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Received 2 Jul 2014 | Accepted 18 Sep 2014 | Published 31 Oct 2014

DOI: 10.1038/ncomms6298

OPEN

Observation of strongly entangled photon pairs from a nanowire quantum dot

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A bright photon source that combines high-fidelity entanglement, on-demand generation, high extraction efficiency, directional and coherent emission, as well as position control at the nanoscale is required for implementing ambitious schemes in quantum information processing, such as that of a quantum repeater. Still, all of these properties have not yet been achieved in a single device. Semiconductor quantum dots embedded in nanowire waveguides potentially satisfy all of these requirements; however, although theoretically predicted, entanglement has not yet been demonstrated for a nanowire quantum dot. Here, we demonstrate a bright and coherent source of strongly entangled photon pairs from a position-controlled nanowire quantum dot with a fidelity as high as 0.859 ± 0.006 and concurrence of 0.80 ± 0.02 . The two-photon quantum state is modified via the nanowire shape. Our new nanoscale entangled photon source can be integrated at desired positions in a quantum photonic circuit, single-electron devices and light-emitting diodes.

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There are demanding requirements for an ‘ideal’ entangled-photon source for implementing ambitious schemes in quantum information processing, such as that of a quantum repeater¹. The source should meet the following criteria: high brightness combined with high-fidelity entanglement², on-demand generation³, high extraction efficiency⁴, directional⁵ and coherent emission⁶, as well as position control at the nanoscale⁷. It is extremely difficult to meet all of these requirements in a single device. Good candidates are semiconductor quantum dots embedded in nanowires.

The high refractive index of a nanowire waveguide around a quantum dot ensures that the emitted light is guided in the desired direction and a tapered end makes the light extraction very efficient⁴. With such a design, efficient single-photon generation has been demonstrated from a single nanowire quantum dot⁵. In addition, the emission mode-profile was shown to be directional and Gaussian^{8,9}, a key requirement for efficient long-distance quantum communication in well-established telecommunication technology. Nanowires can be controllably positioned in uniform arrays^{10,11}, with the ability to independently control the dot size and waveguide shell around it¹². Silicon segments and substrates can be included in the design^{13–16} and electrical contacts have been demonstrated on single nanowires for single-electron devices¹⁷, light-emitting diodes¹⁸, as well as single-photon avalanche photodiodes¹⁹. A significant advantage of using nanowire waveguides for efficient light extraction over other existing approaches, such as optical microcavities²⁰, is the broad frequency bandwidth of operation²¹, which is needed for achieving bright entangled photon-pair generation via the biexciton–exciton radiative cascade. This approach is especially advantageous for quantum dots emitting over a large spectral range and may also be implemented with II–VI quantum dots where the biexciton binding energy is very large (> 20 meV)²².

A key feature of nanowires with embedded quantum dots grown in the [111]-direction is that the fine-structure splitting is expected to vanish²³, which should result in excellent entangled photon emission via the biexciton–exciton radiative cascade²⁴. Our measurements realize this prediction and demonstrate the generation of strongly entangled photon pairs for the first time from a nanowire quantum dot. Our sources are ready to implement in advanced quantum information processing schemes without the need for any post-growth manipulation²⁵ or temporal post-selection²⁶. Temporal post-selection can be a major source of photon losses and puts additional requirements on the measurement, thus limiting the scalability of quantum dot-based entangled photon sources. For practical applications it is therefore very useful that we can avoid temporal post-selection. Finally, due to the efficient waveguiding and the tapered end, which we created during the bottom-up growth of the nanowire, we measure a light extraction efficiency of $18 \pm 3\%$ for the source. Importantly, due to a recent breakthrough in the nanowire growth¹², this high efficiency is obtained while potentially meeting all of the criteria of an ideal entangled photon source.

Results

Site-controlled quantum dots in tapered nanowire waveguides.

The nanowires were grown by selective-area chemical beam epitaxy, which allows for control of the dot size and position, as well as enabling growth of the waveguide shell around the dot for efficient light extraction (see Methods). This technique has been demonstrated to yield defect-free, pure wurtzite nanowires, which is essential to obtain long single-photon coherence²⁷. Figure 1a shows a scanning electron microscopy (SEM) image of a tapered InP nanowire waveguide containing an InAsP segment, 200 nm

from the nanowire base, defining the optically active quantum dot that we study.

A spectrum taken under the excitation condition used for the quantum-state tomography measurements is depicted in Fig. 1b. By performing cross-correlation measurements²⁸, shown in the inset, and power-dependent measurements (see Supplementary Fig. 1), we identified the biexciton (XX) and exciton (X_A and X_B) transitions. The XX – X_B cascade produces entangled photons. In contrast, a weak cascade is observed for XX – X_A , which does not show entanglement. From these observations, X_A could be either a charged exciton or an exciton with a different hole state than X_B as is permitted by the wurtzite crystal structure. The transitions XX and X_B are resolution limited; single-photon interference measurements show excellent coherence of our entangled photons (see Supplementary Fig. 2 and Supplementary Note 1). Autocorrelation measurements at saturation of the XX and X_B transitions show strong antibunching, indicative of nearly perfect single-photon pairs from the XX – X_B cascade (see Supplementary Fig. 3).

Light extraction efficiency. From the single-photon detector counts, 55 kilocounts per second for XX and 15 kilocounts per second for X_B , under pulsed excitation at 80 MHz, we calculate a collection of 7.9 million XX and 2.0 million X_B photons per second into the first objective when taking our $0.7 \pm 0.1\%$ setup efficiency at ~ 900 nm into account. The X_A transition shows the highest intensity of 12.1 million photons per second and saturates our spectrometer’s CCD camera with integration times as short as 1 s under the excitation conditions used throughout our study (Fig. 1b). This radiative recombination pathway from XX competes strongly with the X_B emission, thus reducing the entangled photon-pair generation efficiency. Taking into account this competing recombination pathway for XX , we calculate a light extraction efficiency of $18 \pm 3\%$ for the source. We expect that adding a gold mirror with thin dielectric layer below the nanowire will boost the efficiency nearly twofold²¹. Combining this mirror with further engineering of the nanowire shape promises extraction efficiencies exceeding 90% (ref. 29).

Low fine-structure-splitting system. Using polarization-dependent measurements, presented in Fig. 1c, we obtain an estimation for the excitonic fine-structure splitting, S , by subtracting the XX transition from the X_B transition energy³⁰. We obtain from the sine-function fit a fine-structure splitting of $1.2 \mu\text{eV}$. In the case of nanowires, the small fine-structure splitting is a result of growth on a [111]-oriented substrate and the symmetric hexagonal cross-section of the nanowire core, defining the quantum dot. This small value that we measure for the fine-structure splitting is crucial for the entanglement observation between XX and X_B photons without any temporal post-selection and is representative of the sample where, remarkably, a high percentage ($> 50\%$) of the measured quantum dots show a fine-structure splitting below $2 \mu\text{eV}$ (Supplementary Fig. 4). The period of precession of the X_B spin can be estimated as $h/S = 3.5$ ns (ref. 31), where h is Planck’s constant. This period of precession is much longer than the X_B lifetime of 0.50 ± 0.01 ns as extracted from the XX – X_B cross-correlation measurements without polarization selection (Supplementary Fig. 5). Therefore, the X_B spin precession has only little influence on the correlations in polarization between the two photons.

Polarization-entangled photon pairs. Twelve cross-correlation measurements in the rectilinear, diagonal and circular polarization bases are shown in Fig. 2, where each histogram is composed of 64 ps time bins. In the correlation measurements, the first letter stands for the measured polarization of the XX photon, whereas

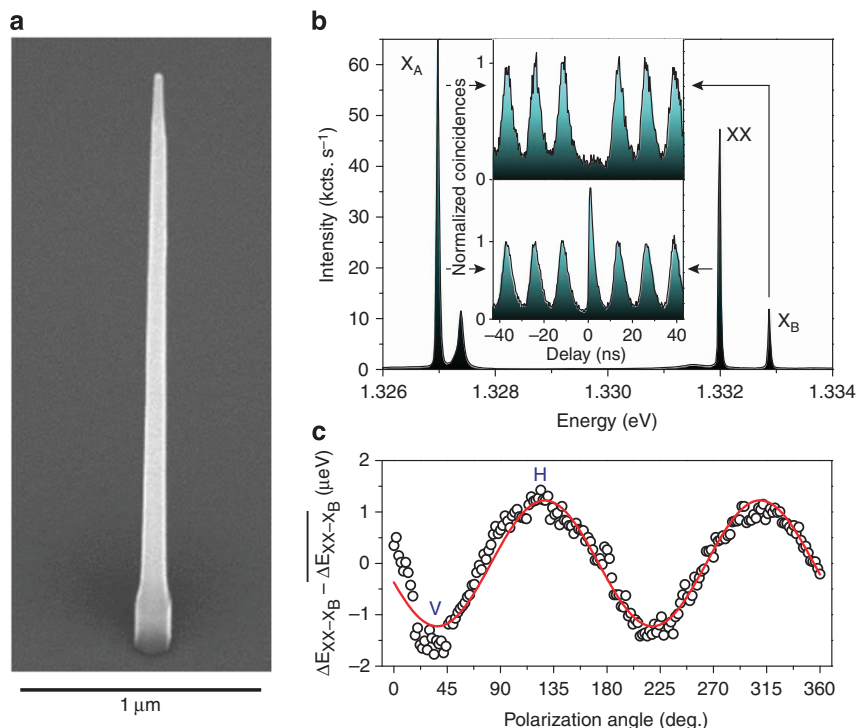


Figure 1 | Nanowire quantum dot sample. (a) Scanning electron microscopy image of a tapered nanowire waveguide with embedded quantum dot. (b) Photoluminescence spectrum of a single InAsP quantum dot embedded in an InP nanowire. The spectrum was taken at the excitation power used for the cross-correlation measurements needed to reconstruct the density matrix (100 nW), which is close to saturation of both XX and X_B transitions. Note that the excitonic transition X_A saturates the CCD camera. (c) Polarization-dependent measurement to determine the excitonic fine-structure splitting. To increase the accuracy of the polarization measurement we plot the relative difference between biexciton XX and exciton X_B emission energy. The amplitude of the sine-function fit indicates a fine-structure splitting of 1.2 μeV.

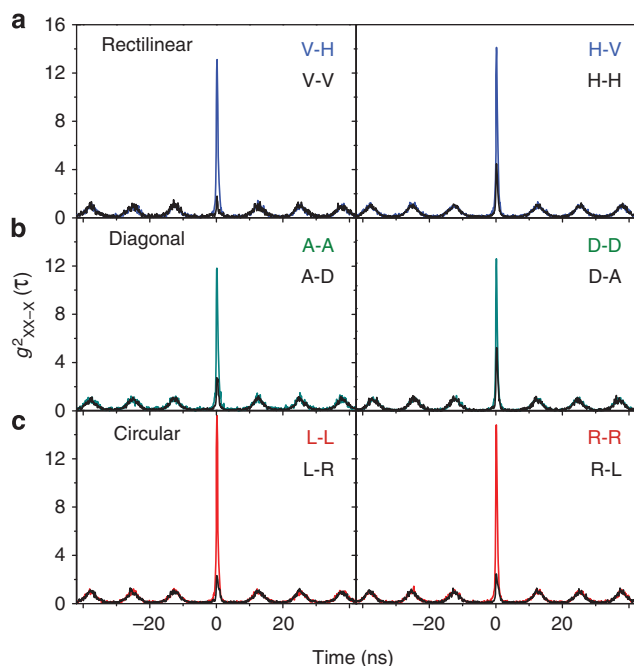


Figure 2 | Cross-correlation measurements for the three different bases: (a) Rectilinear, (b) diagonal and (c) circular. The plotted data are normalized to the Poisson level of the side peaks. Start: biexciton; stop: exciton X_B. The first letter stands for the measured polarization of the biexciton photon, whereas the second letter stands for the measured polarization of the exciton photon.

the second letter stands for the X_B photon. The strong correlations in HV, VH, DD, AA, RR and LL, together with the weak correlations in HH, VV, DA, AD, RL and LR, show that the two photons from the XX–X_B cascade are entangled. Here, H and V are orthogonal linear polarizations (horizontal and vertical), $D = (H + V)/\sqrt{2}$ and $A = (H - V)/\sqrt{2}$ are diagonal and anti-diagonal linear polarizations, whereas $R = (H + iV)/\sqrt{2}$ and $L = (H - iV)/\sqrt{2}$ are righthanded and lefthanded circular polarizations.

The quantum state we observe is different from the state that is measured for self-assembled quantum dots^{7,25,32–35}. Typically, one measures for the XX–X cascade bunching (positive correlations) in HH and RL, and antibunching (negative correlations) in HV and RR. However, we observe the opposite (Fig. 2a,c). Only in the diagonal basis we see the usual correlations: bunching in DD and antibunching in DA. These results show that the two-photon quantum state is closer to $(|HV\rangle + |VH\rangle)/\sqrt{2}$ than to the commonly measured state $(|HH\rangle + |VV\rangle)/\sqrt{2}$ (refs 7,25,32–35).

Quantum-state tomography. We performed quantum-state tomography³⁶ to determine more precisely the quantum state of the photons and the degree of entanglement. The raw cross-correlation measurements needed to reconstruct the density matrix are shown in Supplementary Fig. 6. The resulting density matrix is given in Fig. 3a,b. The concurrence is 0.57 ± 0.02 . A positive value for the concurrence means that the correlations cannot be explained classically and that the photons are quantum entangled. In this calculation, all correlation counts in the full time window of 6.02 ns are taken into account. The two-photon

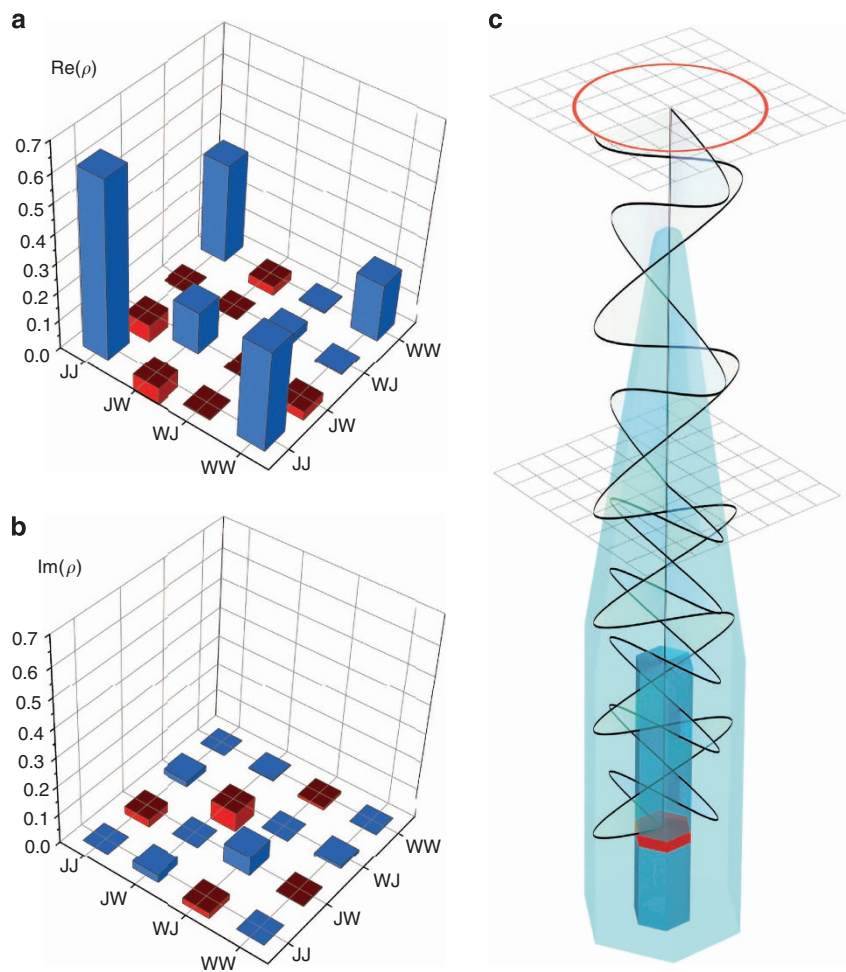


Figure 3 | Quantum-state tomography. Real (a) and imaginary part (b) of the density matrix for the full time window of 6 ns, in the rotated basis. The positive matrix elements are blue, and the negative matrix elements are red. (c) Illustration of the effect of birefringence in the nanowire. The orthogonal waves inside the nanowire experience different refractive indices, and therefore their wavelengths inside the waveguide are unequal. As a result, the polarization of the light emission by the quantum dot (red) is modified leading to a different quantum state. The tapered section of the nanowire is more symmetric and is free of birefringence.

Table 1 Calculated concurrences and the fidelities for five different time windows.			
Time window (% of coincidence counts)	Concurrence	Fidelity to $(JJ\rangle + WW\rangle)/\sqrt{2}$	Fidelity to general maximally entangled state
6.02 ns (100)	0.57 ± 0.02	0.762 ± 0.002 ($\alpha = 33^\circ$, $\beta = 46^\circ$)	0.765 ± 0.002
1.40 ns (80)	0.64 ± 0.02	0.793 ± 0.002 ($\alpha = 33^\circ$, $\beta = 47^\circ$)	0.796 ± 0.002
0.70 ns (59)	0.69 ± 0.02	0.814 ± 0.003 ($\alpha = 34^\circ$, $\beta = 48^\circ$)	0.818 ± 0.003
0.38 ns (35)	0.73 ± 0.02	0.839 ± 0.003 ($\alpha = 35^\circ$, $\beta = 49^\circ$)	0.842 ± 0.003
0.13 ns (10)	0.80 ± 0.02	0.854 ± 0.006 ($\alpha = 36^\circ$, $\beta = 50^\circ$)	0.859 ± 0.006

The fraction of the correlation events taken into account for a certain time window are given in brackets in the first column.

state has a fidelity of 0.762 ± 0.002 to the maximally entangled state $(|JJ\rangle + |WW\rangle)/\sqrt{2}$, where $J = He^{-i\beta} \cos\alpha + Ve^{-i\beta} \sin\alpha$ and $W = -He^{i\beta} \sin\alpha + Ve^{i\beta} \cos\alpha$ are two orthogonal elliptical polarizations. The angles α and β are specified in Table 1. The classical limit is 0.5, so this result shows a strong degree of entanglement, even without temporal selection.

Selection of a narrower time window yields higher values for the concurrence and the fidelity (Table 1). For example, for a time window of 0.13 ns we calculate a concurrence of 0.80 ± 0.02 and a fidelity of 0.854 ± 0.006 . The density matrix for this time window is presented in Supplementary Fig. 7. Temporal selection yields stronger entanglement, because within a narrow time window the

effects of spin precession and dephasing processes are smaller³¹. When we do not restrict our analysis to states of the form $(|JJ\rangle + |WW\rangle)/\sqrt{2}$, but instead calculate the fidelity to a general maximally entangled two-photon state, we find only slightly higher values (Table 1). The maximally entangled states to which the fidelity is maximal are very close to states of the form $(|JJ\rangle + |WW\rangle)/\sqrt{2}$.

Two-photon quantum state modified by birefringence. Why do we measure $(|JJ\rangle + |WW\rangle)/\sqrt{2}$ and not the usual two-photon state that is measured for quantum dots, namely

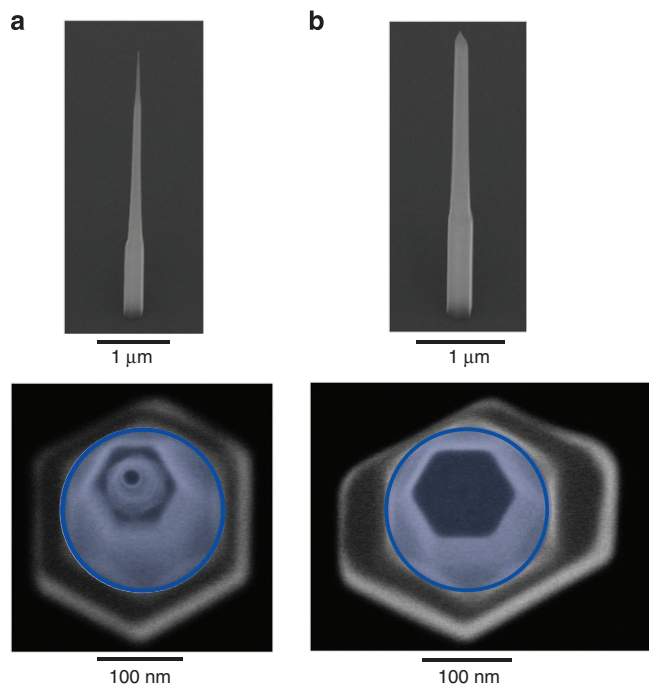


Figure 4 | Nanowire birefringence. SEM images of: (a) symmetric nanowire waveguide, and (b) asymmetric nanowire waveguide. Top panel: side-view SEM images of nanowires with a tilt angle of 45 degrees. Bottom panel: SEM images of the nanowires viewed from the top at a small tilt angle. The blue-shaded circle represents the opening in the SiO₂ mask. The example of the nanowire elongation in **b** is an extreme example that leads to geometric birefringence and corresponding rotation of the quantum state.

$(|HH\rangle + |VV\rangle)/\sqrt{2}$? The most probable reason is that the nanowire has a small anisotropy: it could have a slightly elongated cross-section. An extreme case is shown in the SEM image of Fig. 4b. Such an anisotropy may be formed during the growth of the cladding around the core, and would then be unrelated to the shape of the quantum dot (for details of the growth, see the Methods section). As a comparison, we show a symmetric nanowire waveguide in the SEM image of Fig. 4a. In case of an elongated cross-section the effective refractive indices are different for the polarizations along the short and the long axis of the nanowire. Here, we could imagine that the quantum dot emits photon pairs in the usual entangled quantum state $(|HH\rangle + |VV\rangle)/\sqrt{2}$. As the emitted photons are guided along the nanowire, the two-photon state is modified by birefringence into $(|JJ\rangle + |WW\rangle)/\sqrt{2}$, as is illustrated in Fig. 3c. Thus, HH and VV correlations rotate into predominantly RR and LL correlations, while RL and LR turn mostly into HV and VH , which explains the observations of Fig. 2. For a nanowire waveguide of 6 μm length a difference of effective refractive index of order 0.1 would be enough to explain the magnitude of the observed rotation. Apart from birefringence in the waveguide, the polarization state of the emitted photons could also have been influenced by Γ_7 and Γ_9 hole mixing in the wurtzite quantum dot.

Discussion

In summary, we used a wurtzite nanowire quantum dot to generate single pairs of polarization-entangled photons with a fidelity as high as 0.859 ± 0.006 and a concurrence up to 0.80 ± 0.02 . Furthermore, a high degree of entanglement is maintained (fidelity of 0.762 ± 0.002) without any temporal post-selection. This first observation of entangled photon-pair generation from a nanowire quantum dot, which combines the

desired properties of an ideal entangled photon source, opens new opportunities in quantum optics, integrated quantum photonic circuits^{37,38} and quantum information processing.

To realize an ideal entangled photon source in future work there are several properties of our source to consider. First, quantum dot-entangled photon sources have not yet reached the fidelity or concurrence values of parametric down-conversion sources^{2,39}. However, with recently available post-growth tuning methods to bring the fine-structure splitting of almost any quantum dot near zero²⁵ and two-photon resonant excitation³, the fidelity of these quantum dot sources are approaching that of parametric down-conversion sources. Second, the single-photon coherence of the emitted photon pairs is not yet Fourier-transform limited, which is needed for advanced quantum information-processing schemes. Such Fourier-transform-limited photons may be reached by combining two-photon resonant excitation techniques^{3,40}, cooling of the quantum dot sample to 300 mK²⁸ and by accelerating the quantum dot emission via the Purcell effect²⁰. Finally, the major advantage of tapered nanowire waveguides over other approaches is the light extraction efficiency, which promises entangled photon-pair extraction efficiencies exceeding 90% due to the broadband frequency of operation²⁹. Such efficiencies would surpass the state-of-the-art entangled photon-pair efficiency of 12%²⁰, without the stringent requirements needed to engineer both the exciton and biexciton into resonance with a cavity mode by using post-growth manipulation of pre-selected quantum dots.

Note: Similar work is reported by Huber *et al.*²⁶

Methods

Nanowire quantum dot growth. The InP nanowires containing single InAsP quantum dots were grown using chemical beam epitaxy with trimethylindium and pre-cracked PH₃ and AsH₃ sources. The nanowires were grown on a SiO₂-patterned (111)B InP substrate consisting of circular holes opened up in the oxide mask using electron-beam lithography and a hydrofluoric acid wet-etch. Au was deposited in these holes using a self-aligned lift-off process, which allows the nanowires to be positioned at known locations on the substrate⁴¹. The thickness of the deposited gold is chosen to give 20-nm to 40-nm diameter particles, depending on the size of the hole opening. The nanowires were grown at 420 °C with a trimethylindium flux equivalent to that used for a planar InP growth rate of $0.1 \mu\text{m h}^{-1}$ on (001) InP substrates at a temperature of 500 °C. The growth is a two-step process: (i) growth of a nanowire core containing the quantum dot, nominally 200 nm from the nanowire base, and (ii) cladding of the core to realize nanowire diameters for efficient light extraction (around 200 nm). The quantum dot diameters are determined by the size of the nanowire core. In this study, we investigated quantum dot diameters ranging from ~25 to 30 nm.

The nanowire core was grown for 26 min at a PH₃ flow of 3 s.c.c.m. The dot was incorporated by switching from a PH₃ to an AsH₃ overpressure for 3 s after 15 min of growth. This growth time results in a quantum dot height of ~6 nm as determined in our previous studies¹², using an energy-dispersive X-ray spectroscopy line scan along the nanowire, for a sample with nominally identical growth conditions. We note that our quantum dots are grown during 3 s, resulting in taller quantum dots with longer emission wavelength, as compared with the work of Huber *et al.*²⁶ who used a growth time of 2 s. Our growth conditions result in very small fine-structure splittings as shown in Supplementary Fig. 4.

The nanowire cladding was grown by increasing the PH₃ flow to 9 s.c.c.m. The total growth time was 120 min. To realize the smooth tapering towards the tip, the nanowire was made longer than the diffusion length of indium. Most nanowires, including the quantum dots, have a pure wurtzite crystal structure. The nanowire axis is the wurtzite *c* axis.

Nanowire waveguide elongation. Radial growth is nominally constrained by the oxide opening, and the nanowire cross-section has a hexagonal symmetry. Optimal coupling of the quantum dot emission to the waveguide mode requires diameters exceeding that of the oxide opening. This is achieved by increasing the cladding growth time, which results in the nanowire overgrowing the oxide opening. No longer constrained by the opening, the hexagonal symmetry may be distorted (see Fig. 4). This asymmetry results in a geometric birefringence and concomitant rotation of the polarization state emitted by the quantum dot.

Optical measurements. The optical measurements were performed in a standard confocal microscopy setup where the quantum dot sample is cooled to a temperature of $T = 5 \text{ K}$ in a closed-cycle cryostat. The setup consists of two spectrometers both equipped with red-enhanced single-photon avalanche diodes having

75 ps time resolution, dark count rates as low as 80 counts per second and quantum efficiency of 11.5% at 930 nm⁴². A set of waveplates and polarizers placed in front of each spectrometer was used to perform polarization-dependent cross-correlation measurements. One spectrometer is set to the biexciton (XX) transition and the other to the exciton (X_B) transition. Each correlation measurement was done with 6,000 s of integration to reach over 1,000 correlations in each side peak. For all photoluminescence and correlation measurements, we use a Ti:Sapphire laser emitting 3-ps-long pulses at 750 nm with a repetition rate of 80 MHz to excite the quantum dot.

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Acknowledgements

We acknowledge S. Höfling, T. Braun and R. Trotta for scientific discussions, L. Schweickert for the image illustrating the nanowire birefringence. This research was supported by the Dutch Foundation for Fundamental Research on Matter (FOM projectruimte 12PR2994), ERC, and the European Union Seventh Framework Programme 209 (FP7/2007–2013) under Grant Agreement No. 601126 210 (HANAS).

Author contributions

M.E.R., M.A.M.V. and V.Z. conceived and designed the experiments. K.D.J., M.E.R. and M.A.M.V. performed the experiments. D.D. and P.J.P. fabricated the sample. M.A.M.V., M.E.R. and K.D.J. analysed the data. A.Gu. and A.Gi. developed the detectors. M.E.R., M.A.M.V., K.D.J. and V.Z. wrote the manuscript with input from the other authors.

Additional information

Supplementary Information accompanies this paper at <http://www.nature.com/naturecommunications>

Competing financial interests: The authors declare no competing financial interests.

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How to cite this article: Versteegh, M. A. M. *et al.* Observation of strongly entangled photon pairs from a nanowire quantum dot. *Nat. Commun.* **5**:5298 doi: 10.1038/ncomms6298 (2014).



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