Entangled Photon Pairs from Semiconductor Quantum Dots

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Tomographic analysis demonstrates that the polarization state of pairs of photons emitted from a biexciton decay cascade becomes entangled when spectral filtering is applied. The measured density matrix of the photon pair satisfies the Peres criterion for entanglement by more than 3 standard deviations of the experimental uncertainty and violates Bell's inequality. We show that the spectral filtering erases the "which path" information contained in the photons' color and that the remanent information in the quantum dot degrees of freedom is negligible.

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Entanglement, the intriguing correlations of quantum systems [1–3], is an essential resource of quantum information and communication [4–6]. Entangled photons are particularly attractive for applications due to their noninteracting nature and the ease by which they can be manipulated. Polarization entangled photons are routinely produced by nonlinear optical effects, predominantly by parametric down-conversion [7,8]. Such sources have a large random component whereas quantum data processing schemes require nonrandom, or "event-ready," entangled photons.

Semiconductor quantum dots (QDs) [9–12] provide optically and electrically driven sources of single photons on demand [12–14]. Compatibility with modern electronics makes them potential building blocks for quantum information processing in general [15] and sources for event-ready entangled photons [16,17] in particular.

A QD biexciton decays radiatively through two intermediate optically active exciton states [18,19]. The proposal that the biexciton-radiative cascade could provide a source of event-ready polarization entangled photon pairs was made by Benson *et al.* [16]. Entanglement requires two decay paths with different polarizations, but indistinguishable otherwise. This is the case if the intermediate exciton states are energetically degenerate and if the final state of the QD is independent of the decay path.

The first requirement is difficult to fulfill since the intermediate exciton states are split by the anisotropic electron-hole exchange interaction [20,21]. The two decay paths, which we denote horizontal (H) and vertical (V), have corresponding photon polarizations relative to the asymmetry axis of the QD [see Fig. 1(b)]. Since the paths are spectrally distinguishable the polarization state of the photons cannot be entangled [22]. The second requirement, that the QD final state does not depend on the decay path, has never been tested experimentally.

In this Letter, we show for the first time [23] that the "which path" information can be erased by filtering the photons spectrally and that this procedure produces an entangled polarization state. We also prove that the remnant which path information residing in the myriad degrees of freedom of the QD must be small.

We denote by $|HH\rangle$ ($|VV\rangle$) the two photons' polarization states associated with the H (V) decay path. The final state of the cascade is given by

$$|\psi\rangle = \alpha|p_H\rangle|HH\rangle|d_H\rangle + \beta|p_V\rangle|VV\rangle|d_V\rangle. \quad (1)$$

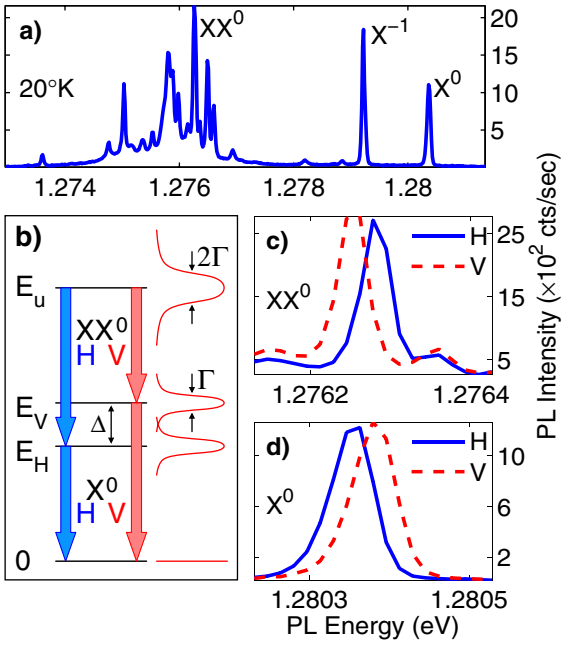

FIG. 1 (color online). (a) PL spectrum of a single QD. (b) Schematic description of the biexciton-radiative cascade in natural QDs with its two colinearly polarized photons (either H or V) and energetically distinguishable paths. (c), (d) High resolution polarization sensitive PL spectra of the biexciton XX^0 and exciton X^0 lines, respectively. Because of spectral diffusion (see text), the measured line widths are larger than the radiative widths, shown schematically in (b).

Here α and β are the amplitudes of the decay paths ($|\alpha|^2 + |\beta|^2 = 1$). The photons' wave packet is denoted by $|p_{H(V)}\rangle$ and the final states of the QD by $|d_{H(V)}\rangle$. All may, *a priori*, depend on the decay path. We neglected the broken paths, $|HV\rangle$ and $|VH\rangle$, which have negligible amplitudes [24]. The density matrix for the two photons polarization state is given by tracing out the $|p\rangle$ and $|d\rangle$ degrees of freedom:

$$\rho = \begin{pmatrix} |\alpha|^2 & 0 & 0 & \gamma \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ \gamma^* & 0 & 0 & |\beta|^2 \end{pmatrix}, \qquad \gamma = \alpha\beta^* \langle p_H|p_V\rangle\langle d_H|d_V\rangle. \tag{2}$$

The phase of γ is a gauge dependent quantity.

The Peres criterion [25] applied to Eq. (2) says that the polarization is entangled provided $\gamma \neq 0$. If $|p_H\rangle|d_H\rangle$ is orthogonal to $|p_V\rangle|d_V\rangle$ the decay paths can be reliably distinguished, and the polarization state is not entangled (though the photons are correlated since there are no HV or VH events). If $\gamma \neq 0$, interrogating $|p\rangle|d\rangle$ does not select a unique path and the state is entangled. Maximal entanglement, $|\gamma| = \frac{1}{2}$, is obtained when the two paths have equal weights and $|p_H\rangle|d_H\rangle$ is parallel to $|p_V\rangle|d_V\rangle$.

In general, entanglement need not imply violation of Bell's inequality [2] but, for ρ of Eq. (2) it does. This follows from the results of Horodecki *et al.* [26] that yield for the measurement of the optimal Bell operator B [3]:

$$\langle B\rangle = \mathrm{Tr}(B\rho) = 2\sqrt{1 + 4|\gamma|^2}, \tag{3}$$

violating Bell's inequality $-2 \leq \langle B\rangle \leq 2$ for all $\gamma \neq 0$.

Suppose $\langle p_H|p_V\rangle = 0$. Then $\gamma = 0$ and the state is not entangled. Nevertheless, one can entangle such a state by applying a projection P on the wave packet. This replaces $|\psi\rangle$ of Eq. (1) by $P|\psi\rangle/|P|\psi\rangle|$ and gives

$$\gamma' = \frac{\alpha\beta^*\langle p_H|P|p_V\rangle}{|P|\psi\rangle|^2}\langle d_H|d_V\rangle. \tag{4}$$

The choice $\alpha^* P|p_H\rangle = \beta^* P|p_V\rangle$ gives $\gamma' = \frac{1}{2}\langle d_H|d_V\rangle$. Maximal entanglement is then obtained when the final state of the dot $|d\rangle$ does not depend on the decay path. We see that $|\gamma'|$ can be significant even though γ is negligible. If the final state of the QD can reliably distinguish between the decay paths (e.g., due to involvement of phonons in the cascade) no entanglement will ever arise.

Second-order perturbation theory within the dipole and rotating wave approximation [27] gives

$$A_H \equiv \alpha\langle \mathbf{k}_1, \mathbf{k}_2|p_H\rangle = \frac{e^{i\phi_H}\Gamma/2\pi}{(|\mathbf{k}_1| + |\mathbf{k}_2| - \epsilon_u)(|\mathbf{k}_2| - \epsilon_H)}, \tag{5}$$

with a similar expression for A_V. The index u denotes the initial biexciton state. The momentum of the photons is labeled $\mathbf{k}_1$, $\mathbf{k}_2$ (using units in which $\hbar = c = 1$), and $\epsilon_j = E_j - \frac{i}{2}\Gamma_j$, $j = u, H, V$ are complex energies with radiative widths $\Gamma_u = 2\Gamma_{H/V}$. P is implemented by a function W representing two spectral windows of width w, centered at $(E_V + E_H)/2$ and $E_u - (E_V + E_H)/2$. Plugging this into Eqs. (4) and (5) gives

$$\gamma' = \frac{\iint d\mathbf{k}_1 d\mathbf{k}_2 A_H^* W A_V}{\iint d\mathbf{k}_1 d\mathbf{k}_2 A_H^* W A_H + \iint d\mathbf{k}_1 d\mathbf{k}_2 A_V^* W A_V}. \tag{6}$$

The denominator is the photon-pair detection probability.

Spectral diffusion, due to changes in the QD environment, causes the energies E_j to fluctuate slowly relative to the radiative time. This leads to inhomogeneous broadening of the spectral lines [see Figs. 1(c) and 1(d)]. One may worry that it restores the which path information in the decay path, if the spectral filters are narrower than the linewidth. This is not the case, since in a given radiative cascade both photons are shifted equally relative to the energy of their fixed spectral filters. Therefore, large fluctuations result in a rapid decrease in the probability of detecting both photons.

Numerical calculations of the joint detection probability $|P|\psi\rangle|^2$ and the measure of entanglement $|\gamma'|$ with and without spectral fluctuations are shown in Fig. 4.

For the measurements we used planar microcavity (MC) embedded QD samples. The samples were grown by molecular beam epitaxy on a (100) oriented GaAs substrate. One layer of strain-induced InAs QDs was deposited in the center of a one wavelength GaAs microcavity formed by two unequal stacks of alternating quarter wavelength layers of AlAs and GaAs, respectively. The height and composition of the QDs were controlled by partially covering the InAs QDs with a 3 nm layer of GaAs and subsequent growth interruption. To improve photon collection efficiency, the microcavity was designed to have a cavity mode, which matches the QD emission due to ground state e-h pair recombinations. Unlike previous studies of radia-

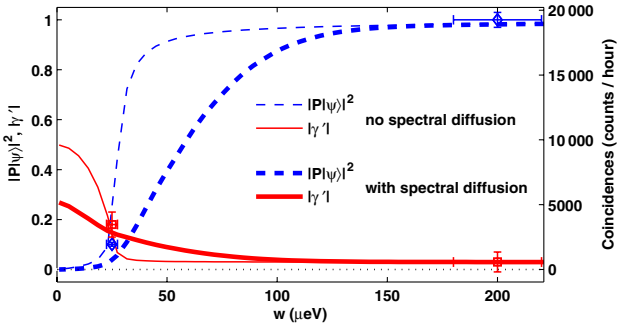

FIG. 4 (color online). Measured (symbols) and calculated (lines) normalized number of coincidences $|P|\psi\rangle|^2$ (dash-lines) and off diagonal density matrix element $|\gamma'|$ (solid-lines) vs the spectral window width w. For the calculations we used $\Gamma = 1.6$ μeV and $\Delta = 27$ μeV, as experimentally determined. Thick (thin) line represents calculation which includes (ignores) spectral diffusion averaged over normally distributed photon energy shifts, with width of 50 μeV at half-maximum.

tive cascades in QDs [28], our sample was not masked or patterned laterally to prevent obscuration of the emitted photon polarizations.

We used a diffraction limited low temperature confocal optical microscope for the photoluminescence (PL) studies of single MCQDs [9,19]. Temporal correlations between emitted photon pairs were measured using a wavelength and polarization selective Hanbury Brown–Twiss (HBT) arrangement [19]. We used a 1 m monochromator in each arm of the HBT setup to obtain spectral resolution of ~ 15 μeV. The polarization state of the emitted light was monitored by the use of liquid crystal variable retarders and high quality polarizers.

In Fig. 1(a) we present PL spectrum of a single, resonant MCQD. The MCQD was excited by a continuous-wave HeNe laser. The spectrum is composed of sharp lines with linewidths of roughly 50 μeV due to the spectral diffusion. We identified most of the observed spectral lines using power and energy dependence polarization sensitive magnetospectroscopy. Here, we are only interested in the neutral single exciton line (X^0) and the neutral biexciton line (XX^0). In Figs. 1(c) and 1(d) we present high resolution polarization sensitive PL spectra of the lines X^0 and XX^0. Figure 1 demonstrates that the neutral spectral lines XX^0 and X^0 are composed of two cross-linearly polarized split doublets with detuning of $\Delta = 27 \pm 3$ μeV. We used the HBT setup to measure polarization sensitive temporal intensity correlations between photons emitted from all the observed spectral lines. The autocorrelation measurements of the lines (not shown), show a deep antibunching notch at coinciding times ($t = 0$), demonstrating that each line is a spectral source of single photons [12–14]. The intensity cross-correlation measurements between the neutral exciton X^0 and the neutral biexciton XX^0 lines are presented in Figs. 2(a) and 2(b) for two different colinear (cross-linear) polarization arrangements. For these measurements, the excitation intensity was tuned such that both lines were essentially equal in strength. Under these conditions, we detect around 50 000 photons/sec from each line. When the two polarizers are colinearly oriented along the major QD axes (HH or VV) an asymmetric trace is obtained in which the positive temporal part shows an antibunching notch, while the negative part shows a strong, enhanced bunching peak. This asymmetrical shape, an experimental signature of an optical cascade, reveals the temporal sequence of these events. While emission of a horizontally (vertically) polarized XX^0 photon is followed by emission of a horizontally (vertically) polarized X^0 photon, the opposite never happens [16,17]. The coincidences at positive times are thus due to pairs of photons which do not occur in the same radiative cascade. When the polarizers are cross-linearly polarized the bunching trace at negative times is replaced by a deep (down to zero within the experimental uncertainty) anti-bunching-like trace, while the positive trace remains the same. This is exactly as anticipated by the considerations of Fig. 1(b). The linear polarization states of the two photons emitted

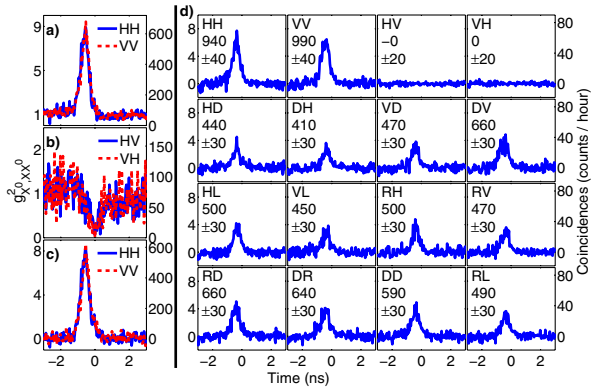

FIG. 2 (color online). (a) [(b)] The temporal intensity cross-correlation measurements between the exciton X^0 and the biexciton XX^0 spectral lines for collinear [cross-linear] polarizations. (c) The same-cascade correlation measurements obtained by the difference between the collinearly polarized curves in (a) and the average curve of the cross-linearly polarized measurements in (b). (d) Tomographical measurements of the same-cascade intensity cross-correlation functions with spectral resolution of 25 μeV. D stands for linear polarizer at 45° relative to the H direction and R (L) stands for right (left) hand circular polarizer. The integrated numbers of coincidences in each measurement are indicated.

during the same biexciton-exciton radiative cascade are completely correlated: both are collinearly polarized and there are no cross-polarized two photon states. It follows that all the measured coincidences in Fig. 2(b) are due to photon pairs from two distinct radiative cascades. The polarizations of such photons are completely uncorrelated, as can be easily verified by inspecting the correlation traces at positive time. Hence, by subtracting the cross-polarized measurements, one obtains the net ''same-cascade'' correlation functions, which include only photon pairs emitted in the same radiative cascade [Fig. 2(c)].

From the temporal correlation measurements we obtain the radiative lifetime of the exciton [19] $T_{X^0} = 0.8 \pm 0.2$ nsec, or $\Gamma = 1.6 \pm 0.4$ μeV for its radiative width.

We performed two sets of independent measurements of the intensity cross-correlation function in 16 different combinations of the polarizers in front of the two detectors. The spectral projection of the photons was implemented by the two monochromators' slits. In the first set the monochromators' slits were opened to spectral resolution of 200 μeV, as independently determined by measuring the spectral lines of a low pressure mercury lamp. In the second set the resolution was set to 25 μeV. This set is shown in Fig. 2(d).

The normalized integrated numbers of coincidences from these polarization tomography measurements [29] are then used to generate the H-V base density matrix of the photon pair generated in the same cascade. In Fig. 3 we show the density matrices obtained by tomography of same-cascade correlations. The experimentally obtained density matrices were fitted to the form of Eq. (2). With

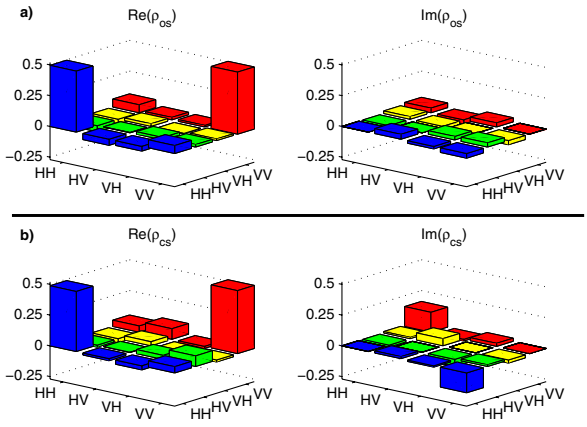

FIG. 3 (color online). The measured two photons' density matrix for photon pairs from a biexciton cascade: (a) [(b)] obtained with spectral window of 200 [25] μeV.

open slits we obtained $|\alpha|^2 = |\beta|^2 = 0.50 \pm 0.02$ and $\gamma' = 0.03 \pm 0.04 + i(0.00 \pm 0.04)$. This agrees with the estimate of $|\gamma'| = \Gamma/2\Delta \approx 0.03$. With closed slits we obtained $|\alpha|^2 = |\beta|^2 = 0.50 \pm 0.04$ and $\gamma' = 0.05 \pm 0.05 + i(0.17 \pm 0.05)$ hence $|\gamma'| = 0.18 \pm 0.05$ is significantly different from zero. The photon pairs of the biexciton cascade are therefore entangled with confidence level greater than 3 standard deviations of the measurement uncertainty. Substituting $|\gamma'|$ in Eq. (3) gives 2.13 ± 0.07 which violates Bell's inequality.

While the above analysis holds for photon pairs emitted from the same radiative cascade, one can also ask whether the raw correlations show evidence for entanglement [30]. The answer depends on the measurements' temporal window width. The events in a wide window will be dominated by distinct cascades with uncorrelated polarizations. In a sufficiently narrow time window, however, same-cascade events will dominate, and the two photon state should be entangled. For a temporal window of 0.6 nsec, centered around the antibunching notch [Fig. 2(b)], we minimize the number of events from distinct cascades while maintaining reasonable same-cascade statistics. The density matrix obtained from the raw correlations in this case has partial transpose with negative eigenvalue of -0.14 ± 0.06, clearly satisfying Peres criterion for entanglement.

The measured data points at 25 μeV and 200 μeV are in quantitative agreement with the calculations shown in Fig. 4. This supports the absence of significant which path information in the final state of the QD.

The entangled photons generated in this work are not event ready for two reasons: first, we used continuous excitation and, second, the erasure introduces randomness. However, neither is fundamental. The excitation can be triggered on demand and the randomness can be overcome by spectrally monitoring the discarded photons without demolishing the entangled pair. Reducing the detuning [21] and increasing the radiative width through the Purcell effect [13,27] is also a possibility.

In summary, we have demonstrated that the biexciton-exciton radiative cascade in single semiconductor quantum dots is a source for entangled photon pairs. The quantum tomography data satisfy the Peres criterion for entanglement and violate Bell's inequality. We show that the which path information residing in the photons color can be erased and that there is no remanent which path information left in the quantum dot.

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