Correlated and entangled pairs of single photons from semiconductor quantum dots*

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(Received 4 August 2006; accepted 4 November 2006; published online 27 April 2007)

Entangled photon pairs are emitted from a biexciton decay cascade of single quantum dots when spectral filtering is applied. We show this by experimentally measuring the density matrix of the polarization state of the photon pair emitted from a continuously pumped quantum dot. The matrix clearly satisfies the Peres criterion for entanglement. By applying in addition a temporal window, the quantum dot becomes an entangled light source. © *2007 American Institute of Physics*.
[DOI: 10.1063/1.2722769]

I. INTRODUCTION

Entanglement, the intriguing correlations of quantum systems,[1,2] is an essential resource of quantum information and communication.[3,4] 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.[5,6] Such sources have, however, a large random component, whereas applications require nonrandom entangled photons.

Semiconductor quantum dots (QDs) provide optically and electrically driven sources of single photons on demand.[7–9] Compatibility with modern electronics makes them potential building blocks for quantum information processing[10] and sources for "event-ready" entangled photons.[11,12]

In a recent Letter[13] we demonstrated that the polarization state of pairs of photons emitted from a biexciton decay cascade becomes entangled when spectral filtering is applied. Though suggestive evidence for entanglement was claimed by Stevenson *et al.*,[14] who measured correlations between photons from radiative cascades in QDs with almost degenerate exciton states, entanglement without spectral filtering has not been demonstrated yet.[13,15]

For our demonstration, we use continuous wave (cw) optical pumping, since this way one maximizes the emission rate from a spontaneously decaying source of photons. Here, we show that by adding a temporal filter, a single QD undergoing a cw excitation becomes a physical source of entangled light.

A QD biexciton decays radiatively through two intermediate optically active exciton states.[16,17] 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.[18] 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.[19] We overcome this requirement by using spectral filtering.[13] The requirement that the QD final state does not depend on the decay path has been recently shown experimentally.[13]

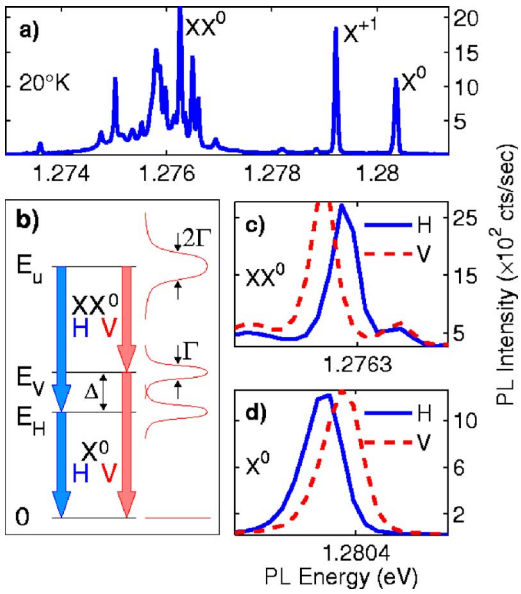

FIG. 1. (Color online) (a) Photoluminescence (PL) spectrum of a single QD. (b) Schematic description of the biexciton radiative cascade in natural QDs with its two colinearly polarized photons (either H or V) and energetically distinguishable paths. (c), (d) High-resolution polarization-sensitive PL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

*This paper is based on a talk presented by the authors at the 18th International Conference on the Physics of Semiconductors, which was held 24–28 July 2006, in Vienna, Austria. Contributed papers for that conference may be found in "Physics of Semiconductors: 28th International Conference on the Physics of Semiconductors," AIP Conference Proceedings No. 893 (AIP, Melville, NY, 2007); see http://proceedings.aip.org/proceedings/confproceed/893.jsp

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II. EXPERIMENT

For the measurements we used planar microcavity (MC) embedded QD samples.[20] 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 radiative cascades in QDs,[22] 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 the single MCQDs.[17] Temporal correlations between emitted photon pairs were measured using a wavelength- and polarization-selective Hanbury-Brown-Twiss (HBT) arrangement.[17] 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 (LCVRs) and high-quality polarizers.

In Fig. 1(a) we present the 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. Changes in the QD electrostatic environment cause the energy levels of the QD to fluctuate slowly relative to the radiative time. This, in turn, leads to inhomogeneous broadening of the spectral lines.

We identified most of the observed spectral lines using power and energy dependence polarization-sensitive magneto PL spectroscopy.[21] 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 $\Delta=27\pm3$ 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.[7–9] The intensity cross-correlation measurements between the neutral exciton X^0 and the neutral biexciton XX^0 lines are presented in Fig. 2 for 16 different combinations of the polarizers in front of the two detectors. For these measurements, the excitation intensity was tuned such that both lines were essentially equal in strength.

From the temporal correlation measurements we obtain the radiative lifetime of the exciton[17] $T_{X^0}=0.8\pm0.2$ ns, or $\Gamma=1.6\pm0.4$ μeV for its radiative width.

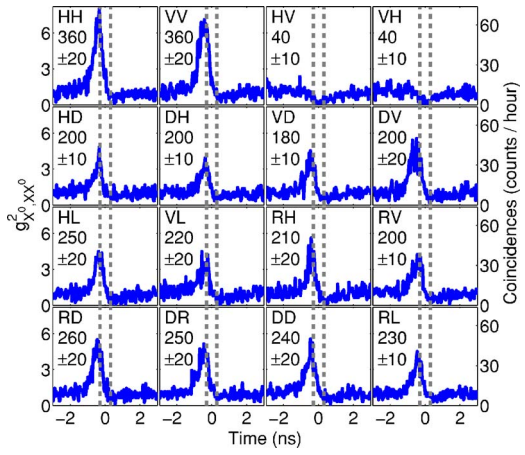

FIG. 2. (Color online) Tomographical measurements of the temporal intensity cross-correlation functions of the exciton X^0 and the biexciton XX^0 spectral lines with spectral resolution of 25 μeV. D stands for linear polarizer at 45° relative to the H direction, and $R(L)$ stands for right (left)-hand circular polarizer. The integrated numbers of coincidences with a temporal window (dashed lines) of 0.6 ns, centered around the antibunching notch in each measurement, are indicated.

Spectral filtering was implemented by the two monochromators' slits, which were closed to obtain spectral width of roughly 25 μeV. Under these conditions, we detect around 5000 photons/s from each line. This amounts to about 10% of the open slit counts.

As schematically described in Fig. 3, it is crucial that the spectral windows be comparable but smaller than the detuning between the two paths. One may worry that the "which path" information in the decay path will not be erased if the spectral filters are narrower than the linewidth due to the spectral diffusion. 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.

The filters are represented in Fig. 3 by the shaded square.

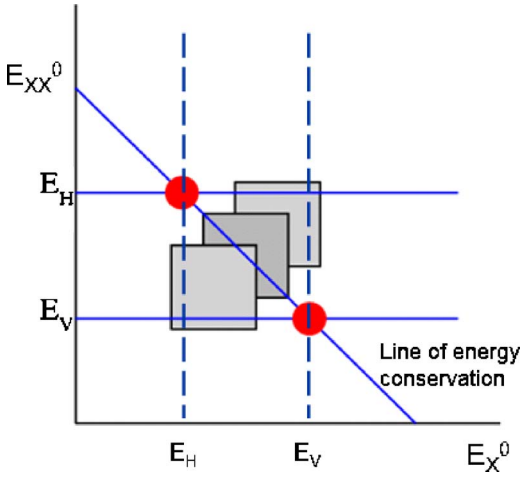

FIG. 3. (Color online) Schematic representation of the spectral filtering and the effect of spectral diffusion. The filter is represented by the shaded square. The spectral diffusion is represented by the meandering of the window along the diagonal as shown by the tiled squares. The diagonal line is the line of total conservation of energy. The solid circles are located where the amplitudes for the H and V decay paths are maximal. As the filter meanders the intersection with the diagonal line decreases. This means effective reduction in the width of the spectral window.

The spectral diffusion can then be represented by the meandering of the square along the diagonal as shown by the tiled squares. The straight lines represent the resonance transitions of the two decay paths. The diagonal line is the line of total conservation of energy. The solid circles are located where the amplitudes are large because both the resonance condition and total conservation of energy are satisfied. The solid circles lie outside the shaded square which selects events which are common to both the *H* and *V* paths. As the filter meanders and gets into the resonance with one of the decay paths, it also gets far from the solid circles so the detection of the two photons has small probability. One way to see this is to look at the intersection with the diagonal line. This means that spectral diffusion leads to effective reduction in the width of the spectral window. Therefore, large fluctuations result in a rapid decrease in the probability of detecting both photons.[13]

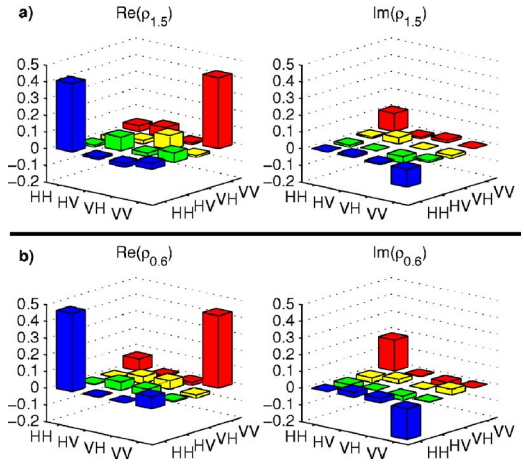

FIG. 4. (Color online) The density matrix of the photon pair polarization state as obtained from the tomography: (a) for temporal window of 1.5 ns and (b) for window of 0.6 ns.

III. RESULTS AND DISCUSSION

By inspecting Fig. 2, we note that 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.[11,12] When the polarizers are cross-linearly polarized (*HV* or *VH*), the bunching trace at negative times is replaced by a deep (down to zero within the experimental uncertainty) antibunching-like trace. This is exactly as anticipated by the considerations of Fig. 1(b). The linear polarization states of the two photons emitted during the same biexciton-exciton radiative cascade are completely correlated, both are collinearly polarized, and the state of the photons emitted in the same radiative cascade does not contain a cross-polarized component. The positive trace, however, in which the exciton photon is detected prior to the biexciton photon, is due to pairs of photons which do not occur in the same radiative cascade. The trace here is the same in both the cross- and copolarized measurements. It therefore follows that all the measured coincidences in the *HV* and *VH* cascades 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.[13] We showed in Ref. 13 that pairs from the same cascade are entangled, significantly violating Bell's inequality.

When the QDs are excited continuously, it is impossible to distinguish, however, between a pair of photons from the same radiative cascade and a pair of photons from distinct ones. Therefore, in order to use a continuously excited QD as an entangled light source, the raw measurements should be considered. Clearly, the events in a wide temporal window will be dominated by distinct cascades with uncorrelated polarizations. In a sufficiently narrow time window, however, events originating from the same cascade will dominate, and the two photon state should be entangled.

We apply a temporal window of 1.5 ns, in which merely all the same cascade pairs were included, and a window of 0.6 ns, centered around the antibunching notch of the raw, spectrally filtered correlation measurements. The number of coincidences from these polarization tomography measurements,[5] within each temporal window, were then used to generate the *H*-*V* base density matrix of the emitted photon pairs.

In Fig. 4 we present the density matrices obtained from these measurements. For the wide temporal window [Fig. 4(a)], the events from distinct cascades are still dominant and the partial transpose of the matrix has marginal negative value of -0.03 ± 0.05, which is not sufficient to claim entanglement. For the window of 0.6 ns [Fig. 4(b)] we minimize the number of events from distinct cascades while maintaining reasonable same-cascade statistics. The obtained density matrix has partial transpose with negative eigenvalue of -0.15 ± 0.07, clearly satisfying the Peres criterion for entanglement.[23]

IV. CONCLUSIONS

By using spectral filtering and temporal gating, we demonstrate that cw excited single quantum dots become sources of entangled light. The entangled photons generated in this work are not event ready since we used continuous excitation and since the erasure introduces randomness. If one can tolerate lower rates, the excitation can be triggered on demand. In this case, the temporal gating is not needed. The randomness can then be overcome by monitoring the photons which were filtered out spectrally. The absence of spectrally filtered photons should in principle herald presence of an entangled pair. This monitoring will not demolish the entangled pair.

By reducing the detuning[18] and/or by increasing the radiative width through the Purcell effect,[8] entanglement to a useful degree may be achieved even without spectral filtering.

ACKNOWLEDGMENT

We acknowledge the Israel Science and the U.S.-Israel Binational Science Foundations.

[1]J. S. Bell, Rev. Mod. Phys. **38**, 447 (1966).
[2]J. F. Clauser *et al.*, Phys. Rev. Lett. **24**, 549 (1970).
[3]A. Ekert, Phys. Rev. Lett. **67**, 661 (1991).
[4]C. H. Bennett *et al.*, Phys. Rev. Lett. **70**, 1895 (1993).
[5]D. F. V. James *et al.*, Phys. Rev. A **64**, 052312 (2001).
[6]K. Edamatsu, G. Ohata, R. Shimizu, and T. Itoh, Nature **431**, 167 (2004).
[7]P. Michler *et al.*, Science **290**, 2282 (2000).
[8]C. Santori *et al.*, Nature **419**, 594 (2002).
[9]Z. Yuan *et al.*, Science **295**, 102 (2001).
[10]D. Loss and D. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
[11]O. Benson *et al.*, Phys. Rev. Lett. **84**, 2513 (2000).
[12]D. Fattal *et al.*, Phys. Rev. Lett. **92**, 037903 (2004).
[13]N. Akopian *et al.*, Phys. Rev. Lett. **96**, 130501 (2006).
[14]R. M. Stevenson *et al.*, Nature **439**, 179 (2006).
[15]A. Gilchrist, K. J. Resch, and A. G. White, Nature (to be published), DOI: 10.1038/nature05546 (2007).
[16]E. Moreau *et al.*, Phys. Rev. Lett. **87**, 183601 (2001).
[17]D. V. Regelman *et al.*, Phys. Rev. Lett. **87**, 257401 (2001).
[18]M. Bayer *et al.*, Phys. Rev. B **65**, 195315 (2002).
[19]T. M. Stace *et al.*, Phys. Rev. B **67**, 085317 (2003).
[20]G. Ramon *et al.*, Phys. Rev. B **73**, 205330 (2006).
[21]S. Alon-Braitbart *et al.*, Physica E (Amsterdam) **32**, 127 (2006).
[22]C. Santori *et al.*, Phys. Rev. B **66**, 045308 (2002); S. M. Ulrich *et al.*, Appl. Phys. Lett. **83**, 1848 (2003).
[23]A. Peres, Phys. Rev. Lett. **77**, 1413 (1996).