Room-Temperature Triggered Single Photon Emission from a III-Nitride Site-Controlled Nanowire Quantum Dot

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Ⓢ Supporting Information

ABSTRACT: We demonstrate triggered single photon emission at room temperature from a site-controlled III-nitride quantum dot embedded in a nanowire. Moreover, we reveal a remarkable temperature insensitivity of the single photon statistics, and a $g^{(2)}[0]$ value at 300 K of just 0.13. The combination of using high-quality, small, site-controlled quantum dots with a wide-bandgap material system is crucial for providing both sufficient exciton confinement and an emission spectrum with minimal contamination in order to enable room temperature operation. Arrays of such single photon emitters will be useful for room-temperature quantum information processing applications such as on-chip quantum communication.

KEYWORDS: Nanowire, quantum dot, single photon source, III-nitride, room temperature
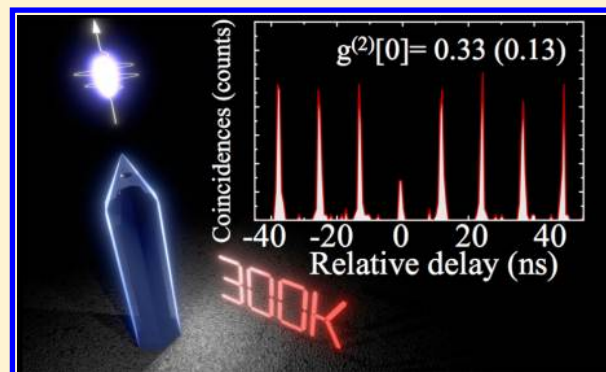
One of the ultimate goals of semiconductor nano-technology, on the way to realizing solid-state quantum information processing, is the development of a site-controlled triggered single-photon emitter that can operate at room temperature. Such a device could be positioned or fabricated into arrays for use in long distance quantum key distribution, on-chip quantum communication, and as a source of on-demand flying qubits for quantum computing.

It has long been suggested that quantum dots[1] (QDs) made from the III-nitride material system could be used as such devices, as they have sufficient band offsets to provide adequate exciton confinement at high temperatures,[2,3] and it is also possible to fabricate them into predefined arrays.[4] Furthermore, the material system itself has several beneficial properties, which include being highly stable[5] (both chemically and thermally), exhibiting a large breakdown voltage,[6] and offering the potential for photon emission from the ultraviolet, across the entire visible, and into the infrared regions of the spectrum,[7] making it an ideal choice for a variety of optoelectronic and electronic applications.

However, due to issues with material quality, there have been no experimental verifications of single photon emission at room temperature from nitride QDs, let alone from those that have had their physical location predetermined. Indeed, while there have been reports of room temperature single photon emission from color centers in diamond,[8] molecules,[9] and from quantum dots,[10,11] there have been no reports of site-controlled QD-based single photon emitters at room temperature. Here, due to recent advances in the fabrication of high-quality site-controlled GaN QDs in nanowires,[12,13] we show that GaN QDs can

indeed be used as room-temperature-operating single photon sources, thus realizing a basic building block for future quantum information applications. Moreover, we reveal the interesting property that the photon statistics are insensitive to temperature, in contrast to other QD-based triggered single photon emitters which exhibit a pronounced degradation at elevated temperatures.[10,14−17]

The structures used in this study were grown by selective area metal−organic chemical vapor deposition (MOCVD) on sapphire (0001) substrates covered with a 25 nm layer of (low temperature) AlN. A 25 nm SiO_2 layer was sputtered onto the substrate surface, and arrays of apertures (25 nm in diameter) were created using electron beam lithography and reactive ion etching. These apertures mark the location for epitaxial growth of GaN nanowires, which were formed at a pressure of 76 Torr and with V/III ratios about 2 orders of magnitude lower than those typically used for GaN thin-film growth.[18,19] A layer of $Al_xGa_{1−x}N$ (with a nominal mole fraction of $x = 0.8$) was grown around the nanowires to form a core−shell type structure, onto which a GaN QD was grown by a short 8 s GaN growth step. Finally, the structures were capped with an additional layer of AlGaN. The complete process results in the total site-control of each QD in a three-dimensional space, as the lateral (XY) position is defined by the nanowire location, and the distance from the substrate (Z) is controlled by the nanowire height (∼700 nm).
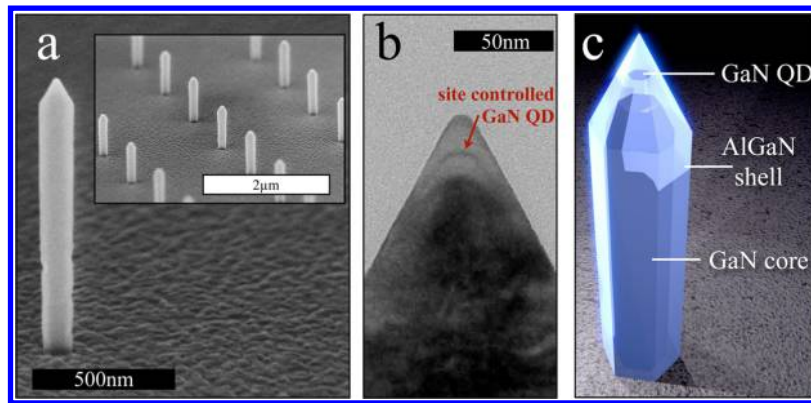
Figure 1. Images of site-controlled nanowire-QDs. (a) SEM image showing a single nanowire grown on a patterned SiO$_2$ substrate by selective area MOCVD. The inset shows an array of nanowires separated by 2 μm (a spacing of 20 μm was used for the optical experiments). (b) TEM image clearly showing the formation of a single QD near the tip of a single nanowire. (c) Schematic of a nanowire containing a single QD.
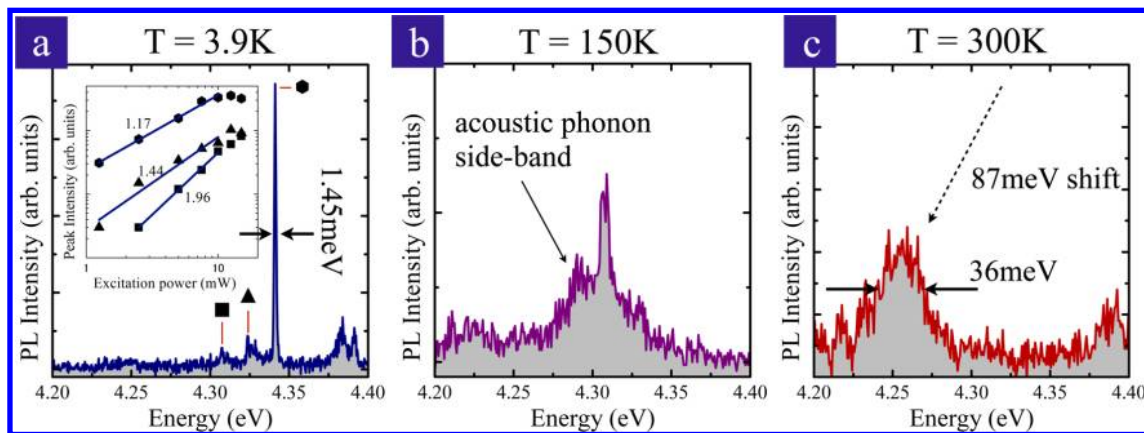

Figure 2. Emission spectra from a single nanowire-QD measured at 3.9 K (a), 150 K (b), and 300 K (c) (measured with excitation powers of 5 mW, 10 mW, and 15 mW, respectively). The inset in (a) shows the power dependence of the peak intensity of the three peaks labeled with a hexagon, triangle, and square, respectively.

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images of nanowires containing single quantum dots, along with a schematic diagram of a structure, are presented in Figure 1. We estimate that the QDs have a height of ~1 nm and a width of ~10 nm: much smaller than typical self-assembled GaN QDs which are usually 2−5 nm in height and can be greater than 20 nm in diameter.[7,20−23] The use of site-controlled nanowire-QDs is crucial for facilitating the optical inspection of a single device at a known location and offers additional benefits such as a high crystal quality due to strain relaxation and enhanced extraction efficiency due to a wave-guiding effect.[24] For the purpose of enabling the optical excitation and measurement of single devices, the nanowires were produced in a regular array separated laterally by distances of 20 μm. The optical properties of the QDs were then measured using microphotoluminescence (μ-PL) spectroscopy via nonresonant excitation with an ultrafast pulsed UV laser as described in the Supporting Information.

The μ-PL spectrum measured from a single device operating at 3.9, 150, and 300 K is presented in Figure 2a, b, and c, respectively. At 3.9 K this particular QD exhibits a low-temperature sharp emission peak at 4.34 eV, with a resolution-limited full width at half-maximum (fwhm) of 1.45 meV (Gaussian fit). In addition to this main peak, we observe a few other peaks with much weaker intensity. The power depend-

ence of these peaks (labeled with a triangle and square), along with that of the main emission peak (labeled with a hexagon) is shown in the inset of Figure 2a. The intensity of the main emission has an approximately linear dependence on the excitation power before saturating at an excitation power of ~15 mW, whereas the additional peaks exhibit a superlinear dependence. This is evidence that the main emission peak is from the exciton ground state and that the lower intensity peaks are from exciton complexes of the dot. Indeed, the lowest energy peak could be the biexciton state of the dot, with a biexciton binding energy of 34 meV, in good agreement with previous studies of GaN quantum dots in nanowires.[12] These peaks have a much lower intensity than the main peak, such that they do not impact greatly on the emission spectrum at low excitation powers. The peaks that are observed at 4.38−4.39 eV are spectrally separated enough from the main emission as to not contaminate the spectrum. Their origin remains unknown at present, and they do not appear in every spectrum.

Upon raising the temperature to 300 K (while tracking the QD emission), the luminescence was observed to red-shift by 87 meV and broaden to ~36 meV. This red-shift is due to the well-known temperature dependent shrinkage of the materials' bandgaps,[25] and the broadening is most probably due to an interaction with acoustic phonons.[26] Indeed, the shape of the emission profile at 150 K is very similar to the acoustic-phonon related sideband observed by Ostapenko et al.[26] when they

investigated single QDs by cathodoluminescence. Neither the shift of the emission line or the line broadening are expected to directly affect the photon statistics, as long as a single state can be resolved.

The high photon emission energy from the QDs is due to the large confinement energy generated in such small dots. High energy UV emitters may be of additional interest as their short wavelength may enable solar-blind operation (i.e., with no background from solar radiation at the earth's surface) and will also facilitate the use of smaller transmission and receiving optics. In fact, the use of small quantum dots provides several additional favorable properties, such as an increase in the exciton binding energy,[27] biexciton binding energy,[12] and a reduction in the radiative lifetime induced by an increased oscillator strength of the tightly confined exciton[28] (or more accurately: due to a suppression of the internal-field induced reduction in oscillator strength found in nitride QDs[22]). We note, however, that if the dots become too small there may be a reduction of the oscillator strength due to a decrease in the coherence volume.

Owing to the fabrication method of the devices, one might expect some spectral contamination from quantum well (QW) luminescence due to a structure that forms on the nanowire sidewall during the QD growth (see the TEM image in Figure 1). However, in the present case, we estimate the thickness of such a QW to be ∼0.5 nm, such that the emission energy is much larger than that of the QDs, and therefore not visible in the measured spectra.

To confirm single photon emission, the photoluminescence from the dots was filtered and directed to a Hanbury-Brown and Twiss (HBT) setup consisting of two photomultiplier tube (PMT) detectors and a 50/50 beam splitter operating at the appropriate wavelength range. A histogram of the relative delay, τ, between photon counts on the two PMTs could then be acquired in order to ascertain the statistics of the light emitted from the QDs. Coincidence histograms of this sort are proportional to the second order correlation function,

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

where $I(t)$ is the intensity of the emitted light at time t. A measurement of $g^{(2)}(0) < 0.5$ verifies the detection of a single photon (an $n = 1$ Fock state) from a single quantum emitter.[29]

Such histograms of the emission from the single dot at 3.9 K, 150 K, and 300 K are presented in Figure 3a, b, and c, respectively. Due to a decrease in emission intensity of the QD at higher temperatures (the total integrated peak intensity of this dot fell by a factor of ∼5.4 when raising the temperature from 3.9 to 300 K—most likely due to increased nonradiative recombination), the measurement was performed using an excitation power of 5 mW at 3.9 K, 10 mW at 150 K, and 15 mW at 300 K (even at these higher excitation powers, the contamination from excited states is expected to be negligible). The suppression of counts at $\tau = 0$ is clear evidence of the single photon nature of the emission. From the data we calculate the second order correlation function of the peak at zero time delay, $g^{(2)}[0]$, to be 0.26 at 3.9 K, 0.35 at 150 K, and 0.33 at 300 K. These values become 0.12, 0.14, and 0.13, respectively, when corrected for background and detector dark counts.[30] It is possible that the residual nonzero value of $g^{(2)}[0]$ can be explained by fast repopulation and subsequent emission from the dot. Indeed, measurements of the emission lifetime
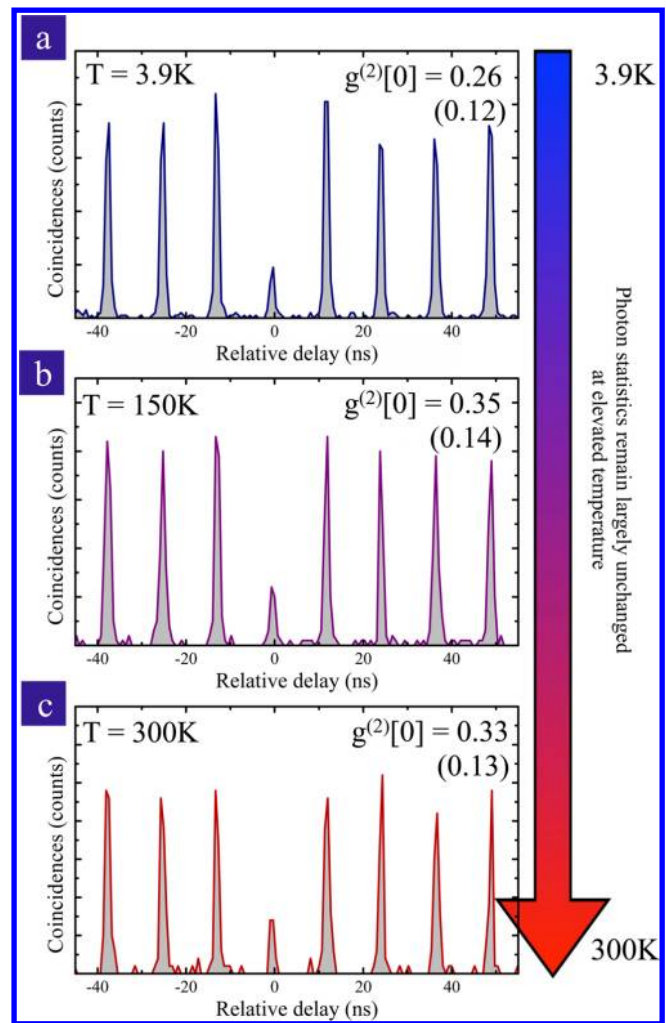

Figure 3. Coincidence counts histograms measured at 3.9 K (a), 150 K (b), and 300 K (c) from the same QD in Figure 2. The $g^{(2)}[0]$ values in parentheses are corrected for background and detector dark counts. The suppressed height of the peak at delay 0 is clear evidence of single photon emission, and the single photon statistics remain largely unaffected by temperature.

(see the Supporting Information) suggest a radiative lifetime of ∼300 ps, which is in good agreement with previous investigations of small GaN QDs,[22,31] and fast enough that some repopulation may occur from a continuum of states related to the dot.[32−34] We expect to be able to suppress this residual in the future via resonant excitation.[13,35,36]

A particularly striking quality of this single photon emission is that there is almost no degradation of the $g^{(2)}[0]$ value with increasing temperature. Degradation of the measurable $g^{(2)}[0]$ for triggered single photon sources at elevated temperatures has been reported in many nonsite-controlled QD systems[10,14−17] and is usually attributed to spectral contamination from thermally broadened excited states or other (spatially close) QDs emitting in the same energy range. In the case of the site-controlled nanowire QDs under investigation here, a relatively contamination-free spectrum is observed even at such high temperatures. The temperature resilience of the photon statistics in the current devices is likely due to a combination of three crucial factors:

1. The use of a wide-bandgap material system such as GaN/AlGaN, which enables exciton confinement at high temperature.
2. The use of site-controlled QDs to reduce the QD density. This acts to eliminate spectral contamination caused by emission overlap from neighboring QDs.
3. The use of small QDs, which exhibit large energy separation between various excited states, preventing significant spectral contamination at high temperatures from states related to the same QD.

This robustness of the single photon statistics could lend these devices to be used in environments with unstable temperature conditions (such as inside computers), where fan- or electronic-cooling could be used to further maintain temperature stability. Operation will eventually require the electrical injection of carriers into the quantum dots, which may also enable denser packing of the nanowires.

Finally, we note that, as well as allowing high-temperature single photon emission, the structure of these devices provides several additional benefits such as a fast radiative lifetime (due to increased exciton oscillator strength in small nitride QDs[28]), which in the present case suggests that operation at GHz frequencies is possible (although at present we detect in the range of ∼2 kHz, which can be corrected for the single photon emission probability,[37] and detection efficiency/throughput of our experimental system to be ∼0.5 MHz). The site-controlled nature provides the potential to independently position, probe, and optically manipulate individual single photon emitters, offering huge advantages over traditional stochastically forming QDs, which must be first detected, marked, and sometimes even relocated before investigation or device fabrication.

As a maturing material system, III-nitride QDs are significant nano structures for future photonic and electronic quantum information processing systems that may operate at room temperature.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Experimental procedures and additional data. This material is available free of charge via the Internet at http://pubs.acs.org/.

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Author Contributions
M.H. and K.C. contributed equally to this work. M.H. performed the optical experiments with assistance from K.C. and S.K, carried out the data analysis with S.K., and prepared the manuscript with contributions from all authors. K.C. and M.A. performed the sample fabrication. Y.A. supervised the entire project.

Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

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