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Generation of single photons and correlated photon pairs using InAs quantum dots

C. Santori[1,2], **D. Fattal**[1], **J. Vuckovic**[1], **G. S. Solomon**[1], and **Y. Yamamoto**[1,3,*]

[1] E. L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA
[2] Institute of Industrial Science, University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8904, Japan
[3] National Institute of Informatics, Tokyo, Japan

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This article reviews recent work on non-classical light generation using semiconductor quantum dots. Experimental results for single-photon generation are presented, including suppression of the two-photon probability, coherence properties and two-photon interference. An experiment demonstrating generation of polarization-correlated photon pairs from biexciton recombination is also reviewed.

1 Introduction

High-quality single-photon sources and sources of entangled photons are needed for proposed applications in quantum information such as quantum cryptography [1], quantum networking [2] and linear-optical quantum computation [3]. Many recent experiments have shown that single quantum emitters are promising candidates for single-photon devices. Semiconductor quantum dots (QDs) [4] offer several possible advantages, including large dipole moments, narrow linewidths that can be close to Fourier-transform-limited, and the possibility of integration into monolithic optical microcavity structures [5–10]. The main difficulty with QDs is that they interact with a solid-state environment, necessitating cryogenic operation temperatures, and yet environment-induced decoherence is still a problem.

Nevertheless QDs have already shown promise as practical single-photon sources suitable for two-photon interference applications [11], and may also be useful as sources of polarization-entangled photons pairs. This article reviews work performed in our group on both of these applications. The first section presents experimental results on single-photon generation using quantum dots embedded in pillar microcavities. The second section describes experiments demonstrating polarization-correlated photon pairs produced from a biexciton cascade, and discusses the difficulty of obtaining polarization entanglement by this method.

2 Single-photon generation

An ideal single-photon source produces exactly one photon in a definite quantum state. In contrast, for a "classical" source such as laser pulses attenuated by a linear filter, the photon number typically follows a Poisson distribution. One method to suppress photon number fluctuations is to use instead a nonlinear filter, such as a single quantum emitter. This emitter ideally absorbs only one photon from an excitation pulse and emits a new photon at a different frequency.

Our device consists of a single QD embedded in an optical microcavity, designed to enhance the spontaneous emission properties of the QD through the Purcell effect [12]. The structure is shown schematically

* Corresponding author E-mail: yamamoto@loki.stanford.edu

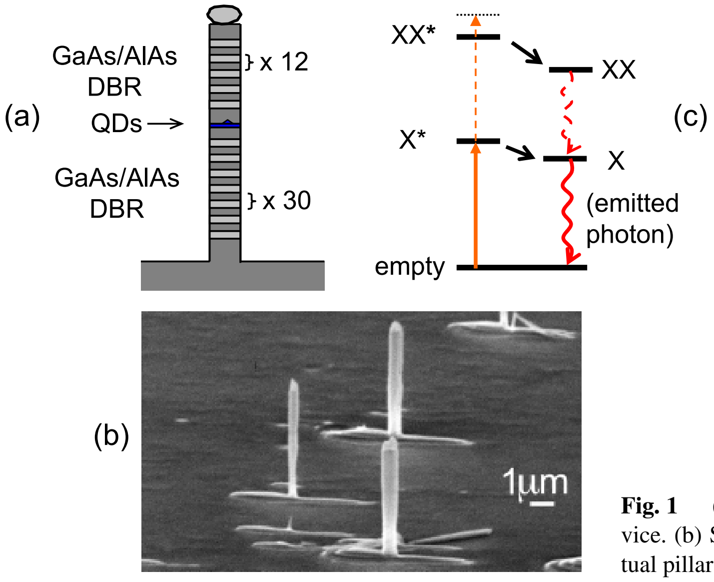

Fig. 1 (a) Schematic diagram of single-photon device. (b) Scanning-electron microscope image of actual pillar structures. (c) Optical excitation scheme.

in Fig. 1a. One or more InAs quantum dots, surrounded by a GaAs matrix, are sandwiched between two distributed-Bragg-reflector (DBR) mirrors, formed from alternating layers of GaAs and AlAs. The quantum dots and DBR mirrors were grown together by molecular-beam epitaxy (MBE). The quantum-dot layer had a density of approximately 25 μm^{-2}, and this layer was centered within a GaAs spacer layer one optical wavelength thick between the DBR mirrors. Other details of the structure are given elsewhere [10,11]. The wafer was etched to form micropillars, about 5 µm tall, using sapphire dust particles as etch masks. This produced pillars with a random distribution of sizes, shapes and locations, as seen in Fig. 1b. The resulting three-dimensional optical microcavities had measured quality factors as large as 1270 and mode volumes of only a few cubic wavelengths. For a quantum dot on resonance with such a cavity, the spontaneous emission rate was observed to increase by a factor as large 5.

The operation scheme is shown in Fig. 1c. A short (2–3 ps) optical pulse generated by a tunable Ti-sapphire laser raises the quantum dot into an excited state containing one electron-hole pair. The quantum dot quickly relaxes with a timescale on the order of 10 ps to its lowest single-exciton state. This state decays through a much slower spontaneous emission process (100–300 ps) to emit a single photon. The spontaneous emission from the single-exciton spectral line is collected and sent through a narrow-band (0.1 nm) filter. This protects against events in which the quantum dot receives multiple excitations. In these events, multiple photons are emitted, but each photon has a unique wavelength, as a result of the electrostatic interactions between particles inside the quantum dot [13]. The sample was cooled to temperatures ranging from 3 to 10 K using a liquid-helium continuous-flow cryostat.

It is convenient to describe the performance of a single-photon device by three parameters: the probability of emitting two photons in the same pulse normalized by the two-photon probability for a Poisson distribution with the same mean ($g^{(2)}$), the mean-squared overlap between the wave-packets of two different photons emitted by the device (a measure of photon state purity), and the efficiency.

2.1 Two-photon suppression

Two-photon suppression is typically measured with a "Hanbury Brown and Twiss"-type setup. The collected photoluminescence is spectrally filtered to select a single emission line, and then sent to a beamsplitter, with each output measured by a photon counter (avalanche photodiode). Coincidence-counting electronics generate a histogram of the relative delay $\tau = t_2 - t_1$ between photon detection events at counters 1 and 2. Such a histogram, obtained from one of our best devices, is shown in Fig. 2. Counts at $\tau = 0$ correspond

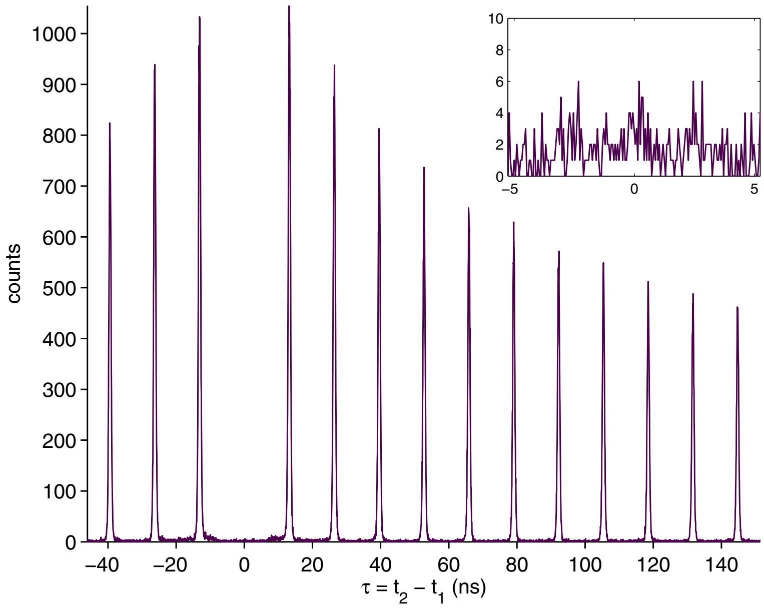

Fig. 2 Photon correlation ($g^{(2)}$) data from a single-photon device, showing a histogram of the relative delay τ between photon detections at two counters in a Hanbury Brown-Twiss setup.

to events where two photons were detected from the same pulse. Few such events should occur for a good single-photon source. The peaks at $\tau = nT_{\mathrm{rep}}$, where n is a nonzero integer, and $T_{\mathrm{rep}} = 13\,\mathrm{ns}$ is the laser repetition period, correspond to events where one photon was detected from each of two different pulses. These peaks can provide information about long-timescale memory effects in the quantum dot, and are useful for normalization.

The results in Fig. 2 indicate that the two-photon probability for this device was a factor of 40 smaller than for an equivalent Poisson-distributed source [10]. This was computed by comparing the central-peak area with the areas of the side peaks far from $\tau = 0$. No backgrounds were subtracted. Theoretically, the two-photon suppression from this incoherent excitation scheme should be even larger, since in the ideal case, a two-photon event can occur only if the quantum dot is excited once, emits a photon, and is excited again, all within a 2 ps excitation pulse. In practice, the observed two-photon suppression is probably limited by weak, broadband background emitted from the sample. Another interesting feature is the increased size of the innermost side peaks near $\tau = 0$. This indicates a "bunching" or "blinking" behavior with a timescale on the order of 50 ns. We have studied these long-term memory effects in more detail in [14]. This behavior likely originates from charge fluctuation within the quantum dot. Such blinking behavior is unwanted in a single-photon devices, since it decreases the efficiency.

2.2 Coherence properties

The purity of the photon state depends on at least four factors: the relaxation rate from the higher-energy single-exciton state excited initially by the laser to the lowest-energy exciton state, the spontaneous emission rate, the pure dephasing rate caused by interactions with phonons or nearby charge fluctuations, and polarization splitting. A simple way to estimate photon-state purity is to compare the spectral linewidth with the spontaneous-emission lifetime. Fig. 3a shows the time-resolved photoluminescence, measured using a streak-camera system, averaged over many laser excitation pulses for two quantum dots, designated A and

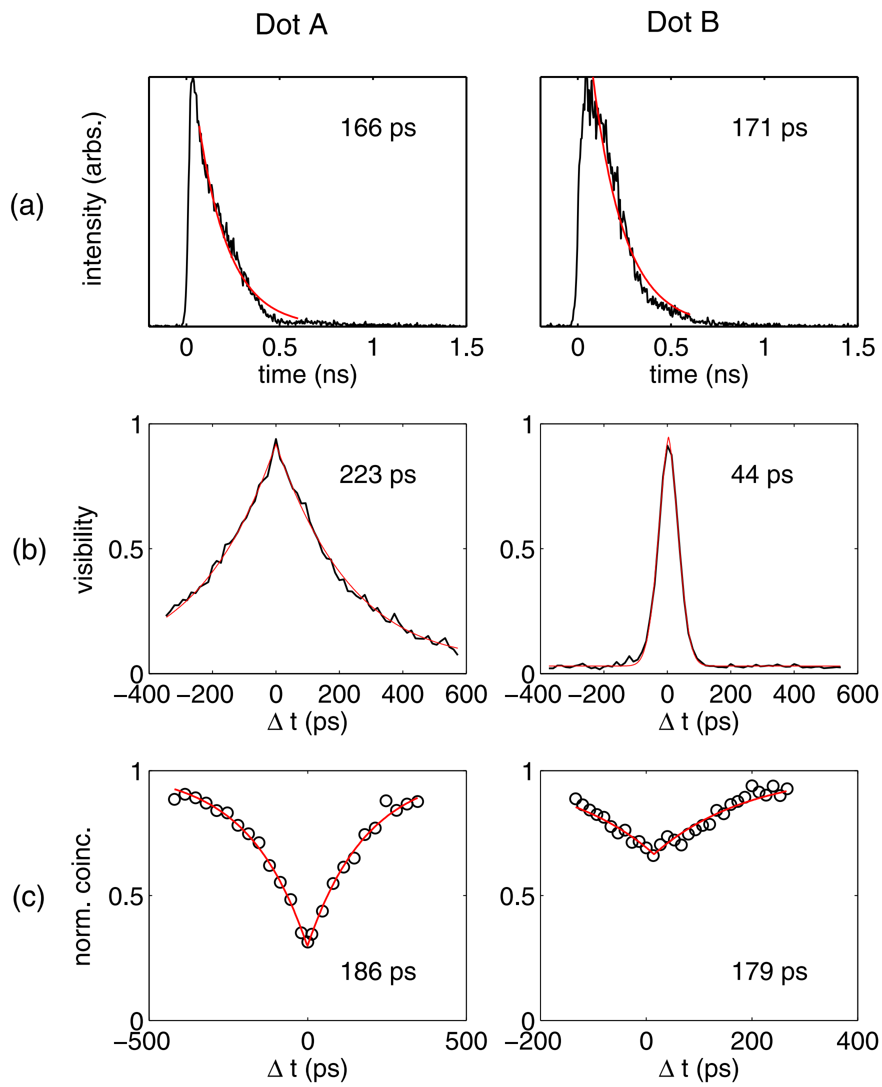

Fig. 3 Measurements on dot A and B to characterize photon state purity: (a) Streak camera measurements, showing mean intensity vs. time following a laser pulse; (b) Interferometer measurements, showing fringe contrast vs. path-length difference; (c) Two-photon interference measurement, showing normalized coincidence rate vs. path-length difference. This normalized rate is approximately twice the probability for two photons that collide at the beamsplitter to exit in different directions.

B, emitting at 932 and 919.5 nm, respectively, and excited by laser pulses tuned to 909 and 905 nm. The spontaneous emission lifetimes τ_s were estimated to be 166 ps and 171 ps, respectively.

We have observed a wide range of emission linewidths for single quantum dots, both when comparing different quantum-dot samples, and when comparing different quantum dots on the same sample. Some emission lines can be too narrow to measure with a typical grating spectrometer, and thus we have used instead a Michelson interferometer setup, where the interference fringe contrast is measured by an avalanche photodiode as the interferometer path-length difference is scanned. The measured curve is approximately proportional to the magnitude of the Fourier transform of the intensity spectrum. Fig. 3b shows that dot B has a broad Gaussian lineshape, while dot A has a narrow, almost Lorentzian lineshape (appearing as a two-sided exponential in the interferometer scan). Dot B is severely broadened; one possible explanation might be interaction with nearby surface charges [15]. For dots A and B, the $1/e$ coherence lengths τ_c were estimated to be 223 ps and 44 ps, respectively. The ratio $2\tau_r/\tau_c$, which equals unity for an ideal spontaneous emission process, is approximately 1.5 and 8 for dots A and B, respectively.

Most quantum-information applications of single-photon sources require pure photon states because they employ two-photon interference in their schemes. A simple example of two-photon interference is the Hong-Ou-Mandel experiment [16, 17]: if two independent photons with identical wavepackets collide at a 50–50 beamsplitter, they always exit together, randomly choosing a side. They never exit in opposite directions. However, if the photons do not have identical wavepackets, they can behave independently, and the two-photon interference effect is reduced. A useful way to characterize photon state purity is to define a two-photon interference visibility, $V_2 = (P_{\text{same}} - P_{\text{opp}})/(P_{\text{same}} + P_{\text{opp}})$, where P_{same} is the probability for two photons to exit in the same direction, and P_{opp} is the probability for them to exit in opposite directions. It can be shown that V_2 is equal to the mean magnitude-squared overlap between the wavepackets of the two photons. A simple model predicts that this overlap should be

$$V_2 = \frac{\Gamma}{\Gamma + 2\alpha} \frac{r}{r + \Gamma} , \tag{1}$$

where Γ is the spontaneous emission rate, α is an additional pure dephasing rate, and r is the relaxation rate between the state initially excited by the laser pulse and the lowest-energy single-exciton state. For $r \to \infty$, this is equal to the inverse of the ratio $2\tau_r/\tau_c$ measured above. It is possible, however, that the mean overlap between two consecutively emitted photons could be larger, if some of the spectral broadening results from a slow spectral-diffusion process.

As described in [11], we tested this experimentally by exciting the quantum dot twice by laser pulses 2 ns apart and arranging for the photons emitted in response to collide sometimes at a beamsplitter. The beamsplitter outputs were measured with photon counters. The probability of two photons that meet at the beamsplitter to exit in opposite directions was estimated from measured coincidence rates. A small delay was also added which allowed the overlap between the photons to be adjusted. The results for dots A and B are shown in Fig. 3c. The results indicate maximum overlaps of 0.70 and 0.33 for dots A and B, respectively. These increase to 0.81 and 0.38 after correcting for known imperfections in the setup. These values are larger than the inverses of the $2\tau_r/\tau_c$ ratios given above. The widths of the coincidence dips, estimated by fitting a simple function of the form $1 - a\exp(-|\Delta t|/\tau_m)$, are 186 ps and 179 ps for dots A and B, respectively. These are close to the measured spontaneous emission decay rates, as expected theoretically. Measurements on other dots having different spontaneous emission lifetimes were reported in [11].

2.3 Efficiency

Efficiency remains a difficulty for this single-photon source. Observed Purcell effects suggest that most of the spontaneous emission is coupled to a single cavity mode, but this does not guarantee that when light escapes from the cavity, it will be efficiently collected. Measurements have indicated that the total efficiency for such devices, measured after the first collection lens, can be as high as 8%, and it has been inferred from simulations that the efficiency of light to escape from the top of the pillar can be higher [9]. Perhaps the efficiency could be improved with refinements to the fabrication process. However, some of the inefficiency must be unrelated to the collection issue. For example, the blinking effect noted in Fig. 2 shows that the single-exciton state is not deterministically prepared. Furthermore, for most applciations, a specific polarization is needed. If the emission polarization is random, polarization filtering results in additional loss.

2.4 Further improvements

Finding devices with small $g_0^{(2)}$ values is not difficult, but the efficiency and photon state purity depend critically on the microcavity characteristics, and on non-ideal aspects of quantum dots. Both the efficiency and photon state purity could potentially be increased by improving the optical cavities. Photonic-bandgap cavities could be used, for example [18]. However, eq. (1) suggests a limit on how much the spontaneous emission rate Γ should be increased through the Purcell effect. The two-photon interference visibility reaches a maximum when $\Gamma = \sqrt{2\alpha r}$. When Γ is increased further, the photon wavepackets become too short relative to the time uncertainty associated with the initial relaxation into the lowest-energy single-exciton state. For reasonable parameter values $1/\alpha = 1\,\text{ns}$ and $1/r = 10\,\text{ps}$, this gives $\tau_r = 1/(\Gamma_{\max}) \approx 70\,\text{ps}$. This is not much shorter than the shortest lifetimes already observed with the present devices. Therefore, the benefits of improved cavities will be lost unless the time uncertainty in the excitation process can be reduced. This time uncertainty is a result of the incoherent excitation scheme we have adopted. This scheme was chosen for its experimental simplicity. Eventually, however, one must consider coherent excitation schemes, such as vacuum-stimulated Raman processes [19], that have been studied in the development of atomic single-photon sources. Such schemes might be possible with quantum dots, as well [20].

3 Photon pair generation

We have recently demonstrated that linear-optical techniques and photodetection can be applied to a single-photon source to observe polarization entanglement [21]. However, it may also be possible for a quantum dot to emit a pair of entangled photons without the need for post-selection.

In [22], it was suggested that the two-photon cascade that results from the decay of a biexciton state (two electrons and two holes) could produce polarization-entangled photon pairs. This would be similar to the entanglement observed from two-photon cascades in atoms. Pure polarization entanglement can occur only if the intermediate single-exciton states are nearly degenerate (the splitting is much smaller than the natural linewidth), so that the two decay paths cannot be distinguished based on the photon energies. The exciton spin decoherence rate must also be small compared with the radiative decay rate.

Here, we describe an experiment demonstrating polarization correlation in a prefered linear basis, but not entanglement, for photon pairs produced through a biexciton cascade [23]. We then discuss why entanglement was not observed, and possible remedies.

For this experiment, quantum dots in simple mesa structures were used. Fig. 4a shows a spectrum from a quantum dot designated dot N. Many spectral lines appear, corresponding to various multiparticle configurations. As described in [24], photon-counting cross-correlation measurements can help in identifying these lines. As in the usual Hanbury Brown-Twiss setup, the collected photoluminescence is sent through a beamsplitter, but in this case each output has an independently tunable spectral filter. If both filters are tuned to the single-exciton line (X), as in Fig. 4b, continuous-wave laser excitation produces a symmetric "antibunching" dip in the coincidence rate, centered at zero delay ($\tau = 0$). However, as shown in Fig. 4c and d, if peaks C' or C'' are used as the "start" triggers, while peak X is used again as the "stop" trigger, an asymmetric dip appears near $\tau = 0$, consistent with the two states having different total charge [24]. In Fig. 4e, the biexciton peak XX was used as the "start" trigger, and X as "stop". In this case, a large correlation peak is seen near $\tau = 0$, the signature of a two-photon cascade. This correlation peak becomes larger relative to the uncorrelated floor far from $\tau = 0$ as the excitation power is decreased, until it approaches a maximum value limited by background and dark counts.

To make a deterministic source of entangled photon pairs, it would be necessary to deterministically excite a biexciton state containing two electrons with opposite spin, and two heavy holes (spin $\pm\frac{3}{2}$) with opposite spin. In the experiment described next, we produce this biexciton state with a low efficiency, but in those cases where a biexciton is produced, we can test whether the emitted photons have polarization correlations or entanglement. This experiment was performed using pulsed excitation, with the laser frequency tuned above the GaAs bandgap (710 nm).

Fig. 5 shows cross-correlation histograms measured between the biexciton and single-exciton spectral lines for polarization combinations in a special linear basis, rotated 18 $^\circ$ relative to the wafer axes. The H and V intensities were unequal, but the integration times were adjusted to obtain approximately the same

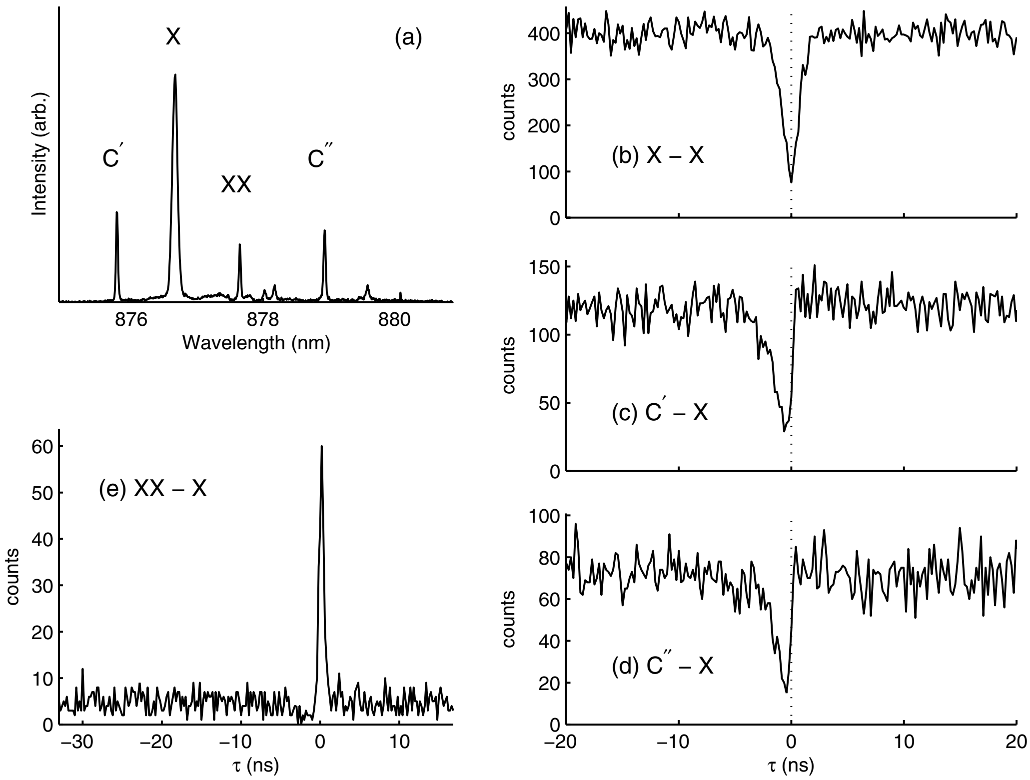

Fig. 4 (a) Photoluminescence spectrum of dot N under continuous-wave, above-band excitation. (b) Cross-correlation histogram, with start/stop counters detecting peaks X/X; (c) detecting peaks C'/X; (d) detecting peaks C''/X; (e) detecting peaks XX/X.

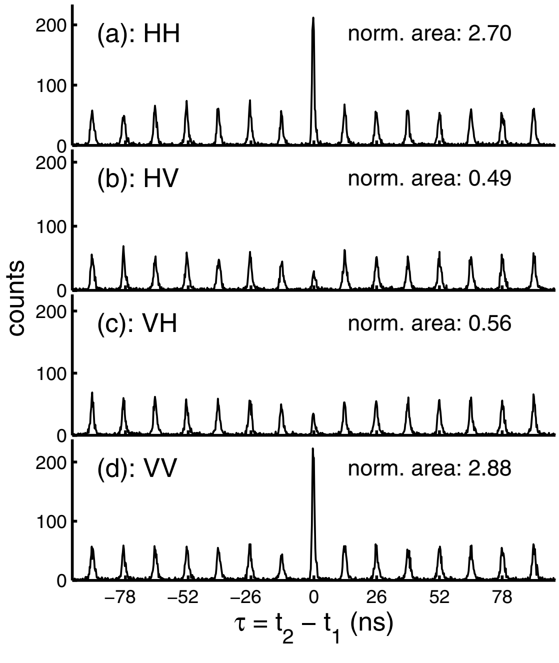

Fig. 5 Cross-correlation histograms under pulsed, above-band excitation, with start/stop counters detecting peaks XX/X, with detection polarizations indicated in the figures. H and V are linear polarizations in a basis rotated 18° degrees relative to the wafer cleave directions, (110) or (1–10).

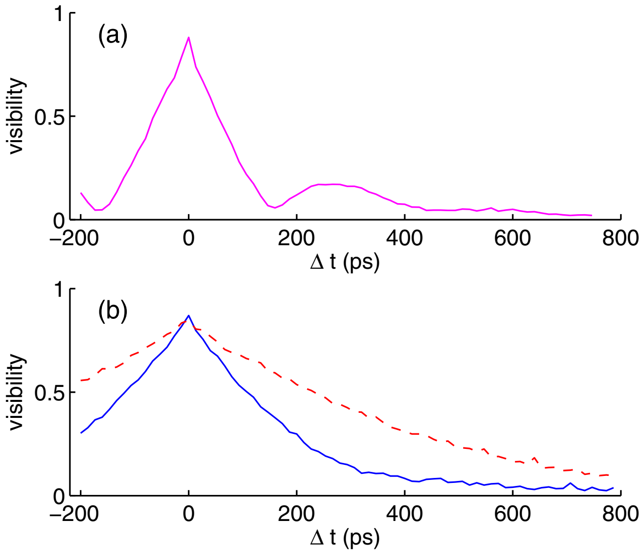

Fig. 6 Measurements with a Michelson-type interferometer, showing fringe contrast vs. path-length difference: (a) dot A with both polarizations detected; (b) dot A with orthogonal linear polarizations designated H' (solid) and V' (dashed) detected separately.

number of uncorrelated coincidences away from $\tau = 0$ in the four cases. Subject to this normalization, it is clear that the correlation peak near $\tau = 0$ is much larger when the two detection polarizations are parallel, than when they are crossed. As described in [23], this polarization correlation is relatively independent of excitation power.

If we could measure such polarization correlations in other bases, such as 45/-45 or right/left circular, this would demonstrate entanglement, but these correlations are absent in the other bases. The existence of a preferred linear basis is evidence of quantum-dot asymmetry. It is known that, for asymmetric quantum dots, the electron-hole exchange interaction lifts the degeneracy of the two bright-exciton states [25]. If this splitting is much larger than the radiative decay rate, then polarization entanglement cannot be observed unless one measures a time window much shorter than the spontaneous emission lifetime. For the particular quantum dot studied here, the spectral line is much broader than the natural linewidth, and it is impossible to determine if such a splitting exists. However, we have more recently observed splittings on the order of 10 μeV for emission lines of quantum dots in microcavities. A splitting is demonstrated in Fig. 6 using a Michelson interferometer to resolve a doublet. Such splittings appear common, and may be another useful way to distinguish neutral-exciton emission from emission from singly-charged exciton states, which must have a two-fold Kramers degeneracy. It is still possible that entanglement might be observed if the single-exciton splitting were removed, through application of a magnetic field parallel to one of the asymmetry axes of the quantum dot, or through application of external strain. Such experiments will hopefully clarify whether biexciton cascades can ever produce entangled photons, and how important decoherence is for this scheme.

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