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Author(s)	Dorenbos, S. N.; Sasakura, H.; van Kouwen, M. P.; Akopian, N.; Adachi, S.; Namekata, N.; Jo, M.; Motohisa, J.; Kobayashi, Y.; Tomioka, K.; Fukui, T.; Inoue, S.; Kumano, H.; Natarajan, C. M.; Hadfield, R. H.; Zijlstra, T.; Klapwijk, T. M.; Zwiller, V.; Suemune, I.
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Position controlled nanowires for infrared single photon emission

S. N. Dorenbos,^{1,a)} H. Sasakura,^{2,3} M. P. van Kouwen,^{1,4} N. Akopian,¹ S. Adachi,⁵ N. Namekata,⁵ M. Jo,² J. Motohisa,⁴ Y. Kobayashi,⁴ K. Tomioka,^{4,6} T. Fukui,^{4,6} S. Inoue,⁵ H. Kumano,^{2,3} C. M. Natarajan,⁷ R. H. Hadfield,⁷ T. Zijlstra,¹ T. M. Klapwijk,¹ V. Zwiller,¹ and I. Suemune^{2,3}

¹Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands

²Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0021, Japan

³CREST, Japan Science and Technology Agency, Kawaguchi 332-0012, Japan

⁴Graduate School of Information Science Technology, Hokkaido University, Sapporo 060-0814, Japan

⁵Institute of Quantum Science, Nihon University, Tokyo 101-8308, Japan

⁶Research Center for Integrated Quantum Electronics, Hokkaido University, Sapporo 060-8628, Japan

⁷School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh EH14 4AS, United Kingdom

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We report the experimental demonstration of single-photon and cascaded photon pair emission in the infrared, originating from a single InAsP quantum dot embedded in a standing InP nanowire. A regular array of nanowires is fabricated by epitaxial growth on an electron-beam patterned substrate. Photoluminescence spectra taken on single quantum dots show narrow emission lines. Superconducting single photon detectors, which have a higher sensitivity than avalanche photodiodes in the infrared, enable us to measure auto and cross correlations. Clear antibunching is observed [$g^{(2)}(0)=0.12$] and we show a biexciton–exciton cascade, which can be used to create entangled photon pairs. © 2010 American Institute of Physics. [doi:10.1063/1.3506499]

Semiconductor quantum dot (QD) structures are attractive candidates for solid-state single photon and/or entangled-photon pair generation.^{1–3} Nanowire QDs (NW-QDs) are promising candidates for such sources because of the controllability of doping, shape, and material freedom.^{4,5} Fine structure splitting is predicted to be absent, which makes NW-QDs ideal for the creation of entangled photon pairs.⁶ Single photon emission from a NW-QD has been shown at wavelengths shorter than 1000 nm.⁷ However, a single photon NW-QD emitter at telecommunication wavelengths and a detailed study of its emission lines has not been reported, because until recently a single photon detector (SPD), with a high enough signal to noise ratio at infrared wavelengths and an adequate timing resolution was lacking. In this letter, we report on the fabrication and characterization of a regular array of InAsP QD embedded in an InP NW, emitting around 1.3 μm and characterization of the QD photoluminescence (PL) using superconducting SPDs (SSPDs). We demonstrate controlled positioning of the NWs by growing them in a regular array. Control of the position is important for uniform growth, which is necessary for uniform QDs. SSPDs offer single photon detection with low dark counts, excellent timing resolution, and decent efficiency in the infrared, without the need for gating. In addition, SSPDs have very short dead times (10 ns) and no after pulsing. These characteristics enable us to perform auto and cross correlation experiments.

Arrays of InAsP QDs embedded in InP NWs are synthesized by selective area metal organic vapor phase epitaxy (SA-MOVPE).⁸ A metal catalyst is usually used (i.e., Au) to grow NW structures, however with SA-MOVPE a catalyst is not needed, preventing diffusion of the metal into the NW. A (111) InP wafer is covered by 30 nm of SiO₂. By electron

beam lithography and wet-etching, 40–60 nm diameter openings are created to form NW nucleation-sites. At a growth rate of 3 nm/s, first a 1 μm long segment of InP is grown by adding trimethylindium and tertiarybutylphosphine (TBP) to the MOVPE reactor at 640 °C. Subsequently the temperature is lowered to 580 °C and arsine (AsH₃) is added to the reactor (V/III ratio 340, partial pressure TBP:AsH₃:1) to grow 8 to 10 nm InAsP to form the QDs. The QDs are embedded in an InP shell, grown at 580 °C. To finalize the NW, a second 1 μm segment of InP is grown at 640 °C. A scanning electron microscope image of the array is shown in Fig. 1(a). A schematic of an individual NW is shown in Fig. 1(b).

NW-QD PL spectroscopy is performed by micro-PL measurements. The sample is cooled to 5 K. The NW-QDs are excited with a continuous wave Ti:sapphire laser tuned at a wavelength of 920 nm, at a power of ~ 3 kW/cm² directed along the NW growth direction and focused with an aspheric lens (numerical aperture=0.5). The NW-QD emission collected by the same aspheric lens is dispersed by a double grating spectrometer ($f=1.0$ m) and detected with a cooled InGaAs photodiode array. An exposure time of 1 s yields a spectrum with a high signal-to-noise ratio. A typical spectrum is shown in Fig. 1(c). We identify an intense peak centered at 1211.6 nm (1.0254 eV) with a line width of 46 μeV full width at half maximum and we name it X⁰ (neutral exciton), for reasons which will appear later. As expected, the fine structure splitting normally induced by QD lateral anisotropy and associated exchange interaction⁹ is not observed in polarization dependent measurements, within our setup resolution of less than 5 μeV . An additional line, labeled XX⁰ (neutral biexciton), can be seen at 1211.3 nm (1.02328 eV). The radiative lifetime of the X⁰ and XX⁰ transitions are measured by time-resolved PL measurements using a streak camera, see Fig. 1(d). A monoexponential fit reveals a lifetime of

^{a)}Electronic mail: s.n.dorenbos@tudelft.nl.

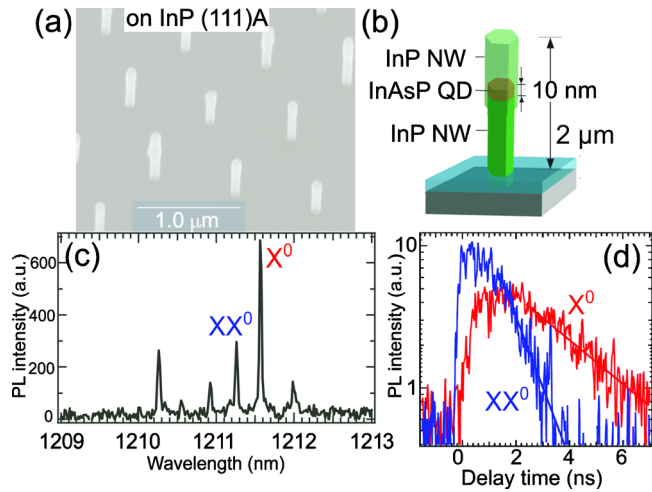


FIG. 1. (Color online) (a) Scanning electron microscope image of the regular array of NWs. (b) Schematic of the NW, consisting of two $1 \mu\text{m}$ long InP segments and a 10 nm InAsP section forming the QD. (c) Spectrum of the NW-QD. The origin of the X^0 line and the XX^0 line follows from cross correlation measurements. (d) Time resolved measurement of the X^0 and XX^0 line. The solid line is a fit with a monoexponential decay, which gives a lifetime of 2.8 ns and 0.87 ns , respectively.

2.8 ns for the X^0 and 0.87 ns for the XX^0 . The long lifetime indicates absence of fast nonradiative processes, revealing the high quality of NW-QDs. We note that the ratio of the lifetime is $\tau_{X^0}/\tau_{XX^0}=3.2$, which indicates a larger dot.¹⁰ We also note the appearance of cascaded emission of XX^0 and X^0 , which we will prove with a cross correlation measurement.

Single photon emission is demonstrated with a Hanbury Brown–Twiss (HBT) experiment [see Fig. 2(a)]. The NW-QD is excited with continuous wave He–Ne laser at 633 nm at a power of $\sim 150 \text{ kW}/\text{cm}^2$, close to saturation power. In each arm of the beam splitter (BS) the emission is filtered with a 0.5 nm wide band pass filter (BPF) to select the X^0 line and sent to an SSPD, chosen for their sensitivity at the wavelength of interest. A detailed description of our SSPDs is given in Refs. 11–13. The detectors are operated at 4.2 K and current biased near the critical current (I_c). The following characteristics are observed: for detector 1, at $0.85 \cdot I_c$ the dark count rate is $70 \text{ counts per second}$, together with 4% efficiency at a wavelength of $1.3 \mu\text{m}$. Detector 2 is operated at $0.82 \cdot I_c$, where it shows $40 \text{ dark counts per second}$ and an efficiency of 3% . The second order correlation function of the output of the two detectors is measured with a time to

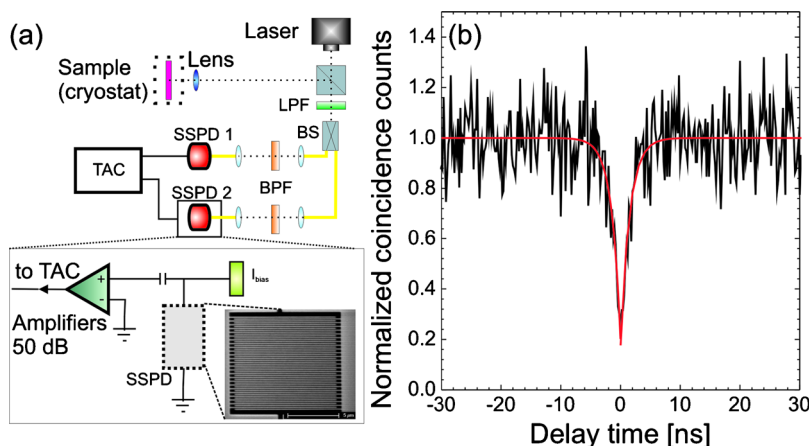


FIG. 2. (Color online) (a) Correlation measurement setup. The emission is filtered with a long pass filter (1100 nm), and coupled to a BS. In each arm of the BS a 0.5 nm broad BPF is used to select one emission line, which is sent to an SSPD. A TAC records coincidence counts. (inset) SSPD operation schematic, showing the current supply (I_{bias}), the amplification stage, and a SEM picture of the device, consisting of a 100 nm wide and $500 \mu\text{m}$ long wire, meandering in an area of $10 \times 10 \mu\text{m}^2$. (b) Normalized histogram of the autocorrelation measurement of the X^0 . Excitation wavelength is 633 nm and the time bins are 200 ps . The solid line is a fit, indicating a dip of the correlation function of $g^{(2)} \times (0) = 0.12$. Count rates ranging from 5 to 15 kHz are observed, the integration time was 30 min .

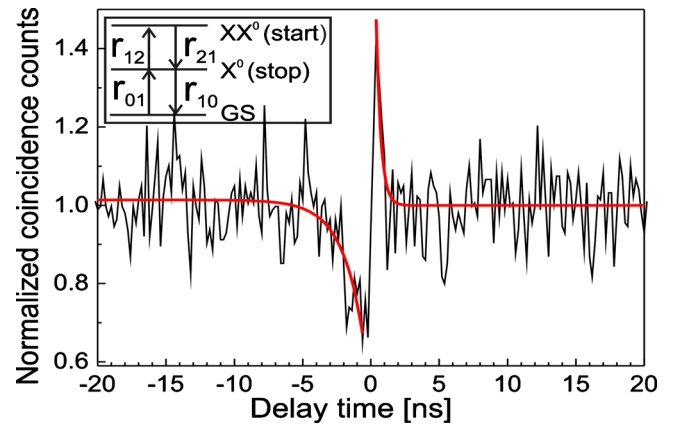


FIG. 3. (Color online) Cross correlation function of the XX^0 (start) and X^0 (stop). Excitation wavelength is 920 nm . The biexciton–exciton cascade is modeled as a three level system (see inset). The solid line is a fit according to this model. This configuration leads to a peak for positive times [$g^{(2)} \times (0.4) = 1.5$], where the probability of emitting a photon (X^0) is enhanced. For negative times the probability is decreased resulting in a dip: $g^{(2)} \times (-0.2) = 0.65$.

amplitude converter (TAC). A histogram of the coincidence counts with time bins of 200 ps and an integration time of 30 min is shown in Fig. 2(b). The dip at zero time delay indicates the emission of nonclassical light. The data is fitted through a second-order correlation function $g^{(2)}(\tau) = 1 - ae^{-\tau/t}$, a accounts for background light and dark counts of the SSPD, $t = t_l + t_p$, where t_l is the lifetime of the NW-QD and t_p is the inverse pumping rate. The function at zero delay gives $g^{(2)}(0) \leq 0.2$, far below the threshold of 0.5 , i.e., unambiguously proving that the NW-QD is a single photon emitter. To correct the data for the background light, the data is fitted with $g_b^{(2)}(\tau) = 1 + \rho^2 [g^{(2)}(\tau) - 1]$, with $\rho = S/(S+B)$ related to the signal to noise ratio.¹⁴ Here a value of $g_b^{(2)}(0) \leq 0.12$ is obtained with $\rho = 0.97$. From the monoexponential decay of the fit the time constant of this measurement can be extracted, which is $t = 1.51 \text{ ns}$. Measurements with different t_p would allow to extract t_l , which can be compared to the streak camera measurement.

With cross correlation experiments a biexciton–exciton cascade can be identified. In the inset of Fig. 3 a schematic of the biexciton–exciton cascade is shown. Upon excitation with rate r_{01} and r_{12} , the dot is filled with two electron-hole pairs (XX^0), neglecting resident charges. At a rate r_{21} one electron-hole pair recombines, leaving the X^0 behind, which

subsequently recombines at a rate r_{10} . In the first arm of the HBT the biexciton is filtered, while the exciton is filtered in the second arm, with the use of BPFs. The output signals of the SSPDs are connected such that the biexciton serves as the start and the exciton as the stop of the TAC. The NW-QD is excited with a continuous wave Ti:sapphire laser ($\lambda = 920$ nm) at a power close to saturation power. Again the second order correlation function is measured and shown in Fig. 3. The difference with Fig. 2(b) can be immediately seen as the function is not symmetric around a time delay of 0 ns. As has been shown before in Ref. 3 this is the signature of cascaded emission. For positive time the peak indicates an enhanced probability of emitting X^0 after emission of XX^0 . For negative times, the dip represents a decrease in probability of the emission of the next XX^0 after the emission of X^0 because the system has to be re-excited. The cascade is modeled as a three level system (see Fig. 3 inset), which implies the cross-correlations can be fitted to two decay paths with their own typical time-constants. From the fitting we conclude that the dip reaches $g^{(2)}(-0.2) = 0.65$ and the peak has a value of $g^{(2)}(0.4) = 1.5$. The function does not reach zero at $\tau = 0$ because of stray background light. The exponential decay obtained from the fit leads to a lifetime of 0.34 ns for the biexciton and 1.24 ns for the exciton. The discrepancy with the streak camera measurement is attributed to a difference in pump power. It has been confirmed that the exciton as well as the biexciton show antibunching under these conditions (not shown).

We have shown that the use of SSPDs enables (cross-) correlation experiments with single photon sources emitting in the infrared because of the SSPD's high efficiency, low dark count rate, and high timing resolution. Arrays of NW-QDs with very good properties can be fabricated without the use of catalysts and can form arrays of nonclassical light emitters in the infrared. Further studies will reveal whether the absence of fine structure splitting gives rise to entanglement between the biexciton and exciton. Tapering the NWs would improve the extraction efficiency.¹⁵ Due to the specific properties of NWs, an electrically controlled source¹⁶ is now within reach.

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