ACS Photonics

Letter

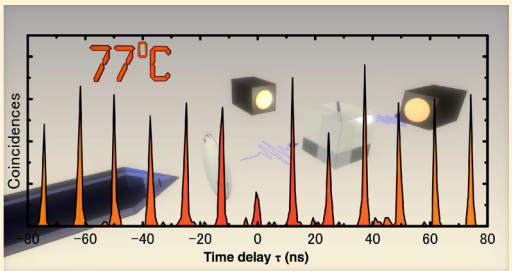
pubs.acs.org/journal/apchd5

Single Photons from a Hot Solid-State Emitter at 350 K

Mark J. Holmes,*[,†] Satoshi Kako,[‡] Kihyun Choi,[†] Munetaka Arita,[†] and Yasuhiko Arakawa*[,†,‡]

[†]Institute for Nano Quantum Information Electronics and [‡]Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8505, Japan

ABSTRACT: Sources of single photons are of central importance for the realization of several quantum information technologies including teleportation, cryptography, true random number generation, metrology, and some varieties of quantum computing. In principle the generation of single photons can be achieved via an optical transition in a quantum two-level system sufficiently separated from its environment. Solid-state semiconductor quantum dots are convenient structures that can provide such two-level systems, with engineered and tunable transition energies, but cryogenic temperatures are required in the vast majority of experiments in order to facilitate both carrier confinement and spectral isolation. The large-scale on-chip integration of such devices, however, due to inherent system heating, will require individual elements that can operate at temperatures in excess of room temperature. Here we report single-photon emission from an isolated state in a position-controlled GaN nanowire quantum dot at an unprecedented ambient temperature of 350 K (170 °F, 77 °C).



KEYWORDS: *nanowire, site-control, quantum dot, single-photon source*

Much progress has been made toward the isolation of individual quantum states (and the transitions between them) for both fundamental research into the quantum nature of reality and the realization and development of future quantum technologies.[1] One example is the single-photon emitter, which, being able to emit individual quanta of light on demand and at the same time possibly act as an interface between flying and stationary qubits, holds the potential to be the workhorse for a number of emerging quantum technologies. Practically, several architectures have been used to realize isolated single-photon emission including atoms/ions,[2] molecules,[3] crystal defects,[4−6] and both colloidal[7] and epitaxially grown quantum dots[8] (QDs). Of these, epitaxially grown quantum dots[9] are of particular interest due largely to their solid-state nature, which, in addition to the possible engineering of the QD electronic states, allows the prospect of site-controlled growth and integration into other nanostructures and nanocavities for the enhancement/modification of their emission properties.[10−12]

While scientific curiosity compels us to push materials to their limits and investigate single-quantum structures at extreme temperatures, it is also technologically desirable to have sources of single photons that operate at high temperature to overcome the need for costly and cumbersome cryogenic cooling, a feat made difficult for solid-state QDs due to phonon-induced emission broadening, which usually leads to the overlap of states and therefore an inherent spectral contamination. To date, there have been no reports of the successful observation of isolated states or indeed of single-photon emission at temperatures beyond room temperature from quantum dots[7,13−16] or any other type of structure, be they engineered or naturally occurring. Isolated single-photon emission at temperatures above 70 °C could in theory be used in computer processing units, data centers, and other environments susceptible to temperature fluctuations.

The realization and observation of such a system is achieved here using a small position-controlled GaN/$Al_{0.8}Ga_{0.2}N$ nanowire quantum dot.[17] The III-nitride material system is well known for its widespread application in LED lighting, lasers, and power electronics, but is chosen for this study due the large degree of quantum confinement achievable in its quantum dots, which makes them suitable for high-temperature single-photon emission.[15,16,18,19] The structures are fabricated by selective-area metal organic chemical vapor deposition and consist of GaN/$Al_{0.8}Ga_{0.2}N$ core−shell-type nanowires with a GaN quantum dot inclusion near the tips.[15,17] The dots themselves typically have a lateral dimension of ∼10 nm and a vertical dimension of ∼1 nm, resulting in emission in the deep ultraviolet at energies of 4.0−4.6 eV. Spatial isolation of a single-quantum structure is achieved through absolute control of the structure areal density during fabrication, in this case down to levels as low as 2.5 × 10^5 cm^{-2} (∼3 orders of magnitude lower than typical self-assembled QDs used for optical studies and 5−6 orders of magnitude lower than dots used as gain media in commercially available QD-based lasers). A scanning electron microscope image of a single structure, along with an optical microscope image, is shown in Figure 1a.

The optical properties of individual nanowire QDs were investigated using microphotoluminescence spectroscopy under excitation with an ultrafast femtosecond pulsed laser as described in the Methods section. The excitation photon energy of 5 eV (λ = 247 nm) is lower than the band gap of the barrier material, leading to carrier injection into the dot via

Received: February 16, 2016
Published: March 21, 2016

DOI: 10.1021/acsphotonics.6b00112
ACS Photonics 2016, 3, 543−546

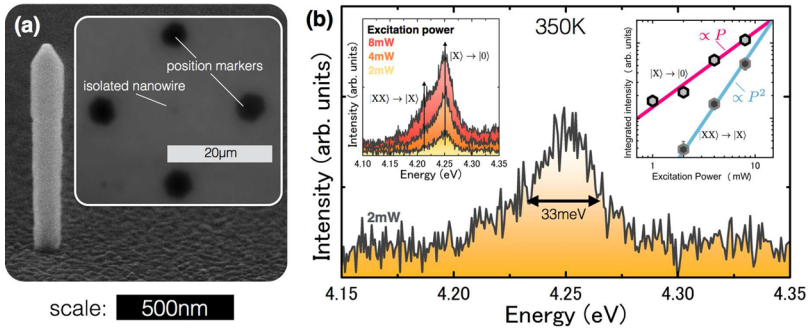


Figure 1. (a) Scanning electron microscope image of a single position-controlled nanowire. The inset shows an optical microscope image taken from directly above the sample using the experimental setup, revealing the spatial isolation of a single structure and position markers. The position markers (also formed during growth) are used for the easy optical registration of the nanowire, which is more difficult to observe due to its submicrometer diameter. (b) Emission spectrum from a single structure measured at 350 K. The emission line width is 33 meV. The figure insets show the increase of the side emission peak as the excitation power is increased and a logarithmic plot of the power dependences of the integrated intensities of the two peaks, revealing their linear and quadratic natures, respectively.

continuum states.[20,21] This excitation energy was chosen, as it was found to provide a slightly more efficient pumping of the dot and therefore a better signal to background ratio at higher temperatures. The emission spectrum of a structure heated to an ambient temperature of 350 K is shown in Figure 1b, exhibiting a clear peak centered at ∼4.25 eV with a full width at half-maximum of 33 ± 1.7 meV. This line width is induced by interactions between the dot-bound exciton and lattice vibrations, a coupling that is relatively large in these structures due to both the comparatively small QD size[22] and a non-negligible piezoelectric coupling mechanism in QDs made from materials such as GaN, which exhibit a non-centrosymmetric crystal structure.[23] Although 33 meV may seem fairly broad for a single quantum dot (the room-temperature inhomogeneous broadening of *ensembles* of infrared emitting QDs can reach <20 meV[24]), it is about an order of magnitude narrower than the typical room-temperature line widths of diamond-based nitrogen vacancy centers[4] and defects in SiC[5] and is comparable to the line widths of other single-photon-emitting diamond impurities.[6]

Under increasing excitation power a lower energy shoulder peak becomes apparent—separated from the main emission by approximately 40 meV—and begins to increasingly impose itself on the emission spectrum (see the figure inset). This is in good agreement with state of the art theoretical[25] and experimental studies,[26,27] which lead to the prediction of a biexciton state with a binding energy of 30−50 meV for a GaN quantum dot of such size and emission energy. Indeed, the integrated intensity of the peaks (also shown in the figure inset) exhibits a clear linear and quadratic excitation power dependence, evidence that their origins are excitonic ($|X\rangle \rightarrow |0\rangle$) and biexctionic ($|XX\rangle \rightarrow |X\rangle$) transitions of the position-controlled quantum dot, respectively.[18] Nevertheless, despite the high-temperature thermal broadening of the emission, it is clear from the figure that a combination of low excitation power and appropriate spectral filtering can be used for the isolation of a single emission peak.

Confirmation of the single-photon nature of the emission was achieved via measurement of the intensity autocorrelation, which is proportional to the second-order coherence at time delay τ,

$$g^{(2)}(\tau) = \frac{\langle I(\tau)\, I(t+\tau)\rangle}{\langle I(t)\rangle^2} \qquad (1)$$

where I is the emission intensity (see Figure 2). During this measurement the emission shown in Figure 1 was filtered using

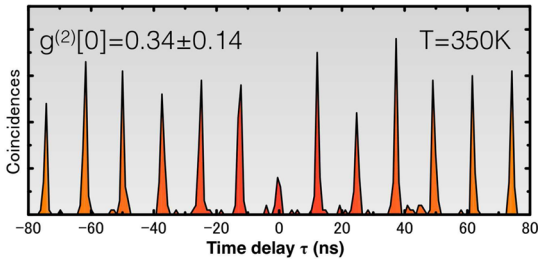


Figure 2. Autocorrelation histogram measured at 350 K. The suppression of coincidence counts at time delay $\tau = 0$ reveals a clear antibunching in the photon statistics, and the measured $g^{(2)}[0]$ value of 0.34 ± 0.14 verifies the presence of a single-quantum emitter.

the exit slit of the spectrometer set to the peak center and with a bandwidth of 36 meV. The photon statistics exhibit a strong antibunching through the suppression of coincidence counts at time delay $\tau = 0$, immediately revealing the nonclassical nature of the emission. Indeed, the value of the second-order coherence of the peak at zero time delay, $g^{(2)}[0]$, is measured to be 0.34 ± 0.14—below the threshold of 0.5 required to verify single photon emission[28]—therefore confirming the presence of a single-quantum emitter even at such high temperatures (the experimental error is given by the standard deviation of the intensities in the side peaks of the autocorrelation histogram). The residual $g^{(2)}[0]$ value is due to unfiltered spectral background contamination, analysis of which permits the calculation of a corrected $g^{(2)}[0]$ value of ∼0.06, which is zero to within the experimental error. In truth, such analysis merely provides an indication that the measured transition in itself is, to the best of our knowledge, acting as a pure source of single photons. It is the $g^{(2)}[0]$ value of 0.34 from the experiment that provides the most important information on the extent to which the single transition is measurable (in this case corresponding to ∼84% of the total collected signal).

Additional properties of the main emission line were measured as a function of temperature as shown in Figure 3. Emission is observable at temperatures up to 400 K, albeit with a significantly reduced intensity. As the temperature is raised, the emission exhibits the usually observed monotonic shift to

DOI: 10.1021/acsphotonics.6b00112
ACS Photonics 2016, 3, 543−546

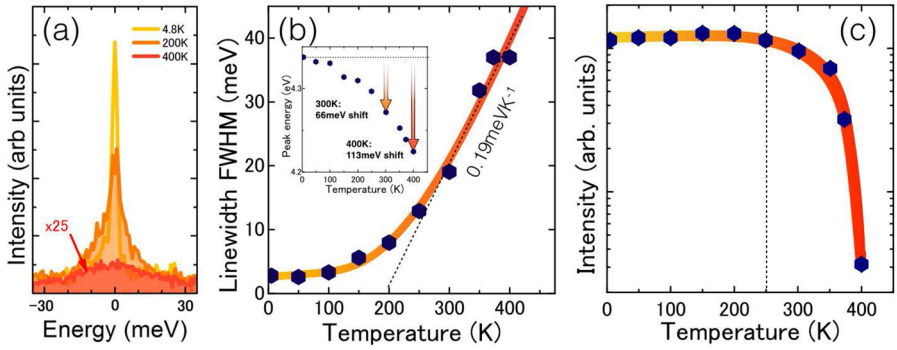


Figure 3. Properties of the isolated emission line as a function of temperature over a range of ~2 orders: (a) peak line shape, (b) line width (the figure inset shows the monotonic red-shift with increasing temperature), and (c) intensity. The solid lines in (b) and (c) are guides to the eye.

lower energy, and the intensity begins to drop rapidly at temperatures above 250 K. No signal is detectable at 425 K, possibly revealing an operational limit for such site-controlled GaN nanowire QDs. Although it is likely that carrier escape is the main cause for the reduction in intensity, the exact mechanism remains uncertain at present. The rapid decrease in intensity renders higher temperature autocorrelation measurements unfeasible with current detector technology. As discussed earlier, the strong acoustic phonon interactions in III-nitride QDs lead to significant line broadening. The spectra shown in Figure 3 reveal the emergence and eventual dominance of phonon wings in the emission line, with a corresponding linear increase in emission line width at a rate of ~0.19 meV K^{-1}. Previous studies into such phonon-wing broadening in III-As[29] and CdTe/ZnTe[30] QDs over smaller temperature ranges report broadening rates of 0.02 and ~0.06 meV K^{-1}, respectively, highlighting the extent of the line broadening in the current material system.

The fact that such semiconductor nanostructures can support isolated transitions and hence be used as sources of single photons under such extreme ambient conditions provides a possible key to unlock a variety of solid-state quantum technologies at high temperatures. Although the current example involves emission in the ultraviolet, it is particularly noteworthy that, in addition to conventional high-power lighting applications, the III-nitride material system can in principle be used for quantum light emission across a broad wavelength range from the deep ultraviolet to the near-infrared.[16,31,32]

METHODS

Sample Preparation. Sample growth was performed using selective-area metal organic chemical vapor deposition (MOCVD) at a pressure of 76 Torr in a horizontal quartz reactor using trimethylgallium, trimethylaluminum, and amonia as precursors. The substrate used for growth consisted of a 25 nm thick low-temperature AlN layer grown on sapphire. A 25 nm SiO_2 layer was deposited, into which arrays of 25 nm diameter apertures were opened using standard electron beam lithography and reactive ion etching. The apertures act as site-controlled centers for nanowire growth, and control over the aperture separation determines the locations of the grown structures (the apertures were separated by 20 μm to ensure spatial QD isolation for the optical experiments). GaN nanowires were then grown (~100 nm diameter, ~1 μm in length) before being capped with $Al_{0.8}Ga_{0.2}N$. Next a short growth period resulted in the formation of a GaN QD at the nanowire apex, which was then once again capped with $Al_{0.8}Ga_{0.2}N$. Additional growth information can be found in ref 17.

Optical Experiments. Optical experiments were performed using conventional microphotoluminescence spectroscopy. The sample was held in a cryostat complete with a temperature controller such that the temperature could be varied and held stable from 4 to 500 K. A frequency-tripled femtosecond-pulse tunable Ti:Al_2O_3 laser at ~247 nm (200 fs pulses at 80 MHz) focused obliquely onto the sample at ~60° to normal was used for optical excitation, and the emission from individual structures was collected using a piezo-mounted 50× magnification objective lens with a numerical aperture of 0.4. The laser wavelength corresponds to an energy of ~5 eV, which is less than the band gap of the $Al_{0.8}Ga_{0.2}N$ barrier material (~5.6 eV). Excitation of carriers into the dot thus occurs via the continuum states that arise due to the 0D−2D transition from dot to wetting layer. Even though the structures are spatially isolated by design, a confocal spatial filter (pinhole) was used to further cut out any stray background emission from neighboring structures and alignment markers. The emission was analyzed using a 300 mm spectrometer equipped with a liquid-nitrogen-cooled charge coupled device (CCD) camera and a 1200 mm^{-1} reflection grating. Autocorrelation was performed using a Hanbury-Brown & Twiss setup consisting of two photomultiplier tubes (PMTs) especially designed for use in the ultraviolet, a 50/50 beamsplitter, and timing electronics to register coincidence counts. The exit slit of the spectrometer was used as a spectral filter with tunable bandwidth in order to filter the emission before measurement. The correction of the $g^{(2)}[0]$ value was performed as

$$(g^{(2)}_{\mathrm{meas}}[0] - 1) = \rho^2(g^{(2)}_{\mathrm{corr}}[0] - 1) \quad (2)$$

where ρ, corresponding to the ratio between the signal from the QD emission and the total measured light within the spectral window, is estimated to be ~0.84 for the spectrum shown in Figure 1.

AUTHOR INFORMATION

Corresponding Authors

*E-mail: holmes@iis.u-tokyo.ac.jp.
*E-mail: arakawa@iis.u-tokyo.ac.jp.

Author Contributions

M.J.H. performed the optical experiments with assistance from S.K, carried out the data analysis, and prepared the manuscript with contributions from all authors. K.C. and M.A. performed

DOI: 10.1021/acsphotonics.6b00112
ACS Photonics 2016, 3, 543−546

the sample fabrication. Y.A. conceived and supervised the entire project.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was supported by the Project for Developing Innovation Systems of the Ministry of Education, Culture, Sports, and Technology (MEXT), Japan, and by JSPS KAKENHI projects 15H05700 and 15H06141. The authors acknowledge insightful discussions with S. Iwamoto and S. Sergent and the assistance of G. Kang and F. Le Roux.

■ REFERENCES

(1) O'Brien, J. L.; Furusawa, A.; Vuckovic, J. Photonic quantum technologies. *Nat. Photonics* **2009**, *3*, 687−695.
(2) Kuhn, A.; Hennrich, M.; Rempe, G. Deterministic Single-Photon Source for Distributed Quantum Networking. *Phys. Rev. Lett.* **2002**, *89*, 067901.
(3) Lounis, B.; Moerner, W. E. Single photons on demand from a single molecule at room temperature. *Nature* **2000**, *407*, 491.
(4) Mizuochi, N.; Makino, T.; Kato, H.; Takeuchi, D.; Ogura, M.; Okushi, H.; Nothaft, M.; Neumann, P.; Gali, A.; Jelezko, F.; Wrachtrup, J.; Yamasaki, S. Electrically driven single-photon source at room temperature in diamond. *Nat. Photonics* **2012**, *6*, 1−5.
(5) Castelletto, S.; Johnson, B. C.; Ivády, V.; Stavrias, N.; Umeda, T.; Gali, A.; Ohshima, T. A silicon carbide room-temperature single-photon source. *Nat. Mater.* **2013**, *12*, 1−6.
(6) Aharonovich, I.; Castelletto, S.; Simpson, D. A.; Su, C.-H.; Greentree, A. D.; Prawer, S. Diamond-based single-photon emitters. *Rep. Prog. Phys.* **2011**, *74*, 076501.
(7) Michler, P.; Imamoglu, A.; Mason, M. D.; Carson, P. J.; Strouse, G. F.; Buratto, S. K. Quantum correlation among photons from a single quantum dot at room temperature. *Nature* **2000**, *406*, 968.
(8) He, Y.-M.; He, Y.; Wei, Y.-J.; Wu, D.; Atatüre, M.; Schneider, C.; Höfling, S.; Kamp, M.; Lu, C.-Y.; Pan, J.-W. On-demand semiconductor single-photon sourcewith near-unity indistinguishability. *Nat. Nanotechnol.* **2013**, *8*, 213−217.
(9) Arakawa, Y.; Sakaki, H. Multidimensional quantum well laser and temperature dependence of its threshold current. *Appl. Phys. Lett.* **1982**, *40*, 939.
(10) Yoshie, T.; Scherer, A.; Hendrickson, J.; Khitrova, G.; Gibbs, H. M.; Rupper, G.; Ell, C.; Shchekin, O. B.; Deppe, D. G. Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity. *Nature* **2004**, *432*, 200.
(11) Hennessy, K.; Badolato, A.; Winger, M.; Gerace, D.; Atatüre, M.; Gulde, S.; Fält, S.; Hu, E. L.; Imamoglu, A. Quantum nature of a strongly coupled single quantum dot−cavity system. *Nature* **2007**, *445*, 896−899.
(12) Nomura, M.; Kumagai, N.; Iwamoto, S.; Ota, Y.; Arakawa, Y. Laser oscillation in a strongly coupled single-quantum-dot-nanocavity system. *Nat. Phys.* **2010**, *6*, 279−283.
(13) Fedorych, O.; Kruse, C.; Ruban, A.; Hommel, D.; Bacher, G.; Kummell, T. Room temperature single photon emission from an epitaxially grown quantum dot. *Appl. Phys. Lett.* **2012**, *100*, 061114.
(14) Bounouar, S.; Elouneg-Jamroz, M.; Hertog, M. d.; Morchutt, C.; Bellet-Amalric, E.; Andre, R.; Bougerol, C.; Genuist, Y.; Poizat, J. P.; Tatarenko, S.; Kheng, K. Ultrafast room temperature single-photon source from nanowire-quantum dots. *Nano Lett.* **2012**, *12*, 2977−2981.
(15) Holmes, M. J.; Choi, K.; Kako, S.; Arita, M.; Arakawa, Y. Room-temperature triggered single photon emission from a III-nitride site-controlled nanowire quantum dot. *Nano Lett.* **2014**, *14*, 982−986.
(16) Deshpande, S.; Frost, T.; Hazari, A.; Bhattacharya, P. Electrically pumped single-photon emission at room temperature from a single InGaN/GaN quantum dot. *Appl. Phys. Lett.* **2014**, *105*, 141109.
(17) Choi, K.; Arita, M.; Kako, S.; Arakawa, Y. Site-controlled growth of single GaN quantum dots in nanowires by MOCVD. *J. Cryst. Growth* **2013**, *370*, 328−331.
(18) Kako, S.; Santori, C.; Hoshino, K.; Götzinger, S.; Yamamoto, Y.; Arakawa, Y. A gallium nitride single-photon source operating at 200K. *Nat. Mater.* **2006**, *5*, 887−892.
(19) Kako, S.; Holmes, M. J.; Sergent, S.; Bürger, M.; As, D. J.; Arakawa, Y. Single-photon emission from cubic GaN quantum dots. *Appl. Phys. Lett.* **2014**, *104*, 011101.
(20) Toda, Y.; Moriwaki, O.; Nishioka, M.; Arakawa, Y. Efficient Carrier Relaxation Mechanism in InGaAs/GaAs Self-Assembled Quantum Dots Based on the Existence of Continuum States. *Phys. Rev. Lett.* **1999**, *82*, 4114.
(21) Holmes, M. J.; Kako, S.; Choi, K.; Podemski, P.; Arita, M.; Arakawa, Y. Probing the Excitonic States of Site-Controlled GaN Nanowire Quantum Dots. *Nano Lett.* **2015**, *15*, 1047.
(22) Takagahara, T. Electron-phonon interactions and Excitonic Dephasing in Semiconductor Nanocrystals. *Phys. Rev. Lett.* **1993**, *71*, 3577.
(23) Krummheuer, B.; Axt, V. M.; Kuhn, T.; D'Amico, I.; Rossi, F. Pure dephasing and phonon dynamics in GaAs- and GaN-based quantum dot structures: Interplay between material parameters and geometry. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2005**, *71*, 235329.
(24) Guimard, D.; Lee, H.; Nishioka, M.; Arakawa, Y. Growth of high-uniformity InAs/GaAs quantum dots with ultralow density below $10^7 cm^{-2}$ and emission above 1.3μm. *Appl. Phys. Lett.* **2008**, *92*, 163101−4.
(25) Hönig, G.; Callsen, G.; Schliwa, A.; Kalinowski, S.; Kindel, C.; Kako, S.; Arakawa, Y.; Bimberg, D.; Hoffmann, A. Manifestation of unconventional biexciton states in quantum dots. *Nat. Commun.* **2014**, *5*, 5721.
(26) Renard, J.; Songmuang, R.; Bougerol, C.; Daudin, B.; Gayral, B. Exciton and biexciton luminescence from single GaN/AlN quantum dots in nanowires. *Nano Lett.* **2008**, *8*, 2092−2096.
(27) Choi, K.; Kako, S.; Holmes, M. J.; Arita, M.; Arakawa, Y. Strong exciton confinement in site-controlled GaN quantum dots embedded in nanowires. *Appl. Phys. Lett.* **2013**, *103*, 171907.
(28) Buckley, S.; Rivoire, K.; Vuckovic, J. Engineered quantum dot single-photon sources. *Rep. Prog. Phys.* **2012**, *75*, 126503.
(29) Borri, P.; Langbein, W.; Schneider, S.; Woggon, U.; Sellin, R. L.; Ouyang, D.; Bimberg, D. Ultralong Dephasing Time in InGaAs Quantum Dots. *Phys. Rev. Lett.* **2001**, *87*, 157401.
(30) Besombes, L.; Kheng, K.; Marsal, L.; Mariette, H. Acoustic phonon broadening mechanism in single quantum dot emission. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2001**, *63*, 155307.
(31) Tomić, S.; Pal, J.; Migliorato, M. A.; Young, R. J.; Vukmirović, N. Visible Spectrum Quantum Light Sources Based on $In_xGa_{1-x}N$/GaN Quantum Dots. *ACS Photonics* **2015**, *2*, 958−963.
(32) Kim, J.-H.; Ko, Y.-H.; Gong, S.-H.; Ko, S.-M.; Cho, Y.-H. Ultrafast single photon emitting quantum photonic structures based on a nano-obelisk. *Sci. Rep.* **2013**, *3*, 2150.

DOI: 10.1021/acsphotonics.6b00112
ACS Photonics 2016, 3, 543−546