Entanglement of photons due to nonlinear optical response of quantum wells

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We study photons entangling in the short-time response of a quantum well excited by a short intense pulse. At the time scales, where the biexciton effect is not yet pronounced, the Pauli principle is responsible for many-body correlations among excitons, giving rise to the production of entangled photons with a yield $\sim 10^{-2}$. The quantum-field theoretical two-particle density matrix in second quantization is used to calculate the entanglement for arbitrary emission angles of the entangled pairs of photons. At the time scales, where the heavy-light hole splitting is resolved, the resonances corresponding to different two-exciton states develop, which allow for a simple kinematic theory relating the states of the outgoing photons with the respective two-exciton states. The resonant response can be expected at symmetric emission angles for resonances with the heavy-heavy and light-light two-exciton states with remarkably nontrivial dependence of entanglement on the emission angles and on the ellipticity parameters of the excitation. We show that the emitted entangled two-photon states are always in a triplet state.

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I. INTRODUCTION

Current entangled-photon sources are mainly based on the parametric down conversion inside nonlinear crystals,[1,2] such as β-barium borate crystals[3,4] or lithium niobate structures.[5] These sources suffer from two serious limitations. First, since the far off-resonant three-photon scattering contains two far off-resonant virtual states, the entangled-photon production yield is very low,[6,7] which limits the brightness of entangled-photon sources based on nonlinear crystals, leading to low signal-to-noise ratios and long measurement times.[5] Second, parametric down conversion produces entangled photons with a wavelength that is twice as long as the pump photons, which limits the operating wavelength.[6] Therefore, there is the permanent interest in alternative semiconductor structures to produce entangled photons. Quantum-dot (QD) structures have already been used to produce successfully entangled photons,[8–10] which make use of the relaxation of two excitons into one bound biexciton on a QD. QD structures are very attractive due to the possibility to achieve a sub-Poissonian temporal correlation $g^{(2)}(\tau)$ between two pairs of emitted entangled photons.[11] Depending on the application, such as transmission over low-noise optical fibers or over noisy atmospheric channels, QD or quantum well (QW) structures are preferred, respectively. For example, a quantum communication link over satellites could be set up in the visible region,[12] for which QW structures seem to be ideal due to their potentially very high entangled-photon production yield.

A current limitation of QD structures is the low operating temperature regime, which is mainly due to the decoherence arising from exciton-phonon and hyperfine interactions.[13–16] QW structures face the additional decoherence source due to Coulomb interactions, even at low temperatures. We therefore investigate the possibility to use the short-time response for the production of entangled photons at time scales for which the biexciton binding energy E_{xx} cannot be resolved anymore, i.e., $T \ll \hbar / E_{xx}$ according to the Heisenberg uncertainty principle. In order for this method to be effective, a microcavity would make a favorable effect for quicker extraction of entangled photons. In this paper, we are concerned with the possibility to produce entangled photons without the need of Coulomb interaction. As we show, it turns out that the Pauli exclusion principle, i.e., quantum statistics, which is instantaneous, is sufficient to produce entangled photons from a QW structure with a high yield. This channel of entangling photons is different from the well-known method that is based on the bound biexciton state.[8,9,11,17–20] The similar mechanism was considered recently in Ref. 21 in the context of the exciton polaritons (that is, in the relatively long-time limit). In the present paper, we focus primarily on a single QW, when the photon density of states is not affected by the presence of the cavity. In this case, we can use perturbation theory to describe the two-photon scattering. Thus the calculations presented in this paper provide the basic insight into the entanglement production mechanism due to the Pauli principle.

II. TWO-PHOTON DENSITY MATRIX

The excitation of the semiconductor QW in a cavity by the external field and emission of the photons due to the radiative recombination are driven by the interaction of the QW with the photonic modes of the cavity. In order to provide the consistent description of these processes, one needs to take into account both the photonic modes propagating outside the structure and the modification of the photonic density of states by the cavity. This is achieved by quantizing electromagnetic field in the whole space while taking into account the one-dimensional (1D) spatial modulation of the refractive index $n(z)$ with z axis coinciding with the growth direction. Let the left (right) end of the cavity be situated at $z=z_L$ (respectively, $z=z_R$) so that $n(z)=n_0$ as $z<z_L$ and $z>z_R$. Outside the structure, the electromagnetic field obeys the Maxwell equations for the empty space. The translational invariance in the direction perpendicular to the growth direction leads to the conservation of the in-plane component of the wave vector $\mathbf{k}_\parallel$. Thus the state of the field can be char-

acterized by $\mathbf{k}_\parallel$, the frequency ω, and the polarization state $\boldsymbol{\epsilon}_h$ with $h=p,s$. The unit vectors $\boldsymbol{\epsilon}_p$ and $\boldsymbol{\epsilon}_s$ correspond to p and s polarizations, i.e., to the polarizations in the plane spanned by $\boldsymbol{\epsilon}_z$ and $\mathbf{k}_\parallel$ and perpendicular to this plane, respectively.

For propagating modes, one still needs to fix the sign of the z component of the wave vector, $k_z=\sqrt{\omega^2/c^2-k_\parallel^2}$. We do this by using the solutions of the standard 1D scattering problems, that is $u_+(z)\propto\exp(ik_z z)$ as $z>z_R$ and $u_-(z)\propto\exp(-ik_z z)$ as $z<z_L$. We combine all the "quantum numbers" characterizing the state of the electromagnetic field into the single index $\hat{k}=(\mathbf{k}_\parallel,\omega,h,g)$, where $g=\pm$ enumerates the solutions of the scattering problem. Using these notations, we present the quantized field as (see, e.g., Ref. 22) in the units with $\hbar=1$, $c=1$,

$$\mathbf{A}=\frac{1}{(2\pi)^{3/2}}\sum_{\hat{k}}\boldsymbol{\epsilon}_{\hat{k}}\frac{1}{\sqrt{2\omega_{\hat{k}}}}u_{\hat{k}}(z)e^{i\mathbf{k}_\parallel\boldsymbol{\rho}}a_{\hat{k}}^\dagger+\text{H.c.}, \tag{1}$$

where $u_{\hat{k}}(z)$ is the spatial distribution of the field along the z axis found as the solution of the respective 1D problem, $\boldsymbol{\rho}$ is the coordinate in the plane of the cavity, and $a_{\hat{k}}^\dagger$ is the respective photon creation operator. The summation over $\hat{k}$ implies the integration over the continuous quantum numbers and summation over the discrete ones.

The entanglement of the photons produced in the course of the radiative relaxation of the pumped semiconductor is found considering the two-photon density matrix. Assuming that the system is in the coherent regime, the density matrix is written as

$$\rho_{\hat{q}_1,\hat{q}_2}^{\hat{k}_1,\hat{k}_2}(t)=\langle\Psi(t)|a_{\hat{q}_1}^\dagger a_{\hat{q}_2}^\dagger a_{\hat{k}_1}a_{\hat{k}_2}|\Psi(t)\rangle, \tag{2}$$

where $|\Psi(t)\rangle$ is the state of the semiconductor-photon system. This state is naturally represented as a superposition of the states corresponding to different numbers of photons. Such representation is in the direct relation with the perturbation theory with respect to the external field. We consider the lowest nonvanishing terms of the perturbational series. This approximation is analogous to the $\chi^{(3)}$ approximation, which is valid if the excitation is not too strong, for example, if the excitation pulse area is much smaller than π. In this approximation, the nonzero contribution to the density matrix results from the two-photon states entering $|\Psi(t)\rangle$. These states are mapped to vacuum by the operator $a_{\hat{k}_1}a_{\hat{k}_2}$. Thus one has

$$\rho_{\hat{q}_1,\hat{q}_2}^{\hat{k}_1,\hat{k}_2}(t)=\Psi_{\hat{q}_1,\hat{q}_2}^*(t)\Psi_{\hat{k}_1,\hat{k}_2}(t) \tag{3}$$

with $\Psi_{\hat{k}_1,\hat{k}_2}(t)=\langle 0|a_{\hat{k}_1}a_{\hat{k}_2}|\Psi(t)\rangle$, where $|0\rangle$ is vacuum of the combined system, that is, there are no photons and the semiconductor has full valence band and empty conduction band. In order to account for free dynamics of the field and the semiconductor, we switch to the interaction picture,

$$\Psi_{\hat{k}_1,\hat{k}_2}(t)=\langle 0|a_{\hat{k}_1}(t)a_{\hat{k}_2}(t)\mathcal{T}_+\exp\left\{-i\int_0^t dt'\tilde{H}_{int}(t')\right\}|0\rangle, \tag{4}$$

where $a_{\hat{k}}(t)$ is the photon annihilation operator in the interaction representation, $\tilde{H}_{int}(t)$ is the Hamiltonian of the light-matter interaction in the interaction representation, and $\mathcal{T}_+$ is the time-ordering operator. In order to derive $H_{int}(t)$, we start from the interaction Hamiltonian written in the electron representation,

$$H_{int}=-\sum_{n,n'}\int d\mathbf{x}c_n^\dagger(\mathbf{x})\mathbf{A}(\mathbf{x})\cdot\mathbf{d}_{nn'}c_{n'}(\mathbf{x}), \tag{5}$$

where $c_n^\dagger(\mathbf{x})$ creates electron in band n and $\mathbf{d}_{nn'}=e/m\langle n|\mathbf{p}|n'\rangle$ is proportional to the matrix element of the momentum operator between the bands n and n'. For the electron states in the valence bands, we introduce the hole operators in the usual way as $v_n^\dagger(\mathbf{x})=c_n(\mathbf{x})$ and enumerate different states in the conduction and valence bands by their spins. It gives for Eq. (5),

$$H_{int}=\sum_{s,\sigma}\int d\mathbf{x}[c_s(\mathbf{x})\mathbf{A}(\mathbf{x})\cdot\mathbf{d}_{\sigma,s}v_\sigma(\mathbf{x}) + v_\sigma^\dagger(\mathbf{x})\mathbf{A}(\mathbf{x})\cdot\mathbf{d}_{s,\sigma}c_s^\dagger(\mathbf{x})]. \tag{6}$$

In order to account for the genuine low-energy excitations, it is convenient to introduce the exciton operators according to $|\mu\rangle=B_\mu^\dagger|0\rangle$, where $|\mu\rangle$ is the hole-electron pair state either bound or unbound corresponding to energy ϵ_μ, i.e., $H_{SC}|\mu\rangle=E_\mu|\mu\rangle$ with H_{SC} being the Hamiltonian of the nonperturbed semiconductor. For simplicity, we assume that the QW can be approximated by a two-dimensional (2D) plane. In this case, the exciton states are characterized by the spin states of the hole and the electron constituting the pair, the center of mass momentum in the plane of the well, $\mathbf{K}$, and other quantum numbers, n_μ, so that $|\mu\rangle=|\sigma_\mu,s_\mu,\mathbf{K}_\mu,n_\mu\rangle$. Denoting by $\phi_\mu(\mathbf{x},\mathbf{x}')$ the exciton (hole-electron) wave function corresponding to the state μ, we represent the exciton operator as

$$B_\mu=\int d\mathbf{x}d\mathbf{x}'\phi_\mu^*(\mathbf{x},\mathbf{x}')c_{s_\mu}(\mathbf{x}')v_{\sigma_\mu}(\mathbf{x}). \tag{7}$$

Using completeness of the exciton wave functions, Eq. (6) can be expressed in terms of the exciton operators,

$$H_{int}=\sum_\mu(\bar{\mathcal{A}}_\mu B_\mu+\mathcal{A}_\mu B_\mu^\dagger), \tag{8}$$

where

$$\bar{\mathcal{A}}_\mu=\int d\mathbf{x}\mathbf{A}(\mathbf{x})\cdot\mathbf{d}_{\sigma_\mu,s_\mu}\phi_\mu(\mathbf{x},\mathbf{x}),$$

$$\mathcal{A}_\mu=\int d\mathbf{x}\mathbf{A}(\mathbf{x})\cdot\mathbf{d}_{s_\mu,\sigma_\mu}\phi_\mu^*(\mathbf{x},\mathbf{x}). \tag{9}$$

We represent the exciton wave function as $\phi_\mu(\mathbf{x},\mathbf{x}')=e^{i\mathbf{K}_\mu\cdot\mathbf{R}_\mu}\tilde{\phi}_\mu(\mathbf{x}-\mathbf{x}')$, where $\mathbf{R}_\mu$ is the in-plane coordinate of

the exciton center of mass and $\tilde{\phi}_\mu(\mathbf{x}-\mathbf{x}')$ is the wave function in the relative coordinates. Using this representation, one can see that the external field is coupled to the states with zero orbital momentum and that only the component of the external field corresponding to $\mathbf{K}_\mu$ contributes into $\mathcal{A}_\mu$ and $\overline{\mathcal{A}}_\mu$.

In order to distinguish between the processes of excitation and radiative exciton recombination, we separate the contributions of the quantized field of emitted photons, $\mathbf{A}^{(q)}(\mathbf{x})$, and of the classical pumping field, $\mathbf{A}^{(cl)}(\mathbf{x})$, into the external field,

$$\mathbf{A}(\mathbf{x}) = \mathbf{A}^{(q)}(\mathbf{x}) + \mathbf{A}^{(cl)}(\mathbf{x}). \tag{10}$$

Both $\mathbf{A}^{(q)}(\mathbf{x})$ and $\mathbf{A}^{(cl)}(\mathbf{x})$ can be expanded over the modes as in Eq. (1) with the only difference that in the expansion of $\mathbf{A}^{(cl)}(\mathbf{x})$, one should use c-number amplitudes instead of the photonic creation and annihilation operators. To simplify the notation in what follows, we will omit the upper index for the quantized field.

In the lowest order of the perturbation theory, we can neglect the processes of reemission and reabsorption. This together with adopting the rotating-frame approximation means that in Eq. (8), the pump field should be paired with the exciton creation operators while the quantized field is paired with the exciton annihilation process. In the latter case, in turn, we need to leave only the part responsible for creation photons, i.e., the part explicitly shown in Eq. (1). Thus, separating the classical and quantized contributions into the electromagnetic field, we use in Eq. (4) the effective interaction Hamiltonian in the form

$$H_{int} = \sum_\mu (\mathcal{A}_\mu^\dagger B_\mu + \mathcal{A}_\mu^{(cl)} B_\mu^\dagger), \tag{11}$$

where $\mathcal{A}_\mu^\dagger$ is found by using the part shown in Eq. (1) in the first equation in Eq. (9) that is,

$$\mathcal{A}_\mu^\dagger = \frac{1}{(2\pi)^{3/2}} \sum_{\hat{k}} \frac{1}{\sqrt{2\omega_{\hat{k}}}} \mathbf{d}_{\sigma_\mu, s_\mu} \cdot \boldsymbol{\epsilon}_{\hat{k}} a_{\hat{k}}^\dagger \int d\mathbf{x} \phi_\mu(\mathbf{x},\mathbf{x}) u_{\hat{k}}(z) e^{i\mathbf{k}_\parallel \cdot \mathbf{x}} \tag{12}$$

and $\mathcal{A}_\mu^{(cl)}(t)$ is found taking into account only the external excitation field in Eq. (9). It should be noted that in this approximation, we neglect the effect of the interaction of light with the QW on the form of the photonic modes in the expression for the density matrix. As a result, the spatial distribution of the exciting field within the cavity is found solving the respective initial value problem for the cavity alone as if the QW were absent.

Expanding the exponential term in Eq. (4), we obtain various terms depending on ordering of the operators B and $B^\dagger$. In what follows, we will be mostly interested in the response along the directions different from the direction of the incident excitation field. Along these directions, the response is not blurred by the nonscattered field and by the linear (single-photon) response. For accordingly chosen $\hat{k}_1$ and $\hat{k}_2$, we obtain for the two-photon amplitudes

$$\begin{aligned}\Psi_{\hat{k}_1,\hat{k}_2}(t) = -\int dt_1 \ldots dt_4 \mathcal{A}_{\nu_3}^{(cl)}(t_3)\mathcal{A}_{\nu_4}^{(cl)}(t_4) \\ \times \langle B_{\mu_1}(t_1) B_{\mu_2}(t_2) B_{\nu_3}^\dagger(t_3) B_{\nu_4}^\dagger(t_4)\rangle \\ \times \langle a_{\hat{k}_1}(t) a_{\hat{k}_2}(t) \mathcal{A}_{\mu_1}^\dagger(t_1) \mathcal{A}_{\mu_2}^\dagger(t_2)\rangle,\end{aligned} \tag{13}$$

where the time intervals under integrations are arranged according to $0 \le t_1 \le \ldots \le t_4 \le t$. Here and below the summation over all hole-electron pair states $\mu_{1,2}$ and $\nu_{3,4}$ is implied.

In order to simplify the integrals in Eqs. (9) and (12), we assume that the QW situated at $z=z_0$ can be described using the δ-functional approximation. Using this assumption, the two-photon amplitude can be presented in the form emphasizing its dependence on the polarization states of the outgoing photons,

$$\Psi_{\hat{k}_1,\hat{k}_2}(t) = -iu_{\hat{k}_1}(z_0) u_{\hat{k}_2}(z_0) \boldsymbol{\epsilon}_{\hat{k}_1} \cdot \overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t) \cdot \boldsymbol{\epsilon}_{\hat{k}_2}, \tag{14}$$

where

$$\begin{aligned}\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t) = \frac{\pi}{\sqrt{\omega_{k_1}\omega_{k_2}}} \tilde{\phi}_{\mu_1}(0)\tilde{\phi}_{\mu_2}(0) \\ \times \int_0^t dt_1 e^{-i(\omega_{k_1}-\omega_{k_2})(t-t_1)} \int_0^{t_1} dt_2 e^{-iE_{\mu_1}(t_1-t_2)} \\ \times [e^{-i\omega_{k_1}(t_1-t_2)} + e^{-i\omega_{k_2}(t_1-t_2)}] \\ \times \delta(\mathbf{K}_{\mu_1} - \mathbf{k}_{1,\parallel}) \delta(\mathbf{K}_{\mu_2} - \mathbf{k}_{2,\parallel}) \\ \times \mathbf{d}_{\sigma_{\mu_1}, s_{\mu_1}} \otimes \mathbf{d}_{\sigma_{\mu_2}, s_{\mu_2}} \mathcal{G}_{\mu_1,\mu_2}(t_2).\end{aligned} \tag{15}$$

Here $\otimes$ denotes the tensor product so that in a particular Cartesian coordinate system $(\mathbf{d}\otimes\mathbf{d}')_{ij}=d_i d_j'$, $\omega_k=k$ is the photon energy and E_μ are the energies of the hole-electron pair states.

The tensor $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ depends only on the direction of propagation of the outgoing photons but neither on their polarizations nor on the choice of the solutions of the scattering problems. This information is contained in the amplitudes $u_{\hat{k}_{1,2}}(z_0)$. They also describe the effect of the photonic density of states modified by the cavity. For example, if the field distribution inside the cavity has the maximum near z_0, this results in accordingly amplified two-particle amplitudes $\Psi_{\hat{k}_1,\hat{k}_2}$. On the contrary, if, for example, for one polarization, s or p, the amplitude is significantly smaller comparing to the other, this can be easily shown to imply significant decrease in the photon entanglement and so on.

The effect of the semiconductor nonlinear response is described by the function $\mathcal{G}_{\mu_1,\mu_2}(t)$, which we present as a sum of the instantaneous and the memory terms to make the analogy with the representation of semiconductor nonlinear response in the $\chi^{(3)}$ approximation[23,24] clearer,

$$\begin{aligned}\mathcal{G}_{\mu_1,\mu_2}(t) = \langle B_{\mu_1} B_{\mu_2} B_{\nu_3}^\dagger B_{\nu_4}^\dagger\rangle P_{\nu_3}^{(1)}(t) P_{\nu_4}^{(1)}(t) \\ + i\int_0^t dt' \langle B_{\mu_1} B_{\mu_2} e^{-iH_{SC}(t-t')} D_{\nu_3,\nu_4}^\dagger\rangle \\ \times P_{\nu_3}^{(1)}(t') P_{\nu_4}^{(1)}(t'),\end{aligned} \tag{16}$$

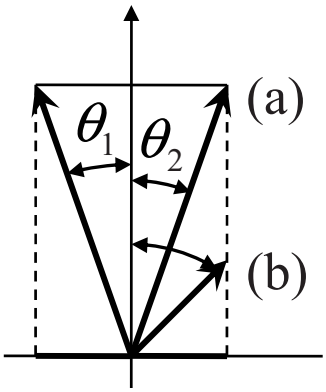

FIG. 1. The wave vectors of the outgoing photons. (a) If the energies of the photons are equal, the resonant directions are symmetric. (b) The angles of propagation are different if the photon energies are not the same.

where[23,24] $D^{\dagger}_{\nu_3,\nu_4}=[B^{\dagger}_{\nu_3},[B^{\dagger}_{\nu_4},H]]$ and $P^{(1)}_{\nu}(t)$ are the exciton polarizations of the linear response created by the action of the external (classical) field,

$$P^{(1)}_{\nu}(t) = -i\int_0^t dt' e^{-iE_\nu(t-t')}\mathcal{A}^{(cl)}_{\nu}(t'). \tag{17}$$

The two-photon amplitudes while are given by the series of long expressions have the transparent structure. Indeed, the function $\mathcal{G}_{\mu_1,\mu_2}(t)$ is related to the amplitude of finding the semiconductor excited through the two-photon absorption in the "two-exciton" state $|\mu_1,\mu_2\rangle \equiv B^{\dagger}_{\mu_1}B^{\dagger}_{\mu_2}|0\rangle$. The kernel $\propto t_1-t_2$ in the integral over t_2 in Eq. (15) has the meaning of the propagator of a photon-exciton system resulted from the radiative recombination of one of the excitons. Finally, the kernel in the integral over t_1 is the propagator of the two-photon state.

The important property of the two-photon amplitudes follows from the fact that the interaction between the electrons and the holes leaves the total momentum intact. That is, particular terms in the sum over ν_3 and ν_4 in Eq. (16) are proportional to $\delta(\mathbf{K}_{\mu_1}+\mathbf{K}_{\mu_2}-\mathbf{K}_{\nu_3}-\mathbf{K}_{\nu_4})$. Assuming the normal incidence of the excitation field, this leads to important restriction on the two-photon amplitudes,

$$\Psi_{\hat{k}_1,\hat{k}_2} \propto \delta(\mathbf{k}_{1,\parallel}+\mathbf{k}_{2,\parallel}). \tag{18}$$

It should be emphasized that since the QW is invariant only with respect to 2D translations, Eq. (18) implies restrictions only for the in-plane projections of the wave vectors of the outgoing photons (see Fig. 1).

III. ENTANGLEMENT OF EMITTED PHOTONS

The application of the perturbation theory for finding the density matrix has the great advantage that, as follows from Eq. (3), the density matrix corresponds to a pure state of the two-photon system. Thus, we can directly apply the standard machinery for evaluating entanglement of two photons as the von Neumann entropy of the reduced density matrix. Next orders of the perturbation theory can be shown to lead to the two-photon density matrix which cannot be presented as a direct product of the two-photon amplitudes. The origin of the mixed states is clear. Next orders take into account not only the pairs of emitted photons but also triples, quadruplets, and so on. The two-photon density matrix is effectively obtained by tracing out the states corresponding to "extra" photon, which, due to entanglement, results in a density matrix corresponding to a mixture.

The dependence of entanglement on the direction of propagation of the outgoing photons is very complex. With this regard, it should be noted that $\Psi_{\hat{k}_1,\hat{k}_2}(t)$ is *not* a bilinear function of two vectors, that is strictly speaking $T_{\hat{k}_1,\hat{k}_2}=u_{\hat{k}_1}\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)u_{\hat{k}_2}$ is not a tensor. Due to the presence of the cavity, the amplitudes $u_{\hat{k}}(z_0)$, which depend nontrivially on the polarization of the field, both the formal eigenvalues and the principal axes of $T_{\hat{k}_1,\hat{k}_2}$ depend on $\boldsymbol{\epsilon}_{\hat{k}_1}$ and $\boldsymbol{\epsilon}_{\hat{k}_2}$. In particular, it is imperative that $\boldsymbol{\epsilon}_{\hat{k}_1}$ and $\boldsymbol{\epsilon}_{\hat{k}_2}$ in Eq. (14) are the unit vectors of s and p polarizations corresponding to the directions $\hat{\mathbf{e}}_1$ and $\hat{\mathbf{e}}_2$, respectively.

In the present paper, we concentrate on the principal features of entanglement of emitted photons. We adopt some simplifications and assumptions which allow us to maximally reduce the technical complications. As the first simplification, we limit ourselves to the case of a single QW without the cavity. In this case, $u_{\hat{k}}(z_0)$ are polarization independent plane waves and up to the choice of the start point of the time axis, we can take $u_{\hat{k}}(z_0)\equiv 1$. This approximation removes the dependence of $T_{\hat{k}_1,\hat{k}_2}$ on the polarization vectors and makes it a legal tensor thus yielding a compact expression for the reduced single-photon density matrix,

$$\rho^{\boldsymbol{\epsilon}'}_{\boldsymbol{\epsilon}}(t;\mathbf{k}_1,\mathbf{k}_2) = \boldsymbol{\epsilon}\cdot\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)(\overleftrightarrow{1}-\hat{\mathbf{e}}_2\otimes\hat{\mathbf{e}}_2)\overleftrightarrow{M}^{\dagger}_{\mathbf{k}_1,\mathbf{k}_2}(t)\cdot\boldsymbol{\epsilon}', \tag{19}$$

where $\overleftrightarrow{1}$ is the unit tensor and the argument of $\rho^{\boldsymbol{\epsilon}'}_{\boldsymbol{\epsilon}}$ shows the dependence of the reduced density matrix on the wave vectors of the *pair* of the outgoing photons. It is seen immediately from this representation that the tensor convoluted with the polarization vectors has at least one zero eigenvalue. Its two remaining eigenvalues determine entanglement as $E_N=-\tilde{\rho}_1\log_2(\tilde{\rho}_1)-\tilde{\rho}_2\log_2(\tilde{\rho}_2)$ with $\tilde{\rho}_{1,2}=\rho_{1,2}/(\rho_1+\rho_2)$.

Two contributions into $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ stemming from the instantaneous and the memory terms in Eq. (16) describe different types of semiconductor dynamics prior to the photons emission. For oblique directions, distinct from the direction of the excitation field, the instantaneous term describes the effect of the Pauli exclusion principle. The memory term accounts for the Coulomb interaction. The nontrivial part of the memory term is related to resonances due to existence of the bound (biexcitons) and "almost bound" states of two holes and two electrons.[20,23,25,26] Often this is the Coulomb interaction, which is regarded to be the main source of entanglement. This is motivated by the standard physical picture that for a pair of *initially disentangled* particles in order to become entangled they must interact. For optically excited semiconductors, however, this picture is not sufficient because the process of excitation of the semiconductor is affected by already existent population of the conduction and

the valence bands owing to the Pauli principle. This is particularly relevant for the problem of entanglement because the effect of the exclusion principle is determined by the electron and hole spin states, which play the direct role in determining the polarization states of emitted photons. In the present paper, we limit ourselves to considering only the effect of the exclusion principle, which allows for complete and closed description. The interplay between the effects of Pauli blocking and of Coulomb interaction will be studied elsewhere (see, however, Appendix for more detailed discussion). To this end, we leave only the instantaneous term in the expression for $\mathcal{G}_{\mu_1,\mu_2}$. Also among the different hole-electron pair states, we take into account only the most prominent heavy-hole and light-hole $1s$ exciton states in the sum over μ_1 and μ_2.

The polarization state of the excitation pulse is specified by the amplitudes of its left circular and right circular components A_+ and A_-, respectively. Using the simplifications described above, the tensor $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ is directly expressed in terms of these amplitudes as

$$\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t) = \frac{(2\pi)^5}{4}\frac{1}{\sqrt{\omega_1\omega_2}}\delta(\mathbf{k}_{1,\parallel}+\mathbf{k}_{2,\parallel}) \times \int_0^t dt_1 e^{-i(\omega_1+\omega_2)(t-t_1)} \times \int_0^{t_1} dt_2 (e^{-i\omega_1(t_1-t_2)}+e^{-i\omega_2(t_1-t_2)})\overleftrightarrow{W}(t_1,t_2), \tag{20}$$

where

$$\overleftrightarrow{W}(t_1,t_2) = (A_+^2\hat{\mathbf{e}}_+\otimes\hat{\mathbf{e}}_+ + A_-^2\hat{\mathbf{e}}_-\otimes\hat{\mathbf{e}}_-)W_1(t_1,t_2) + (\hat{\mathbf{e}}_+\otimes\hat{\mathbf{e}}_- + \hat{\mathbf{e}}_-\otimes\hat{\mathbf{e}}_+)A_+A_-W_2(t_1,t_2) + \hat{\mathbf{e}}_z\otimes\hat{\mathbf{e}}_z A_+A_-W_3(t_1,t_2) \tag{21}$$

with

$$W_1(t_1,t_2) = e^{-iE_h(t_1-t_2)}\mathcal{I}_{hh}e^{-2iE_ht_2} + \frac{1}{9}e^{-iE_l(t_1-t_2)}\mathcal{I}_{ll}e^{-2iE_lt_2},$$

$$W_2(t_1,t_2) = \frac{2}{3}(e^{-iE_h(t_1-t_2)}+e^{-iE_l(t_1-t_2)})\mathcal{I}_{hl}e^{-i(E_h+E_l)t_2},$$

$$W_3(t_1,t_2) = -\frac{8}{9}e^{-iE_l(t_1-t_2)}\mathcal{I}_{ll}e^{-2iE_lt_2}. \tag{22}$$

Here we have taken into account that the exciton energies and their wave functions do not depend on the sign of the spin and have introduced the indices h and l for $|\sigma|=3/2$ and $|\sigma|=1/2$, respectively. The nontrivial part of the contribution of the instantaneous term in Eq. (16) is given by

$$\mathcal{I}_{\sigma_1,\sigma_2} = |Q|^4|\tilde{\phi}_{\sigma_1}(0)\tilde{\phi}_{\sigma_2}(0)|^2\int d\mathbf{q}|\tilde{\varphi}_{\sigma_1}(\mathbf{q})|^2|\tilde{\varphi}_{\sigma_2}(\mathbf{q})|^2, \tag{23}$$

where we have neglected the dependence on the in-plane momentum using the fact that it is much smaller than the inverse exciton Bohr radius and have introduced $\tilde{\varphi}_\sigma(\mathbf{q})$, the Fourier transform of the exciton wave function. Writing down Eq. (22), we have taken into account the structure of the valence band in the semiconductors with the point symmetry T_d and have introduced the common interband dipole moment $Q=-ie/m\langle X|p_x|\Gamma_1\rangle$ (see, e.g., Ref. 27).

The important result immediately following from Eq. (21) is that if the external excitation field is circularly polarized then the emitted photons are disentangled. Indeed, in this case, the tensor $\overleftrightarrow{W}(t_1,t_2)$ is represented as a tensor product and, hence, so is the single-particle density matrix, i.e., it corresponds to a pure state.

Representation (20) explicitly shows the tensor $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ as a superposition of the amplitudes of the radiative decay of different two-exciton states through two channels into the two-photon states. Due to the presence of different characteristic frequencies, the time dependence of the total amplitude has rather a complex form, especially within the transitional regime. However, there are several resonances whose amplitudes increase with time and which define the long-time response and, respectively, the time dependence of entanglement. In order to extract these resonances, we consider the long-time limit in the spirit of the Wigner-Weisskopf approximation. The typical term constituting $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ has the form

$$m_{E,E_2}(t) = \int_0^t dt_1 e^{-i(1+\alpha)\omega(t-t_1)}\int_0^{t_1} dt_2 e^{-i(b\omega+E_x)(t_1-t_2)}e^{-iE_{xx}t_2}, \tag{24}$$

where E_x and E_{xx} stand for single-exciton and two-exciton energies, respectively. Here we have taken into account the momentum selection rule [Eq. (18)] and have introduced $\alpha=\sin(\theta_1)/\sin(\theta_2)$ (see Fig. 1) and b stands for either 1 or α depending on particular exciton-photon channel. The easiest way to find the resonant components is to extend the limits of integration over time and over frequencies from $-\infty$ to ∞. Thus, we find

$$m_{E_x,E_{xx}}(t) \propto e^{-iE_{xx}t}\delta[E_{xx}(1+\alpha-b)-E_x(1+\alpha)] \times \delta[\omega(1+\alpha)-E_{xx}]. \tag{25}$$

As follows from this expression, only such terms in Eqs. (21) and (22) contribute into the long limit which satisfy the special resonant condition. The total energy of the emitted pair must be equal to the energy of the two-exciton state. Additionally there is the special "kinematic" requirement imposed on the energies of the involved single- and two-exciton states.

A. Entanglement of photons along the symmetric directions

First we consider the symmetric case ($\theta_1=\theta_2$, see Fig. 2), when $\alpha=1$ and $b\equiv 1$. The only terms contributing to the long-time limit are with the two-exciton energy equal exactly to the doubled energy of the exciton in the exciton-photon channel. The physical meaning of this condition is transparent. Each of the two photons emitted due to the radiative recombination must be in the resonance but since $\theta_1=\theta_2$ and

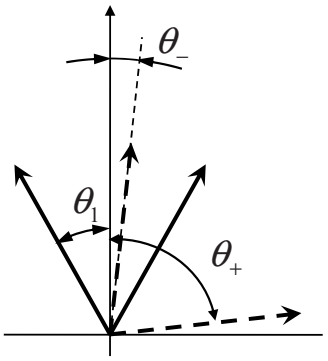

FIG. 2. The emission directions. The symmetric case corresponds to the heavy-heavy and light-light two-exciton resonances. The asymmetric directions $\theta_2=\theta_\pm$ correspond to the heavy-light resonance.

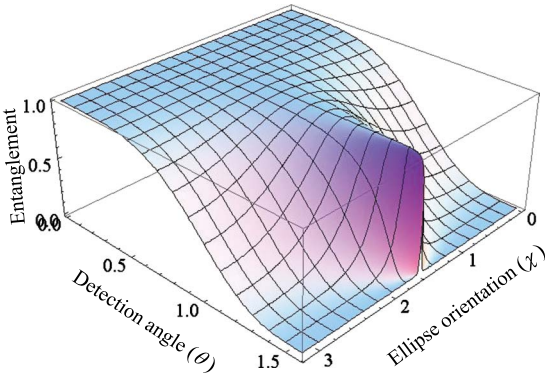

FIG. 3. (Color online) Entanglement near the heavy-hole resonance. Dependence on the detection angle (θ) for the linearly polarized excitation field (solid line left scale) and on ellipticity (β) along $\theta=0$ (dashed line right scale).

by virtue of the momentum selection rule, these photons must be of the same energy. That is the energy of the exciton-photon state should differ from the energy of the two-exciton state by exactly the single-exciton energy. Also we need to consider separately the responses near the energies of the heavy-hole ($\omega_1=\omega_2=E_h$) and light-hole ($\omega_{1,2}=E_l$) excitons. Near $\omega_{1,2}=E_h$, only the first term in W_1 contributes. The non-normalized eigenvalues of the single-particle density matrix are the solutions of the quadratic equation,

$$\rho^2-\frac{\rho}{2}\{\cos(2\chi)\sin^2(\beta)\sin^4(\theta)+[1+\cos^2(\beta)]\times[1+\cos^2(\theta)]^2\}+\sin^4(\beta)\cos^4(\theta)=0, \quad (26)$$

where β and χ are the polar and azimuthal angles on the Poincare sphere describing the polarization state of the excitation field,[28] $A_+=e^{i\chi/2}\cos(\beta/2)$ and $A_-=e^{-i\chi/2}\sin(\beta/2)$. So that $\beta=0,\pi,\pi/2$ correspond to the left and right circular and linear polarizations, respectively, and $\chi/2$ is the angle between the axis of the ellipse of polarization and the projection of $\mathbf{k}_1$ on the plane of the QW.

The dependence of entanglement on the direction of the outgoing photons and on the parameters of the excitation pulse is relatively simple in this case (see Fig. 3). As the special feature the flat maximum near $\theta=0$ should be emphasized. Near this angle one has for arbitrary polarization of the excitation field $\partial E/\partial\theta\propto\sin^3(\theta)$. This makes the entanglement practically angular independent for small angles and being determined by the non-normalized eigenvalues of the reduced density matrix, $\rho_{1,2}=[1\pm\cos(\beta)]^2$. Thus near $\theta=0$, the entanglement reaches its maximum value $E_N=1$ for the linear polarization, $\beta=\pi/2$, and monotonously decreases with decreasing the degree of ellipticity. Moreover, for linearly polarized light when $\chi=\pi/2$ entanglement reaches the maximum, $E_N=1$, and is independent of θ. It should be noted, however, that as follows from Eq. (26), the eigenvalues of the reduced density matrix are $\rho_{1,2}\propto\cos^2(\theta)$ in this case so the signal vanishes in the direction $\theta=\pi/2$.

Using Eq. (14) in the polarization basis, the two-photon states are written as $|\Psi\rangle=\Sigma_{\boldsymbol{\epsilon},\boldsymbol{\epsilon}'}\Psi_{\boldsymbol{\epsilon},\boldsymbol{\epsilon}'}|\boldsymbol{\epsilon},\boldsymbol{\epsilon}'\rangle$, where $\Psi_{\boldsymbol{\epsilon},\boldsymbol{\epsilon}'}$ are found convoluting the polarization vectors of the outgoing photons $\boldsymbol{\epsilon}$ and $\boldsymbol{\epsilon}'$ with $\overleftrightarrow{M}_{\hat{k}_1,\hat{k}_2}$. Along the direction $\theta\approx 0$, where $E_N=1$ can be reached, we obtain

$$|\Psi\rangle\propto -e^{i\chi}[1-\cos(\beta)]|+\rangle|+\rangle-e^{-i\chi}[1+\cos(\beta)]|-\rangle|-\rangle, \quad (27)$$

where $|+\rangle$ ($|-\rangle$) is the state of a photon that is right (left) circularly polarized. As θ is increased, the entanglement is reduced down except for the case $\beta=\pi/2$, $\chi=\pi/2$, where the state is of the same structure as for $\theta=0$, i.e., $|\Psi\rangle\propto|+\rangle|+\rangle-|-\rangle|-\rangle$, as long as $\theta<\pi/2$. Along $\theta=\pi/2$, the two-photon state is $|\Psi\rangle\propto[\cos(\chi)-i\cos(\beta)\sin(\chi)]|s\rangle|s\rangle$, i.e., the two-photon state is completely disentangled. Here the two-photon state is expressed in terms of the s and p polarization eigenstates. Varying the ellipticity of the incoming pulse away from linear polarization $\beta=\pi/2$, the entanglement is monotonously reduced down to $E_N=0$ for $\beta=0$. There the two-photon state reads $|\Psi\rangle=|\theta_1\rangle|\theta_1\rangle$ with $|\theta_1\rangle=[i|s\rangle+\cos(\theta)|p\rangle]/\sqrt{1+\cos^2(\theta)}$, which is also completely disentangled. We would like to emphasize that all these states are triplet, that is transform according to the three-dimensional representation of the rotation group.

For the photons in resonance with the light-hole excitons ($\omega_1=\omega_2=E_l$), the directional dependence of entanglement is more complex. The reason is the interaction of obliquely propagating p-polarized photons with the light-hole excitons with the zero projection of the total spin. Now both terms $\propto W_{1,3}$ contribute resulting in the equation for the non-normalized eigenvalues of the density matrix, which differs from Eq. (26) by the term

$$X\{X[\rho-1+\sin^2(\beta)\sin^2\chi]+2\cos(\chi)\cos^2(\theta)[\rho-\sin^2(\beta)]\} \quad (28)$$

in the right-hand side with $X=8\sin(\beta)\sin^2(\theta)$.

This term becomes important for oblique directions thus yielding a richer structure of the angular dependence of entanglement [see Fig. 4(a)]. Now the maximum value of entanglement is reached not at a fixed angle (say, at $\theta=0$ as in the previous case) but along the direction, which is determined by the ellipticity of the excitation field. This depen-

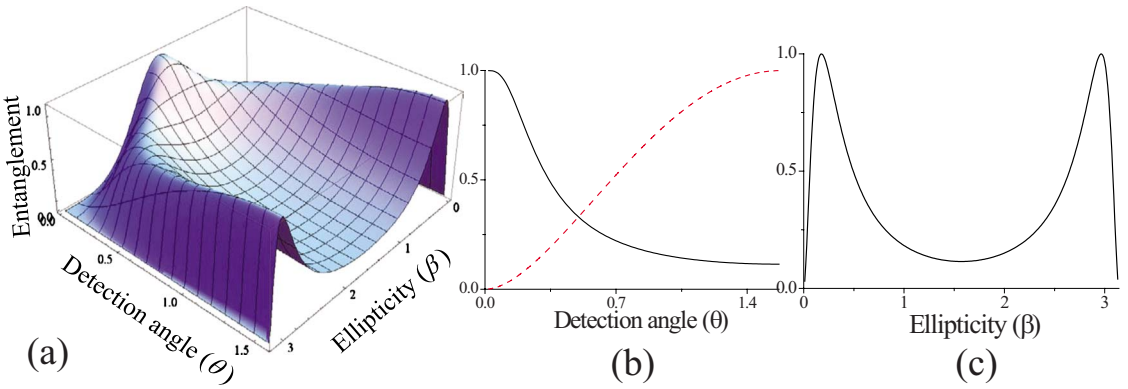

FIG. 4. (Color online) Entanglement (vertical axes, scale from 0 to 1) in the vicinity of the heavy-hole exciton resonance. (a) Dependence on the angle between the outgoing photons and polarization of the excitation field $E_N(\theta,\beta,\chi=0)$. (b) Entanglement as a function of the detection angle for the fixed helicity: $E_N(\theta,\beta=0,\ \chi=0)$ (solid line) and $E_N(\theta,\beta\approx\pi/18,\ \chi=0)$ (dashed line). (c) Dependence on the ellipticity $E_N(\theta=\pi/2,\beta,\ \chi=0)$.

dence is well approximated for θ not too close to $\pi/2$ by the equation

$$\cos^2(\beta) - 8[1-\cos^4(\beta)]\sin^2(\theta) = 0. \tag{29}$$

It should be noted that while the value of entanglement along these directions is significant, $E_N > 0.5$, the maximum value $E_N=1$ is reached only along $\theta=0$ and $\theta=\pi/2$ [see Figs. 4(b) and 4(c)]. For example, for the latter case, the difference between the roots of the characteristic equation is $\propto 65\ \sin^2(\beta)-2$ and it vanishes for $\beta\approx\pi/18$ implying maximum entanglement. Such small angles on the Poincare sphere correspond to polarization close to the circular. This is the surprising result considering that, as has been shown above, circularly polarized excitation always produces disentangled photons.

The two-photon states, at which the maximal entanglement is reached are given by Eq. (27) along $\theta\approx 0$ and by $|\Psi\rangle \propto -[\cos(\chi) - i\ \cos(\beta)\sin(\chi)]|s\rangle|s\rangle + 8\ \sin(\beta)|p\rangle|p\rangle$ along $\theta=\pi/2$.

B. Entanglement along asymmetric directions

The asymmetric case, when $\theta_1 \neq \theta_2$ is less complex. There is only one resonance in this case, when the two-exciton state is made of light-hole and heavy-hole excitons. Respectively, there are only two resonant directions with $\theta_1 \neq \theta_2$ (see Fig. 2) with

$$\sin(\theta_\pm) = \sin(\theta_1)\left(\frac{E_1}{\Delta_{\rm hl}}\right)^{\pm 1}. \tag{30}$$

The only contribution to the two-photon amplitudes in this case originates from the term $\propto W_2$ in Eq. (21) and only from the part $\propto e^{-iE_1(t_1-t_2)}$ in the expression for W_2 in Eq. (22). Thus the reduced single-photon density matrix up to the normalization factor is determined by Eq. (19) with

$$\overleftrightarrow{M}_{\hat{\mathbf{e}}_1,\hat{\mathbf{e}}_2} = A_+A_-(\overleftrightarrow{1} - \hat{\mathbf{e}}_z \otimes \hat{\mathbf{e}}_z). \tag{31}$$

The non-normalized eigenvalues of the single-photon density matrix are 1 and $1+\cos^2(\theta)$. As a result, entanglement monotonously decreases from 1 to 0 (see Fig. 5) as θ changes from 0 to $\pi/2$ while the direction of the detection of the second photon is determined by Eq. (30). It should be noted that for sufficiently large detection angles θ such that $\sin(\theta) > \Delta_{\rm hl}/E_1$ only one resonant direction, namely, corresponding to θ_-, remains. It should be noted that in the asymmetric case, the entanglement is independent of the polarization of the excitation field. However, if the excitation pulse is circularly polarized then the response described by Eq. (31) completely vanishes.

IV. YIELD OF ENTANGLED PHOTONS

Studying entanglement would not be complete without considering yield, which is defined as the ratio of the energy flux carried by the entangled pairs of the photons to the energy flux of the excitation field. For practical purposes, it is more convenient to use an alternative definition,

$$Y = \frac{N_{\rm out}}{N_{\rm in}}, \tag{32}$$

where $N_{\rm out}$ and $N_{\rm in}$ are the number of outgoing and incoming *pairs*, respectively. The number of pairs for the excitation field is formally defined as $N_{\rm in}=(\Phi/\hbar\Omega)^2$, where Φ is the total flux of the external field and Ω is its frequency in the stationary frame. As follows from this definition, yield is the

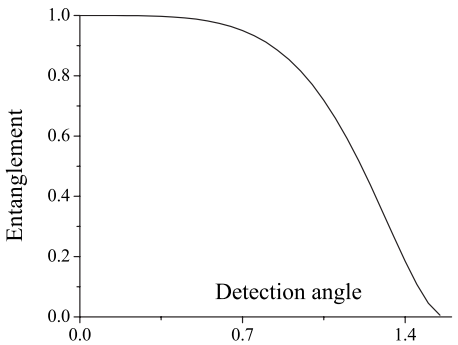

FIG. 5. Angular dependence of entanglement for the nonsymmetric detection.

long-time limit of $\mathrm{Tr}(\rho)$ with ρ being the two-particle density matrix given by Eq. (2). Direct taking the limit $t\to\infty$, however, fails because the density matrix diverges $\propto t^4$ due to the resonances discussed above. The origin of divergence is the perturbatively treated degeneracy of a two-exciton and a single exciton plus photon states. Rigorously this problem is resolved in a full dynamical theory, which accounts for lifting the degeneracy by the exciton-photon interaction. This approach is beyond the scope of the present paper. In order to estimate the yield, we use the observation that in reality, the long-time limit is naturally bounded from above by the coherence time after which the two-particle amplitudes $\Psi_{\hat{k}_1,\hat{k}_2}$ do not provide the correct description of the two-particle states any longer. From the perspective of the dynamical theory, this assumption effectively removes the degeneracy by the value $\sim\gamma=\hbar\tau^{-1}$, where τ is the coherence time. Thus, as usual, the perturbation theory is valid if the exciton-photon coupling is not too strong. Calculating the trace of the single-particle density matrix over the polarization quantum numbers we find (in SI units)

$$Y\sim\frac{4\pi^9 E_{\mathrm{x}}}{3\gamma^3 S}(\rho_1+\rho_2)\left(\frac{T|Q|^2}{\hbar\Omega c^3\epsilon_0^2}\right)^2, \tag{33}$$

where E_{x} is the exciton energy in the stationary frame, $\rho_{1,2}$ are the non-normalized eigenvalues of the density matrix defined by the equations studied above, S is the area of the excitation spot, T is the duration of the excitation pulse, and ϵ_0 is vacuum permittivity. The inverse dependence on the pump area is clear since denser excitation results in more pronounced effect of the exclusion principle. The dependence on the pulse duration is the consequence of the semiconductor response to be determined by the polarizations of the linear response [see Eq. (17)] rather than by the energy input. In the limit of short excitation pulse, the polarizations depend linearly on time so that $\mathrm{Tr}[\rho]\propto T^4$. The total flux of the external field also depends linearly on time so that $N_{\mathrm{in}}\propto T^2$ resulting in $Y\propto T^2$. Substituting the values typical for GaAs, $\gamma=1.5$ meV, $\hbar\Omega\approx E_{\mathrm{x}}=1.5$ eV, $Q=1.3\times10^{-24}$ kg m/s (see, e.g., Ref. 27) and using $T=100$ fs, $S=\pi(20)^2$ μm^2, we find $Y\sim0.01$. Such high value of yield is the result of the resonant transitions between the many-particle states.

V. CONCLUSION

We have studied entangling photons by means of the nonlinear optical response of semiconductor quantum wells. We have considered the excitation of the quantum well by a short pulse and have analyzed entanglement of the outgoing photons emitted in the course of the radiative relaxation. The important result is that the Pauli blocking alone results in entanglement between the polarizations of the emitted photons.

Spectrally, the response is especially strong in the vicinity of several characteristic frequencies corresponding to the resonant decay of different two-exciton states, light-light (ll), heavy-heavy (hh), and light-heavy (lh). Together with the restriction imposed on the momenta of the outgoing photons, this defines the resonant directions along which the entangled photons propagate. We show that the two-photon states are triplets. In the short-time limit, when the effect of the bound two-exciton states is not pronounced, the directions are determined by the relations between the energies of the single-exciton states constituting the pair. Thus, the directions are symmetric for hh and ll states, and are asymmetric of the lh state. It can be conjectured that the effect of the biexcitons would manifest in breaking the symmetry of the resonant directions for hh and ll states.

The dependence of entanglement on the angle between the outgoing photons is especially nontrivial for the ll state owing to the complex form of the dipole moment. Managing the polarization of the excitation field, one can control the direction along which the most entangled photons propagate.

ACKNOWLEDGMENTS

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APPENDIX: COULOMB INTERACTION

The effect of Coulomb interaction in the chosen approximation is given by the second term in Eq. (16). The full description of the complex effects of the Coulomb interaction requires more precise approach than used in the present paper. Therefore, we restrict ourselves to the qualitative discussion of the short-time scale dynamics and show that the Coulomb interaction in this limit does not lead to significant changes in the results presented in the main text.

The main assumption we use for this consideration is that the Coulomb term in Eq. (16) can be taken in the short memory approximation, thus neglecting the frequency shifts and the respective formation of additional skewed resonances, that is the resonances along asymmetric detection directions. This approximation was tested in Ref. 29 for the problem of the 2D Fourier spectroscopy, where it provided very good agreement with the experiment. In the framework of the short memory approximation, the Coulomb term is presented as

$$\mathcal{C}_{\mu_1,\mu_2}\approx\sum_{\nu_3,\nu_4}\langle B_{\mu_1}B_{\mu_2}F(H_{SC})D_{\nu_3,\nu_4^\dagger}\rangle P^{(1)}_{\nu_3}(t)P^{(1)}_{\nu_4}(t), \tag{A1}$$

where $F(H_{SC})$ is either a c function or an analytical function of the nonperturbed Hamiltonian H_{SC}. Using the fact that the Hamiltonian preserves the number of particles with the particular values of spins and expanding B and D in terms of the electron and hole operators, Eq. (A1) can be rewritten as

$$\mathcal{C}_{\mu_1,\mu_2} \approx \sum_{\nu_3,\nu_4} \int d\mathbf{x}_1 d\mathbf{y}_1 \ldots d\mathbf{x}_4 d\mathbf{y}_4 \langle v_{\sigma_{\mu_2}}(\mathbf{x}_2) v_{\sigma_{\mu_1}}(\mathbf{x}_1) v^{\dagger}_{\sigma_{\nu_3}}(\mathbf{x}_3) v^{\dagger}_{\sigma_{\nu_4}}(\mathbf{x}_4)\rangle \langle c_{s_{\mu_1}}(\mathbf{y}_1) c_{s_{\mu_2}}(\mathbf{y}_2) c^{\dagger}_{s_{\nu_3}}(\mathbf{y}_3) c^{\dagger}_{s_{\nu_4}}(\mathbf{y}_4)\rangle$$
$$\times \Phi^{\nu_3,\nu_4}_{\mu_1,\mu_2}(\mathbf{x}_1,\mathbf{y}_1,\ldots,\mathbf{x}_4,\mathbf{y}_4) P^{(1)}_{\nu_3}(t) P^{(1)}_{\nu_4}(t), \tag{A2}$$

where while arranging the electron and hole operators we have taken into account that averaging is taken over semiconductor vacuum, i.e, empty conduction band and filled valence band. The function Φ, whose explicit form essentially depends on the physical origin of short memory, is invariant with respect to homogeneous translations of all coordinates.

One can easily check that the general structure of $\mathcal{C}_{\mu_1,\mu_2}$ is the same as that of the first term in Eq. (16) and, in particular, it does not produce new terms. Thus one again arrives at Eqs. (20) and (22) but with modified $\mathcal{I}_{\sigma_1,\sigma_2}$ comparing to Eq. (23). This modification affects the time dependence of tensor $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}$ during the transitional regime and the value of yield. It, however, leaves intact the properties of entanglement of the resonant terms, which are the object of main interest in the present paper. As is shown in the main text, the reason is that the entanglement due to the resonant terms does not depend on the amplitudes $\mathcal{I}_{\sigma_1,\sigma_2}$, which are factored out after the normalization of the single-photon density matrix.

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