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Controlling the exciton energy of a nanowire quantum dot by strain fields

Yan Chen,[1] Iman Esmaeil Zadeh,[2] Klaus D. Jöns,[2] Andreas Fognini,[2] Michael E. Reimer,[3] Jiaxiang Zhang,[1] Dan Dalacu,[4] Philip J. Poole,[4] Fei Ding,[1,a)] Val Zwiller,[2] and Oliver G. Schmidt[1,5]
[1]*Institute for Integrative Nanosciences, IFW Dresden, Helmholtz Strasse 20, 01069 Dresden, Germany*
[2]*Kavli Institute of Nanoscience, Delft University of Technology, 2600 GA Delft, The Netherlands*
[3]*Institute for Quantum Computing and Department of Electrical and Computer Engineering, University of Waterloo, Waterloo, N2L 3G1, Canada*
[4]*National Research Council, Ottawa, Ontario K1A 0R6, Canada*
[5]*Material Systems for Nanoelectronics, Chemnitz University of Technology, Reichenhainer Strasse 70, 09107 Chemnitz, Germany*

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We present an experimental route to engineer the exciton energies of single quantum dots in nanowires. By integrating the nanowires onto a piezoelectric crystal, we controllably apply strain fields to the nanowire quantum dots. Consequently, the exciton energy of a single quantum dot in the nanowire is shifted by several meVs without degrading its optical intensity and single-photon purity. Second-order autocorrelation measurements are performed at different strain fields on the same nanowire quantum dot. The suppressed multi-photon events at zero time delay clearly verify that the quantum nature of single-photon emission is well preserved under external strain fields. The work presented here could facilitate on-chip optical quantum information processing with the nanowire based single photon emitters. *Published by AIP Publishing.*
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Quantum optical information processing has imposed several key requirements on a good single-photon emitter. First, the emitter should generate single-photons with high brightness and long coherence time. Second, the emitted photons from one source, and much more preferred from independent sources, should be indistinguishable.[1–5] Semiconductor quantum dots (QDs) embedded in bottom-up grown nanowires have the potential to fulfill these requirements.[6,7] For the first requirement, these wires benefit from the recently developed two step growth process combining selective-area and vapor-liquid-solid epitaxy techniques.[8,9] A single QD can be ideally embedded into the nanowire, which overcomes previous random approaches of fabricating waveguides around multiple QDs.[10–12] Bright single photon sources based on the similar nanowires with the extraction efficiencies of up to 42% have been demonstrated.[13] A further improved extraction efficiency exceeding 90% can be achieved if the wire shape is smoothly tapered and a thin dielectric-gold mirror is integrated underneath.[14] The nanowire also features a Gaussian emission profile, which is important for the fiber coupling.[15–17] The significant reduction of nearby traps also results in an ultra-clean spectrum with long coherence times of up to 1.2 ns.[8,18]

For the second requirement, however, the nanowire QDs suffer from inhomogeneous spectral broadening of the exciton emission from different sources due to shape, strain, and composition inhomogeneity during the growth.[7,12] Therefore, the single photons from different sources are distinguishable, and scaling up these sources is extremely challenging.[19] Post-growth tuning is therefore essential in order to achieve indistinguishable photons from the independent sources. Tuning the exciton emission from nanowire QDs will also find important applications in a hybrid system, where the QDs can be coupled to an atomic system either to counteract slow spectral diffusion in QDs or to store photons in a quantum memory.[20]

To date, the methods for engineering the nanowire QD emission are limited. Signorello *et al*. clamped a freestanding nanowire by a Ti contact onto a flexible substrate.[21] By bending the nanowire, they shifted the photoluminescence (PL) signal from GaAs/$Al_{0.3}Ga_{0.7}As$/GaAs core/shell nanowires by over 290 meV. However, on-chip implementation of such a "bending wire" setup is quite challenging, and the core/shell nanowires by their nature are not single-photon sources. Kremer *et al*. fabricated QD-containing nanowires on a piezo substrate by reactive ion etching and managed to tune the QD exciton emission over 1.2 meV by strain.[22] In addition to the fabrication challenges such as rough nanowire surfaces and shape control, the top-down fabrication approach generally has a low yield since the QD locations are not deterministically aligned during the etching. Reimer *et al*. have successfully removed the biexciton binding energy in nanowire quantum dots by a local lateral electric field. Unfortunately, in this configuration, the in-plane emission is blocked by electrical contacts at both the ends of the nanowire to pump away excess charges,[23,24] and is therefore not desirable for on-chip quantum information processing unless new designs are envisioned.

In this letter, we demonstrate wavelength-tunable single-photon sources from nanowire QDs by strain. The studied sample is a single InAsP QD embedded in a bottom-up grown InP nanowire waveguide. The nanowire shape can be modified during growth to control the tapering angle, diameter, and length (Fig. 1). Detailed information on the growth is discussed elsewhere.[15] The tapered nanowires were

a)Author to whom correspondence should be addressed. Electronic mail: f.ding@ifw-dresden.de.

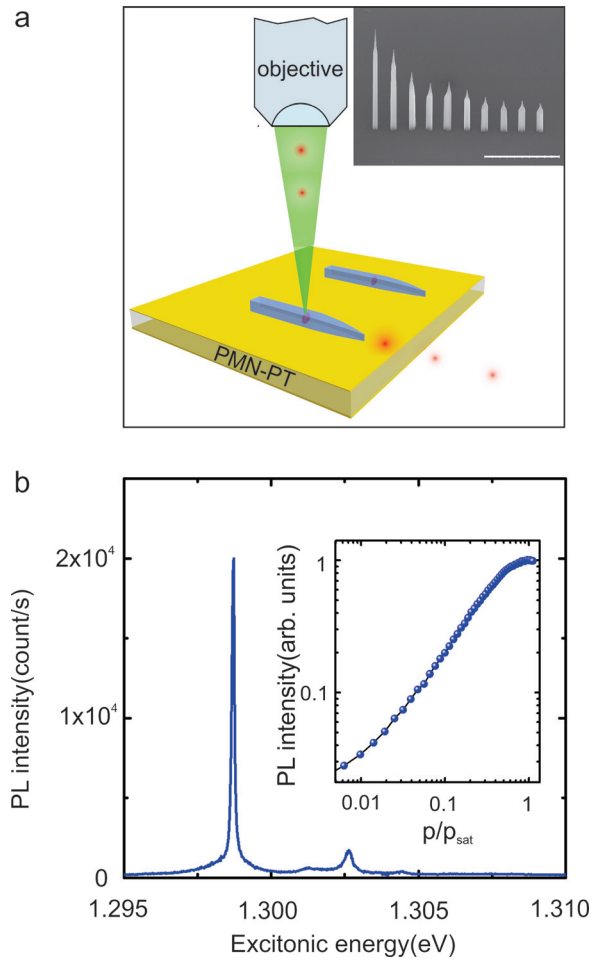

FIG. 1. (a) Sketch of the device. Inset shows a scanning electron microscopy image of the nanowires with different tapering. Scale bar indicates 3 μm. Considering the QD dipole orientation, the emission can be collected either from the top of the nanowire (this work) or along the nanowire axis. (b) Emission spectrum of a single nanowire QD. Inset shows the linear dependence of the integrated emission intensity on the excitation power.

picked-up from the growth substrate by a nano-manipulator and transferred subsequently onto a piezoelectric crystal consisting of [Pb $(Mg_{1/3}Nb_{2/3})$ $O_3]_{0.72}[PbTiO_3]_{0.28}$ (PMN-PT) (see Fig. 1(a)). The choice of PMN-PT is due to its large in-plane strain capabilities and negligible drop of strain at low temperatures.[25]

The device is mounted on the cold-finger of a continuous helium-flow cryostat and all of the optical experiments are carried out at 5 K. A continuous-wave laser at wavelength of 532 nm is focused onto the nanowire by an objective with numerical aperture of 0.42. The radiative dipole of a single nanowire QD lies in a plane perpendicular to the nanowire axis. Therefore, the excitation and collection of the photoluminescence (PL) can be done either along the nanowire axis or from the top of the nanowire (perpendicular to the axis). With both excitation and collection geometries, the peak emissions have exactly the same energies.[26] In this work, we used the geometry as sketched in Fig. 1(a) to demonstrate the strain tuning of the nanowire emission. For on-chip integration, however, the in-plane emission along the axis can be used together with dielectric waveguides to obtain much higher photon fluxes.[27] Fig. 1(b) shows a typical PL spectrum from a single nanowire QD. The integrated intensity of the main emission line has a linear dependence on the laser excitation power (see inset of Fig. 1(b)) and its fine structure splitting[28] is $\sim$5 μeV. We attribute this line to neutral exciton emission.

A bias voltage V applied to the PMN-PT results in an out-of-plane electric field E_p, leading to an in-plane strain in the nanowires that modifies the QD emission energy.[25,29,30] The PL emission spectrum is recorded while sweeping the voltage applied to the piezoelectric substrate. Figure 2(a) shows the typical result for strain tuning of a single nanowire QD. We observe a clear blue shift of the QD emission when increasing the electric field E_p This shift is because increasing the electric field corresponds to compressive strain, which increases the emission energy. Both blue and red shifts can be achieved with our device, simply by changing the polarity of the applied voltages. Two types of devices were used to study the efficiency of strain transfer. In the first type of devices, the nanowires were anchored by a thin silicon nitride (SiN) layer (which is deposited on top of the whole device) as we expected more efficient strain transfer. However, the anchoring layer could affect the guiding properties of the nanowire for on-chip applications. As shown in very recent work, this problem can be solved by embedding the nanowires into structured silicon nitride (SiN) waveguide channels,[27] where the electric dipole moment of the QD embedded in the nanowire matches the fundamental waveguide mode of the SiN. In the second type of devices without anchoring layers, the nanowires were transferred directly onto the PMN-PT substrate. Surprisingly, we observed that the emission energy tuning can be achieved with the same order of magnitude in these two types of devices. We believe that in the devices without anchoring layers, the nanowires and the substrate can be held together by interlayer van der Waals forces, as being observed in 2D material heterojunctions.[31] This finding will greatly simplify the fabrication processes for future devices where in-plane single-photon emitters are needed.

To demonstrate the wavelength-tunable single-photon emission from nanowire QDs, we have measured the second-order autocorrelation functions[32] $g^{(2)}(\tau)$ as E_p is varied from -10 to 25 kV/cm. After spectral filtering of the neutral

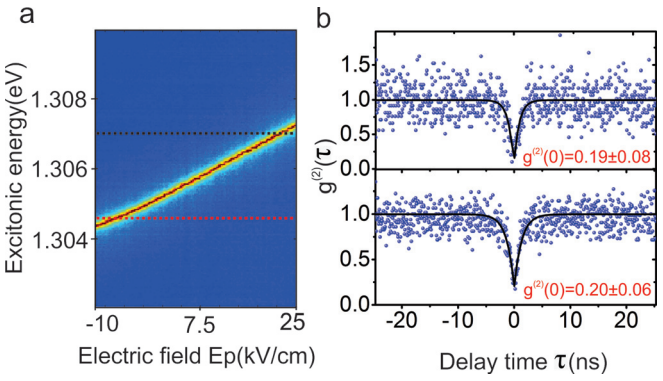

FIG. 2. Strain tuning from a nanowire QD. (a) PL from the nanowire quantum dot as a function of strain. By changing the electric field *E*p applied to the PMN-PT substrate, the exciton energy can be controllably shifted by $\sim$3 meV. (b) Normalized second-order autocorrelation functions measured at two different strain fields (emission energies are indicated by the black and red dashed lines in (a)).

exciton with a spectrometer, a non-polarizing 50:50 beam splitter splits the exciton emission between two single-photon avalanche detectors. Figure 2(b) shows the measured $g^{(2)}(\tau)$ for two different strain fields corresponding to dissimilar emission energies. At an exciton energy of 1.307 meV, a multiphoton emission probability of $g^{(2)}(0)$ of 0.19 ± 0.08 is obtained by fitting the experimental data. When the exciton energy is shifted by 2 meV, the $g^{(2)}(0)$ remains unchanged within the experimental error, with a value of 0.20 ± 0.06 (solid line in Figure 2(b), with no background subtraction in both the cases). The non-vanishing $g^{(2)}(0)$ is most likely due to stray light from background emission, the dark counts, and limited timing resolution of our single-photon detectors.

We can also tune the QD energies from different nanowires into spectral resonance. For this purpose, we transfer two nanowire QDs onto two separate chips. The results are shown in Figure 3(a). For the applied electric fields of 2 kV/cm and 14 kV/cm, the emission energies of two QDs are tuned to the same value. This strain tuning technique of the emission energy that we present opens up a scalable route to realize two-photon interference between two spatially separated nanowire single-photon sources. It is worth mentioning that in the current work, the bulk PMN-PT substrate was used to demonstrate the potential of strain tuning of nanowires on-chip. The structures inherently have large footprints and high operation voltages, which are not desired for photonic on-chip implementations. In future work, the piezo substrate can be replaced by thin PMN-PT films of a few micrometer thickness on silicon or many other substrates.[33] After patterning the piezo film, we can obtain single strain-tunable devices with ultra-small footprints (several tens of microns) and low operation voltages (down to a few volts).

We also would like to address the question of what is the percentage of these nanowire QDs that can be tuned into spectral resonance. We compare the emission tuning range of single nanowire QDs with the inhomogeneous emission broadening of 50 nanowire QDs. The maximum strain tuning range of a single nanowire QD that we achieved in this work is shown in Fig. 4(a). By applying a voltage from −300 V to 800 V to the PMN-PT, the exciton emission energy is shifted by 6.3 meV. The observed non-linear tuning behavior is most likely due to the imperfect anchoring of the nanowire. We measured 50 QDs from another growth run with different emission energies to determine the inhomogeneous spectral broadening. The distribution of their emission energies is shown in Fig. 4(b). The data are fitted by a Gaussian envelope. The standard deviation is 5.7 meV, which is smaller than the strain tuning range. This result suggests that we can tune most nanowire QDs into spectral resonance with each other from the same wafer.

To evaluate the feasibility of on-chip two-photon interference, we conducted a Hong-Ou-Mandel (HOM) measurement on a single nanowire.[2,34] The measurement is carried out in a cryostat, which is cooled down to 300 mK. The

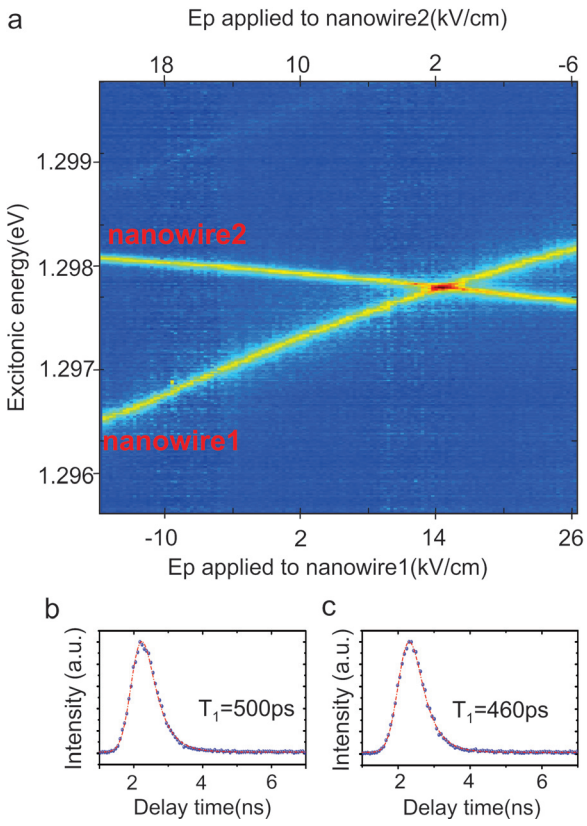

FIG. 3. Two nanowire QDs can be tuned into spectral resonance by strain. (a) Exciton emissions of two nanowires on separate chips are controllably tuned by strain into spectral resonance. (b) and (c) Exciton lifetime of two QD emissions at degeneracy are 500 ps (b) and 460 ps (c), respectively. Red lines are the single exponential fit after taking into account the transit time jitter in the detector.

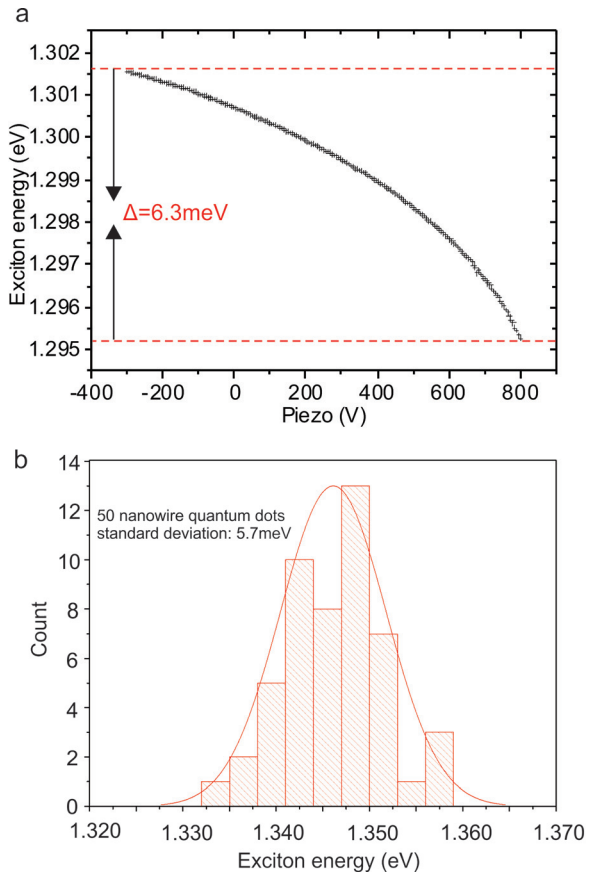

FIG. 4. (a) Exciton emission energy of a single nanowire QD is shifted by 6.3 meV. (b) Inhomogeneous spectral broadening of 50 nanowire QDs from another growth run with different emission energies. The red envelope is a Gaussian fit, with a standard deviation of 5.7 meV.

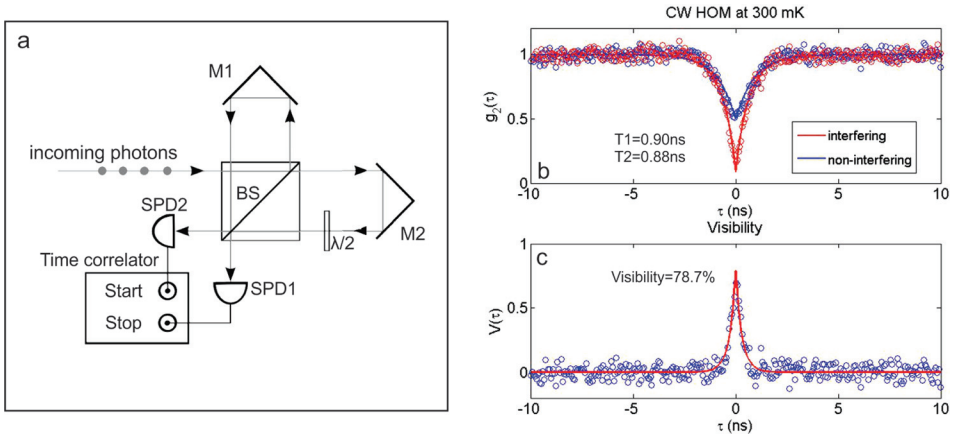

FIG. 5. (a) Sketch of experimental set-up to measure the single photon indistinguishability. Two retroreflective mirrors, M1 and M2, are placed at both the ends of the interferometer. Two single photon detectors, SPD1 and SPD2, are used to characterize the two-photon interference. (b) HOM interference data recorded for 3 h. Red circle indicates the interfering data and the blue line corresponds to the non-interfering case. Solid lines are the theoretical fit. (c) Extracted two-photon interference visibility. Maximal indistinguishability of 78.7% is achieved.

nanowire is excited above the bandgap by a continuous-wave HeNe laser. A schematic of the optical set-up used to perform the HOM measurement is illustrated in Fig. 5(a). First, the emitted photons from a single nanowire QD are directed towards a 50:50 beam splitter. The two photons then take either the short or the long path of the interferometer arms. A half-wave plate is inserted in one of the interferometer arms, so that we can measure two-photon coincidence counts for both cross and parallel polarization by rotating this plate. Finally, a single photon detector is placed at each output arm to record the photon correlations. The histogram is shown in Fig. 5(b). For both cases of two-photon indistinguishability (red circles) and distinguishability (blue circles) measurements, the data are recorded for 3 h. We extract a lifetime and coherence time of 0.90 ns and 0.88 ns, respectively, and a two-photon interference visibility of 78.7% (Fig. 5(c)). For future experiments with two different nanowire quantum dots, resonant excitation can be used to further improve the coherence and the two-photon interference visibility.[35]

In summary, we have demonstrated wavelength-tunable single-photon sources from nanowire quantum dots by using the strain engineering techniques. Due to several important advantages, the tapered nanowires grown in a bottom-up approach are ideal single-photon sources for on-chip quantum photonic applications when laid down on the substrate. With the strain tuning method presented in this work, the inhomogeneity of their emission energies can potentially be overcome. As an example, we demonstrate the spectral resonance of two strain engineered nanowire QDs. Our strain engineering technique is also compatible with nanowires embedded in dielectric waveguides to facilitate complex routing of indistinguishable single photons. To evaluate the feasibility of on-chip two-photon interference, we conducted HOM type measurements and obtained a photon indistinguishability of around 80% from one nanowire under non-resonant excitation. Combined with the recently developed on-chip strain tuning platform with low operation voltages and ultra-small footprints,[33] our work may facilitate important quantum optical experiments such as two-photon interference with on-chip integrated nanowire QDs.[32,36]

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[1]L. M. Duan, M. D. Lukin, J. I. Cirac, and P. Zoller, Nature **414**(6862), 413 (2001).
[2]C. Santori, D. Fattal, J. Vuckovic, G. S. Solomon, and Y. Yamamoto, Nature **419**(6907), 594 (2002).
[3]A. Kuhn, M. Hennrich, and G. Rempe, Phys. Rev. Lett. **89**, 067901 (2002).
[4]J. L. O'Brien, G. J. Pryde, A. G. White, T. C. Ralph, and D. Branning, Nature **426**, 264 (2003).
[5]E. B. Flagg, Phys. Rev. Lett. **104**, 137401 (2010).
[6]T. Heindel, C. Schneider, M. Lermer, S. H. Kwon, T. Braun, S. Reitzenstein, S. Höfling, M. Kamp, and A. Forchel, Appl. Phys. Lett. **96**(1), 011107 (2010).
[7]I. Friedler, C. Sauvan, J. P. Hugonin, P. Lalanne, J. Claudon, and J. M. Gérard, Opt. Express **17**(4), 2095 (2009).
[8]D. Dalacu, K. Mnaymneh, J. Lapointe, X. Wu, P. J. Poole, G. Bulgarini, V. Zwiller, and M. E. Reimer, Nano Lett. **12**(11), 5919 (2012).
[9]D. Dalacu, A. Kam, D. Guy Austing, X. Wu, J. Lapointe, G. C. Aers, and P. J. Poole, Nanotechnology **20**(39), 395602 (2009).
[10]J. Claudon, J. Bleuse, N. S. Malik, M. Bazin, P. Jaffrennou, N. Gregersen, C. Sauvan, P. Lalanne, and J.-M. Gerard, Nat. Photonics **4**, 174 (2010).
[11]A. Dousse, L. Lanco, J. Suffczyński, E. Semenova, A. Miard, A. Lemaître, I. Sagnes, C. Roblin, J. Bloch, and P. Senellart, Phys. Rev. Lett. **101**(26), 267404 (2008).
[12]J. Heinrich, A. Huggenberger, T. Heindel, S. Reitzenstein, S. Höfling, L. Worschech, and A. Forchel, Appl. Phys. Lett. **96**(21), 211117 (2010).
[13]M. E. Reimer, G. Bulgarini, N. Akopian, M. Hocevar, M. B. Bavinck, M. A. Verheijen, E. P. A. M. Bakkers, L. P. Kouwenhoven, and V. Zwiller, Nat. Commun. **3**, 737 (2012).
[14]I. Friedler, P. Lalanne, J. P. Hugonin, J. Claudon, J. M. Gérard, A. Beveratos, and I. Robert-Philip, Opt. Lett. **33**(22), 2635 (2008).
[15]G. Bulgarini, M. E. Reimer, M. B. Bavinck, K. D. Joüns, D. Dalacu, P. J. Poole, E. P. A. M. Bakkers, and V. Zwiller, Nano Lett. **14**(7), 4102 (2014).
[16]G. Bulgarini, D. Dalacu, P. J. Poole, J. Lapointe, M. E. Reimer, and V. Zwiller, Appl. Phys. Lett. **105**(19), 191113 (2014).
[17]N. Gregersen, T. R. Nielsen, J. Claudon, J.-M. Gérard, and J. Mørk, Opt. Lett. **33**(15), 1693 (2008).
[18]M. E. Reimer, G. Bulgarini, R. W. Heeres, B. J. Witek, M. A. M. Versteegh, D. Dalacu, J. Lapointe, P. J. Poole, and V. Zwiller,

"Overcoming power broadening of the quantum dot emission in a pure wurtzite nanowire," preprint arXiv:1407.2833 (2014).
[19]J. Bleuse, J. Claudon, M. Creasey, N. S. Malik, J.-M. Gérard, I. Maksymov, J.-P. Hugonin, and P. Lalanne, Phys. Rev. Lett. **106**(10), 103601 (2011).
[20]N. Akopian, U. Perinetti, L. Wang, A. Rastelli, O. G. Schmidt, and V. Zwiller, Appl. Phys. Lett. **97**(8), 082103 (2010).
[21]G. Signorello, S. Karg, M. T. Bjoürk, B. Gotsmann, and H. Riel, Nano Lett. **13**(3), 917 (2013).
[22]P. E. Kremer, A. C. Dada, P. Kumar, Y. Ma, S. Kumar, E. Clarke, and B. D. Gerardot, Phys. Rev. B **90**(20), 201408 (2014).
[23]M. E. Reimer, M. P. Van Kouwen, A. W. Hidma, M. H. M. Van Weert, E. P. A. M. Bakkers, L. P. Kouwenhoven, and V. Zwiller, Nano Lett. **11**(2), 645 (2011).
[24]M. P. van Kouwen, M. E. Reimer, A. W. Hidma, M. H. M. van Weert, R. E. Algra, E. P. A. M. Bakkers, L. P. Kouwenhoven, and V. Zwiller, Nano Lett. **10**(5), 1817 (2010).
[25]F. Ding, R. Singh, J. D. Plumhof, T. Zander, V. Křápek, Y. H. Chen, M. Benyoucef, V. Zwiller, K. Dörr, G. Bester, A. Rastelli, and O. G. Schmidt, Phys. Rev. Lett. **104**(6), 067405 (2010).
[26]G. Bulgarini, M. E. Reimer, and V. Zwiller, Appl. Phys. Lett. **101**(11), 111112 (2012).
[27]I. E. Zadeh, A. W. Elshaari, K. D. Jöns, A. Fognini, D. Dalacu, P. J. Poole, M. E. Reimer, and V. Zwiller, Nano Lett. **16**(4), 2289 (2016).
[28]V. D. Kulakovskii, G. Bacher, R. Weigand, T. Kümmell, A. Forchel, E. Borovitskaya, K. Leonardi, and D. Hommel, Phys. Rev. Lett. **82**(8), 1780 (1999).
[29]J. Zhang, Y. Huo, A. Rastelli, M. Zopf, B. Hoüfer, Y. Chen, F. Ding, and O. G. Schmidt, Nano Lett. **15**(1), 422 (2015).
[30]J. Zhang, J. S. Wildmann, F. Ding, R. Trotta, Y. Huo, E. Zallo, D. Huber, A. Rastelli, and O. G. Schmidt, Nat. Commun. **6**, 10067 (2015).
[31]G. S. Duesberg, Nat. Mater. **13**(12), 1075 (2014).
[32]R. H. Brown and R. Q. Twiss, Nature **178**(4541), 1046 (1956).
[33]Y. Chen, J. Zhang, M. Zopf, K. Jung, Y. Zhang, R. Keil, F. Ding, and O. G. Schmidt, Nat. Commun. **7**, 10387 (2016).
[34]C. K. Hong, Z. Y. Ou, and L. Mandel, Phys. Rev. Lett. **59**(18), 2044 (1987).
[35]C. Matthiesen, A. N. Vamivakas, and M. Atatüre, Phys. Rev. Lett. **108**(9), 093602 (2012).
[36]R. B. Patel, A. J. Bennett, I. Farrer, C. A. Nicoll, D. A. Ritchie, and A. J. Shields, Nat. Photonics **4**(9), 632 (2010).