected at time t_2 , which projects the emitter on the superposition $\Lambda(\theta_2,\chi_2)^{*\dagger}|1_H\rangle$ of the exciton states $|1_H\rangle$ and $|1_V\rangle$. Second, the superposition state evolves in time until the detection of the second photon at energy $\hbar \omega_1$ at time $t_1 + t_2$. Consequently, this conditional probability will be related to the population in the superposition $\Lambda(\theta_1, \chi_1)^{\dagger} |1_H\rangle$ at time $t_2 + t_1$, knowing that the intermediate levels were in the superposition $\Lambda(\theta_2,\chi_2)^{*\dagger}|1_H\rangle$ at time t_2 . All these probabilities are integrated over the photodetection time window.

The population at time t_1+t_2 in the superposition $|1_H(\theta_1X_1)\rangle$ can be expressed as $[e^{-\gamma_1 t_1}+\langle S_\Delta\rangle(t_2+t_1|t_2)]/2$, where $\langle S_\Delta\rangle(t_2+t_1|t_2)$ is the first value of the vector V of Eq. (8) measured under the transformation of Eq. (10) with $U = \Lambda(\theta_1,\chi_1)^{\dagger}$, after a free evolution during the time t_1 [Eq. (7)] with the assumption of the initial state V^{init} corresponding to the excitonic state $\Lambda(\theta_2,\chi_2)^{*\dagger}|H\rangle$ at time t_2 . Thus by defining the vector $V_0=\{1,0,0\}$, which corresponds to the values of V measured in the eigenbasis with the source in the state $|1_H\rangle$, it follows that

$$V^{init} = M(\Lambda(\theta_2, \chi_2)^{*\dagger})^{-1} V_0,$$

$$V^{measured} = M(\Lambda(\theta_1, \chi_1)^{\dagger}) e^{At} V^{init},$$

$$\langle (t_1|0) = \langle S_\Delta \rangle (t_2 + t_1|t_2)$$

$$= [M(\Lambda(\theta_1, \chi_1)^{\dagger}) e^{At} M(\Lambda(\theta_2, \chi_2)^{*\dagger})^{-1}]_{11},$$
(13)

where $[\ldots]_{ij}$ denotes the matrix element on row *i* and column *j*. The probability $P_{+,+}(\theta_2, \chi_2, \theta_1, \chi_1)$ can therefore be written as follows:

 $\langle S_{\Delta} \rangle$

$$P_{+,+}(\theta_2, \chi_2, \theta_1, \chi_1) = \int_0^{+\infty} \gamma_2 e^{-2\gamma_2 t_2} dt_2$$
$$\times \int_0^{+\infty} \frac{\gamma_1}{2} (e^{-\gamma_1 t_1} + \langle S_\Delta \rangle (t_1|0)) dt_1.$$
(14)

Upon integration, this probability reads in the particular case $\chi_1 = \chi_2 = 0$ as follows:

$$P_{+,+}(\theta_2, \theta_1) = \frac{1}{4} \left[1 + \frac{\gamma_1}{\gamma_1 + 2\Gamma_{flip}} \cos(4\theta_1)\cos(4\theta_2) + \frac{\gamma_1(\gamma_1 + \Gamma_{flip} + \Gamma - \delta\Gamma_{flip})}{(2\,\delta\omega)^2 + (\gamma_1 + \Gamma_{flip} + \Gamma)^2 - (\delta\Gamma_{flip})^2} \times \sin(4\theta_1)\sin(4\theta_2) \right].$$
(15)

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For a perfect quantum dot, $P_{+,+}(0,0)$ tends toward 1/2 as expected.

III. QUANTIFYING TWO-PHOTON ENTANGLEMENT AND DENSITY MATRIX

Entanglement can be quantified by several means such as measurement of the concurrence, tangle of the density matrix, or entanglement witness operators. A nonoptimal entanglement witness, but nevertheless experimentally simple to measure, is the Bell inequality under the CHSH form, which discriminates between states that can be explained by a local hidden variable model (LHVM) or not. The possible violation of Bell inequalities is experimentally easy to verify by measuring the fringes visibility [30] of two-photon coincidences as a function of $(\theta_1 - \theta_2)$, whereas other measurements need the experimental knowledge of the density matrix. Hence we shall first derive the analytical form of the CHSH inequality, then generalize the result to the derivation of the density matrix and one possible entanglement witness [31] by use of the Peres criterion [32].

A. Violation of Bell's inequalities

The CSHS inequality is calculated by measuring the correlation coefficient for four sets of properly chosen angles of a half-wave plate, and therefore the angles χ_i referring to the quarter-wave plate are set to zero and omitted in the rest of this section. From the expression of Eq. (15) one deduces all the probabilities $P_{\pm,\pm}(\theta_2, \theta_1)$ and compute analytically in a straightforward manner the correlation coefficient of the form

$$E(\theta_2, \theta_1) = P_{+,+}(\theta_2, \theta_1) + P_{-,-}(\theta_2, \theta_1) - P_{-,+}(\theta_2, \theta_1) - P_{+,-}(\theta_2, \theta_1).$$
(16)

The generalized Bell's inequality in the Clauser-Horne-Shimony-Holt (CHSH) formulation [33,34] is expressed as a combination of such correlations functions as

$$S(\theta_2, \theta'_2, \theta_1, \theta'_1) = E(\theta_2, \theta_1) - E(\theta'_2, \theta_1) + E(\theta_2, \theta'_1) + E(\theta'_2, \theta'_1),$$
(17)

which, for classically correlated states, is bounded by $|S| \le 2$. In the case of an ideal entangled source, the maximum value of the CHSH coefficient *S* is obtained for every set of polarization directions of each analyzer verifying $\theta_2 = x + \pi/16$ and $\theta'_2 = x + 3\pi/16$; $\theta_1 = x$ and $\theta'_1 = x + \pi/8$, where *x* is an arbitrary rotation of both half-wave plates. In this context and under the assumption of $\delta\Gamma_{flip}=0$, the CHSH parameter *S* is given by the formula

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$$S = \sqrt{2} \left(\frac{\gamma_1}{\gamma_1 + 2\Gamma_{flip}} + \frac{\gamma_1(\gamma_1 + \Gamma_{flip} + \Gamma)}{(\gamma_1 + \Gamma_{flip} + \Gamma)^2 + (2\delta\omega)^2} \right), \quad (18)$$

which is, as expected, independent of the arbitrary rotation x. Violation of Bell's inequalities implies S > 2.

B. Density matrix

In the above section, the Bell inequalities have been derived from the coincidence probabilities when the exciton and biexciton are detected in a linear basis. We will now exploit the more general expression of the joint photodetection probability $P_{+,+}(\theta_2, \chi_2, \theta_1, \chi_1)$ obtained upon integration of Eq. (14). By definition of the density matrix, this probability can also be expressed as

$$P^{\rho}_{+,+}(\theta_2,\chi_2,\theta_1,\chi_1) = \langle H_{XX}H_X | [\Lambda(\theta_2,\chi_2) \otimes \Lambda(\theta_1,\chi_1)] \\ \times \rho [\Lambda(\theta_2,\chi_2) \otimes \Lambda(\theta_1,\chi_1)]^{\dagger} | H_{XX}H_X \rangle,$$

where ρ is the density matrix of the pair of photon in the basis $\{H_{XX}H_X, H_{XX}V_X, V_{XX}H_X, V_{XX}V_X\}$. By identifying $P_{+,+}^{\rho}(\theta_2, \chi_2, \theta_1, \chi_1) = P_{+,+}(\theta_2, \chi_2, \theta_1, \chi_1)$ for 16 well chosen sets of four angles $(\theta_2, \chi_2, \theta_1, \chi_1)$ we construct a linear system of 16 independent equations whose unknown variables are the 16 real values of ρ . In this way, we simply reconstruct the density matrix from the joint photodetection probabilities [35] and obtain a theoretical value of ρ . The same holds for an experimental approach. The calculated density matrix is hence

$$\underline{\rho} = \begin{pmatrix} \alpha & 0 & 0 & d - \iota c_1 \\ 0 & \beta & c_2 & 0 \\ 0 & c_2 & \beta & 0 \\ d + \iota c_1 & 0 & 0 & \alpha \end{pmatrix},$$
(19)

where

$$\alpha = \frac{1}{2} \frac{\gamma_1 + \Gamma_{flip}}{\gamma_1 + 2\Gamma_{flip}},$$

$$\beta = \frac{1}{2} \frac{\Gamma_{flip}}{\gamma_1 + 2\Gamma_{flip}},$$

$$d = \frac{1}{2} \frac{\gamma_1(\gamma_1 + 2\Gamma + \Gamma_{flip})}{(2\,\delta\omega)^2 + (\gamma_1 + \Gamma_{flip} + \Gamma)^2 - (\delta\Gamma_{flip})^2},$$

$$c_1 = \frac{1}{2} \frac{\gamma_1\delta\omega}{(2\,\delta\omega)^2 + (\gamma_1 + \Gamma_{flip} + \Gamma)^2 - (\delta\Gamma_{flip})^2},$$

$$c_2 = \frac{1}{2} \frac{\gamma_1\delta\Gamma_{flip}}{(2\,\delta\omega)^2 + (\gamma_1 + \Gamma_{flip} + \Gamma)^2 - (\delta\Gamma_{flip})^2}.$$
(20)

Note that for a perfect quantum dot $(\delta\omega \rightarrow 0, \Gamma_{flip} \rightarrow 0, \Gamma \rightarrow 0)$, ρ tends, as expected, toward the $|\Phi^+\rangle$ Bell state: $\rho \rightarrow |\phi^+\rangle\langle\phi^+|$.

C. Quantum dot spectroscopy from quantum tomography

An interesting feature of the analytical form of the density matrix arises from the fact that once computed experimentally, one can deduce all the quantum dot parameters provided γ_1 is measured independently. They are expressed as a function of the density matrix elements as follows:

$$\begin{split} \delta \omega &= \gamma_1 \frac{c_1}{4(d^2 + c_1^2 - c_2^2)}, \\ \delta \Gamma_{flip} &= \gamma_1 \frac{c_2}{2(d^2 + c_1^2 - c_2^2)}, \\ \Gamma_{flip} &= \gamma_1 \frac{2\beta}{4\beta - 1}, \\ \Gamma &= \gamma_1 \frac{d(1 - 2d - 4\beta + 4d\beta) + 2(c_1^2 - c_2^2)(2\beta - 1)}{4(d^2 + c_1^2 - c_2^2)(4\beta - 1)}. \end{split}$$
(21)

D. Entanglement witness

Apart from the CHSH inequality other entanglement witnesses can be constructed and following [31] we define an entanglement witness as $Tr[W\rho]$, where *W* is an operator. In the case where *W* is an optimal witness, the above mentioned quantity is negative if ρ is an entangled state. As proposed in [32] we define the partial transpose $\rho_0^{T_2}$ of an arbitrary density matrix ρ_0 as follows:

$$\rho_{0} = \sum_{i,j,k,l=H,V} \underline{\rho}_{0i,j,k,l} |ij\rangle \langle kl|,$$

$$\rho_{0}^{T_{2}} = \sum_{i,j,k,l=H,V} \underline{\rho}_{0k,j,i,l} |ij\rangle \langle kl|.$$
(22)

As demonstrated in [32], if ρ^{T_2} has a negative eigenvalue λ associated to the eigenvector $|\nu\rangle$ then the density matrix ρ represents an entangled state. Thus defining $W = |\nu\rangle\langle\nu|^{T_2}$ we have

$$\operatorname{Tr}[W\underline{\rho}_0] = \operatorname{Tr}[|\nu\rangle\langle\nu|\underline{\rho}_0^{T_2}] = \lambda < 0.$$
(23)

In our case we choose for ρ_0 the density matrix $|\phi^+\rangle\langle\phi^+|$ toward which the biexciton density matrix ρ of our model tends to. This gives a nonoptimal witness but is already less demanding than the Bell's inequalities and with a simple analytical form

$$W = \begin{pmatrix} 0 & 0 & 0 & -1/2 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 1/2 & 0 \\ -1/2 & 0 & 0 & 0 \end{pmatrix},$$
(24)

$$\operatorname{Tr}[W\underline{\rho}] = \beta - d$$

$$= \frac{1}{2} \frac{\Gamma_{flip}}{\gamma_1 + 2\Gamma_{flip}}$$

$$- \frac{1}{2} \frac{\gamma_1(\gamma_1 + 2\Gamma + \Gamma_{flip})}{(2\,\delta\omega)^2 + (\gamma_1 + \Gamma_{flip} + \Gamma)^2 - (\delta\Gamma_{flip})^2}.$$
(25)

IV. RESTORATION OF ENTANGLEMENT THROUGH CAVITY EFFECTS

In the above section we have analytically derived the CHSH inequality as well as the density matrix for the biexciton-exciton cascade emission from a single semiconductor quantum dot. Although we could discuss the density matrix as a function of the internal parameters of the guantum dot (QD), we choose to discuss the CHSH inequality since it is an intuitive entanglement witness with a simple experimental realization. Equation (18) indicates that polarization entanglement in the cascade emission from a biexciton in a self-assembled quantum dot may be affected by the relative contribution of three processes with respect to the exciton radiative lifetime $1/\gamma_1$: the mutual coherence between the two nondegenerate excitonic levels described by a cross-dephasing time $1/\Gamma$, the excitonic energy splitting giving rise to quantum beats with a time period $2\pi/2\delta\omega$ and the incoherent population exchange between the two bright excitons with a decay time $1/\Gamma_{flip}$. Entanglement does not depend on the biexciton radiative rate (γ_2) and among all the dephasing processes taken into account in our model, only the cross dephasing between the excitonic levels affects the visibility of entanglement. The analytical expression of S given by Eq. (18) also confirms that polarization entanglement from the biexciton cascade in self-assembled quantum dots is exclusively affected by the dynamics and mutual coherence of the excitonic states.

For quantum dots with no excitonic energy splitting ($\delta\omega$ =0) and in the absence of cross dephasing (Γ =0) and incoherent population exchange (Γ_{flip} =0), the *S* quantity reaches its maximum value of $2\sqrt{2}$ and the photons emerging from the biexcitonic cascade are maximally entangled [13]. Conversely, for quantum dots whose excitonic states are split, and which are affected by spin-dependent dephasing mechanisms and incoherent population exchange between the exciton bright states, the *S* parameter rapidly decreases so that the two photons emitted are only partially entangled or even only correlated in one preferred polarization basis corresponding to the polarization eigenbasis of the dot [19].

As an example, the characteristic excitonic lifetimes $1/\gamma_1$ of InAs quantum dots embedded in GaAs are typically on the order of 1 ns [36] and the excitonic energy splitting $2\hbar\delta\omega$ is of the order of a few μeV [37] corresponding to quantum beat periods lower than a few hundred picoseconds. Numerous observations also indicate that the exciton spin relaxation is quite negligible on the time scale of the exciton lifetime and may reach values of about 10 ns or even higher [38,39]. The mutual coherence time $1/\Gamma$ is likely to be longer than a few ns [40], since it shall involve hypothetical spindependent dephasing processes. These typical values indicate that the main ingredient affecting entanglement is the excitonic energy splitting; they imply that in an experimental setup involving bare InAs quantum dots, the S quantity is lower than 2 and tests of the Bell's inequalities on the two photons emerging from the biexciton cascade will not lead to any violation of the CHSH inequality (see the dashed line in Fig. 3). Even for relatively small exciton energy splitting $(2\hbar\delta\omega$ higher than a few μ eV), the S value tends to 1.2, a value significantly lower than the $S = \sqrt{2}$ limit of perfectly



FIG. 3. CHSH inequality as a function of the energy splitting of the exciton line, for a single quantum dot in bulk material (dashed line) and subject to a Purcell effect with F=10 (continuous line). The dotted line corresponds to the classical limit of 2. For these two curves, $T_1=1/\gamma_1=1$ ns, $1/\Gamma_{flip}=10$ ns, and $1/\Gamma=2$ ns.

correlated photons without any hidden variables. The incoherent population exchange between the excitonic relay levels destroys entanglement and the emitted photons are a statistical mixture of $\{|HH\rangle, |VV\rangle, |HV\rangle, |VH\rangle\}$ states. Even for bare quantum dots with no exciton energy splitting, entanglement is spoiled by cross dephasing and incoherent population exchange between the two bright excitonic states: the maximum value of *S* on Fig. 3 for such quantum dots reaches only 2.06, a value very close to the classical limit of 2.

Nevertheless, restoration of entanglement and improvement of its visibility can be achieved by reducing the excitonic radiative lifetime of the quantum dot by a factor of Fthrough its introduction in a resonant microcavity and the exploitation of the Purcell effect [41]: by making the excitonic spontaneous emission faster than the quantum beats period $(F\gamma_1 \ge 2\delta\omega)$, the cross-dephasing time $(F\gamma_1 \ge \Gamma)$ and the decay time of incoherent excitonic population exchange $(F\gamma_1 \gg \Gamma_{flin})$, it should be possible to preserve the quantum correlations between the two recombination paths. We consider here that both excitonic transitions releasing either Hor V-polarized photons are accelerated by cavity effects with the same spontaneous emission enhancement factor F. For dots subject to a spontaneous emission enhancement of its excitonic transition by a factor F=10 (see the solid line in Fig. 3), S values higher than 2.6 should be achievable for null exciton energy splitting $(2\hbar \delta \omega = 0)$. In such microcavity source however, violation of Bell's inequalities (S > 2) requires the use of quantum dots with an excitonic energy splitting smaller than 7 μ eV. With a Purcell effect of F = 30, S reaches the value of 2.76 close to its maximum value of $2\sqrt{2}$ for dots with no exciton energy splitting, and Bell's inequalities are violated for quantum dots displaying an energy splitting up to 20 μ eV (see Fig. 4).

Figure 4 shows the value of *S* as a function of $2\hbar \delta \omega$ and *F* for values of γ_1 , Γ , and Γ_{flip} considered above as typical of currently available quantum dots. The results confirm that the main ingredient degrading entanglement is the exciton fine structure. However, reducing the exciton energy splitting within the exciton linewidth is not experimentally sufficient



FIG. 4. CHSH inequality as a function of the energy splitting of the exciton line and its spontaneous emission exaltation *F*, for a single quantum dot with $T_1=1/\gamma_1=1$ ns in bulk material, $1/\Gamma_{flip}=10$ ns and $1/\Gamma=2$ ns.

and hardly allows for violation of Bell's inequalities. Violation of the CHSH inequalities requires a combination of cavity effects enhancing the excitons spontaneous emission and techniques leading to a reduction of the exciton energy splitting (such as growth optimization [23] or use of external magnetic [21] or electric [22] field). For typically available quantum dots, a Purcell factor of the order of 10 exalting equally both exciton transitions, would be sufficient for reaching values of S higher than the classical limit of 2. Yet, the generation of maximally entangled photons $(S=2\sqrt{2})$ with a single quantum dot is precluded by all decoherence mechanisms such as cross dephasing between the exciton states and incoherent population exchange between the two bright excitons. Maximally entangled states could, however, still be obtained out of nonmaximally entangled states by use of entanglement purification [42].

V. SUMMARY AND CONCLUSION

We have shown analytically that in the two-photon cascade from the biexciton in a single semiconductor quantum dot, solely the dynamics and coherence of the excitonic dipole governs the visibility of polarization entanglement. We have derived Bell inequalities under the CHSH form, as well as the density matrix of such a state. In bare quantum dots, polarization entanglement is spoiled not only by the energy splitting of the relay level but also by the incoherent population exchange and cross dephasing between the two bright relay states. The use of a microcavity can restore the generation of polarization-entangled photons from the quantum dot: The presence of the microcavity enhances the spontaneous emission rate of the excitonic transition, so that emission of the second photon arises before any quantum beat, cross dephasing, or incoherent population transfer between the excitonic radiative states. For experimentally accessible regime, violation of Bell's inequalities can be achieved with real quantum dots, provided they exhibit small excitonic energy splitting (lower than a few μeV) and a Purcell factor of the order of 10. Such Purcell factors and excitonic energy splitting have already been achieved, indicating that the possibility of realizing polarization-entangled photons with semiconductor quantum dots embedded in microcavities is totally accessible with available technology.

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- [1] A. K. Ekert, Phys. Rev. Lett. 67, 661 (1991).
- [2] D. Collins, N. Gisin, and H. de Riedmatten, J. Mod. Opt. 52, 735 (2005).
- [3] C. H. Bennett, G. Brassard, C. Crépeau, R. Jozsa, A. Peres, and W. K. Wootters, Phys. Rev. Lett. 70, 1895 (1993).
- [4] P. Shor, SIAM J. Comput. 26, 1484 (1997).
- [5] L. K. Grover, Phys. Rev. Lett. 79, 325 (1997).
- [6] R. Ursin, T. Jennewein, M. Aspelmeyer, R. Kaltenbaek, M. Lindenthal, and A. Zeilinger, Nature (London) 430, 849 (2004).
- [7] O. Landry, J. A. W. van Houwelingen, A. Beveratos, H. Zbinden, and N. Gisin, J. Opt. Soc. Am. B **24**, 398 (2007).
- [8] J.-W. Pan, D. Bouwmeester, and H. Weinfurter, and A. Zeilinger, Phys. Rev. Lett. 80, 3891 (1998).
- [9] M. Halder, A. Beveratos, N. Gisin, V. Scarani, C. Simon, and H. Zbinden, Nat. Phys. 3, 692 (2007).
- [10] X. Li, P. L. Voss, J. E. Sharping, and P. Kumar, Phys. Rev. Lett. 94, 053601 (2005).
- [11] J. Fulconis, O. Alibart, J. L. O'Brien, W. J. Wadsworth, and J.

G. Rarity, Phys. Rev. Lett. 99, 120501 (2007).

- [12] V. Scarani, H. de Riedmatten, I. Marcikic, H. Zbinden, and N. Gisin, Eur. Phys. J. D 32, 129 (2005).
- [13] O. Benson, C. Santori, M. Pelton, and Y. Yamamoto, Phys. Rev. Lett. 84, 2513 (2000).
- [14] E. Moreau, I. Robert, L. Manin, V. Thierry-Mieg, J. M. Gérard, and I. Abram, Phys. Rev. Lett. 87, 183601 (2001).
- [15] C. Simon and J. P. Poizat, Phys. Rev. Lett. 94, 030502 (2005).
- [16] M. Larqué, A. Beveratos, and I. Robert-Philip, Eur. Phys. J. D 47, 119 (2008).
- [17] D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. **76**, 3005 (1996).
- [18] M. Bayer, G. Ortner, O. Stern, A. Kuther, A. A. Gorbunov, A. Forchel, P. Hawrylak, S. Fafard, K. Hinzer, T. L. Reinecke, S. N. Walck, J. P. Reithmaier, F. Klopf, and F. Schäfer, Phys. Rev. B 65, 195315 (2002).
- [19] C. Santori, D. Fattal, M. Pelton, G. S. Solomon, and Y. Yamamoto, Phys. Rev. B 66, 045308 (2002).
- [20] N. Akopian, N. H. Lindner, E. Poem, Y. Berlatzky, J. Avron,

D. Gershoni, B. D. Gerardot, and P. M. Petroff, Phys. Rev. Lett. 96, 130501 (2006).

- [21] R. M. Stevenson, R. J. Young, P. Atkinson, K. Cooper, D. A. Ritchie, and A. J. Shields, Nature (London) 439, 179 (2006).
- [22] B. D. Gerardot, S. Seidl, P. A. Dalgarno, R. J. Warburton, D. Granados, J. M. Garcia, K. Kowalik, and O. Krebs, Appl. Phys. Lett. **90**, 041101 (2007).
- [23] R. Seguin, A. Schliwa, S. Rodt, K. Pötschke, U. W. Pohl, and D. Bimberg, Phys. Rev. Lett. **95**, 257402 (2005).
- [24] A. Berthelot, I. Favero, G. Cassabois, C. Voisin, C. Delalande, Ph. Roussignol, R. Ferreira, and J. M. Gérard, Nat. Phys. 2, 759 (2006).
- [25] J. M. Gérard, B. Sermage, B. Gayral, B. Legrand, E. Costard, and V. Thierry-Mieg, Phys. Rev. Lett. 81, 1110 (1998).
- [26] A. Aspect, Ph. Grangier, and G. Roger, Phys. Rev. Lett. 49, 91 (1982).
- [27] E. S. Fry, Phys. Rev. A 8, 1219 (1973).
- [28] J. Bylander, I. Robert-Philip, and I. Abram, Eur. Phys. J. D 22, 295 (2003).
- [29] G. Lindblad, Commun. Math. Phys. 48, 119 (1976).
- [30] I. Marcikic, H. de Riedmatten, W. Tittel, H. Zbinden, M. Legré, and N. Gisin, Phys. Rev. Lett. 93, 180502 (2004).
- [31] P. Hyllus, O. Gühne, D. Bruss, and M. Lewenstein, Phys. Rev.

A 72, 012321 (2005).

- [32] A. Peres, Phys. Rev. Lett. 77, 1413 (1996).
- [33] D. Bohm, Phys. Rev. 85, 166 (1952).
- [34] J. F. Clauser, M. H. Horne, A. Shimony, and R. A. Holt, Phys. Rev. Lett. 23, 880 (1969).
- [35] D. F. V. James, P. G. Kwiat, W. J. Munro, and A. G. White, Phys. Rev. A 64, 052312 (2001).
- [36] J. M. Gérard, O. Cabrol, and B. Sermage, Appl. Phys. Lett. 68, 3123 (1996).
- [37] W. Langbein, P. Borri, U. Woggon, V. Stavarache, D. Reuter, and A. D. Wieck, Phys. Rev. B 69, 161301(R) (2004).
- [38] M. Paillard, X. Marie, P. Renucci, T. Amand, A. Jbeli, and J. M. Gérard, Phys. Rev. Lett. 86, 1634 (2001).
- [39] K. Kowalik, O. Krebs, A. Lemaître, J. A. Gaj, and P. Voisin, e-print arXiv:0801.0571.
- [40] A. J. Hudson, R. M. Stevenson, A. J. Bennett, R. J. Young, C. A. Nicoll, P. Atkinson, K. Cooper, D. A. Ritchie, and A. J. Shields, Phys. Rev. Lett. **99**, 266802 (2007).
- [41] F. Troiani, J. I. Perea, and C. Tejedor, Phys. Rev. B 74, 235310 (2006).
- [42] J. W. Pan, S. Gasparoni, R. Ursin, G. Weihs, and A. Zeilinger, Nature (London) 423, 417 (2003).