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We report the demonstration of single-photon emission from cubic GaN/AlN quantum dots grown by molecular beam epitaxy. We have observed spectrally clean and isolated emission peaks from these quantum dots. Clear single-photon emission was detected by analyzing one such peak at 4 K. The estimated $g^{(2)}[0]$ value is 0.25, which becomes 0.05 when corrected for background and detector dark counts. We have also observed the single-photon nature of the emission up to 100 K ($g^{(2)}[0] = 0.47$). These results indicate that cubic GaN quantum dots are possible candidates for high-temperature operating UV single-photon sources with the possibility of integration into photonic nanostructures. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4858966]

Generation of non-classical states of light is of fundamental scientific interest in quantum optics, and an important technological challenge in quantum information processing (QIP). Semiconductor quantum dots (QDs) are practical candidates for such applications because of their atomic-like energy structures, optical stability, and capability to be incorporated with other semiconductor structures.[1–3] In particular, III-nitride QDs are promising solid state quantum emitters for use in such optical devices, as their large exciton binding energy and confinement potential may allow such devices to operate at high temperature. Indeed, there are several reports on single-photon emission at high temperatures from III-nitride QDs.[4–6] However, all of these demonstrations utilized polar QDs, which, in general, suffer from the presence of a giant built-in electric field that may act to degrade device performance.[4,7]

The thermodynamically-stable crystallographic phase of III-nitride semiconductors is the hexagonal, Wurtzite (WZ), crystal structure. As a consequence, most of the research has been conducted on III-nitride semiconductors with this WZ structure. However, this structure exhibits a spontaneous polarization oriented along the polar axis, and is piezoelectric in nature. As a result, polar III-nitride semiconductor heterostructures with WZ crystal structure have a giant built-in electric field along the c-axis, due to the combination of the spontaneous and piezoelectric polarizations. The reduction/suppression of this built-in electric field is the main motivation for growing and studying nonpolar heterostructures.[8]

Nonpolar GaN QDs, both with a cubic (Zinc-Blende) structure and a hexagonal crystal structure (using non-polar planes), have been successfully grown by plasma-assisted molecular beam epitaxy (PA-MBE).[8,9] These QDs are expected to be less sensitive to their environment, and exhibit shorter radiative lifetimes because of the absence of giant built-in electric field along the growth axis.[10,11] Although this has been confirmed experimentally, the single-dot spectroscopy on these nonpolar GaN QDs has been so far quite limited,[12–14] and there has been no report of the generation of non-classical light using such non-polar GaN QDs. Recently, atomically-flat ZB AlN without WZ inclusions has been realized by PA-MBE[15] and has allowed the growth of high quality cubic GaN QDs.[16,17] Here, we report experimental evidence for the short radiative lifetime and single photon emission from the cubic GaN QDs, revealing their potential for use as single photon emitters in QIP applications.

The cubic GaN/AlN QDs were grown by PA-MBE on a 3C-SiC/Si(100) substrate. For the purpose of probing, the emission from an individual QD, sub-micron-scale mesas were processed on the sample. The optical properties of the QDs were then measured using micro-photoluminescence (μ-PL) spectroscopy, with non-resonant excitation, via a frequency-tripled Ti:Sapphire pulsed laser (200 fs pulses at 80 MHz with an excitation wavelength of 266 nm). The laser was focused to an elliptical spot ($10\,\mu$m $\times$ $40\,\mu$m) at a steep angle of 60° to the normal of the sample plane. The luminescence from the QDs was then collected by a microscope objective (N.A. = 0.4). The objective was followed by concave mirrors with a pinhole to spatially filter the collected light, which was then sent to a monochromator equipped with both a liquid-nitrogen cooled CCD camera to analyse the spectrum, and a Hanbury-Brown Twiss (HBT) setup (a 50/50 beamsplitter and two photomultiplier tubes) to perform either time-resolved PL (TRPL) studies, or photon autocorrelation measurements. The sample was held in a continuous flow helium cryostat, which enabled control of the temperature from ~4 K to room temperature.

The emission from the QD ensemble, measured on unpatterned areas of the sample, exhibits a Gaussian distribution centered around 3.5 eV with about 200 meV full-width at half-maximum.[16,18] Isolated single emission lines can be found on the higher energy side of the QD ensemble PL between 3.5 eV and 4.3 eV, when probing sub-micron-scale mesas.[18] Figure 1(a) shows one such emission peak from a single cubic GaN QD at 3.687 eV, with a linewidth of 2.1 meV. The excitation power was 20 mW. This linewidth

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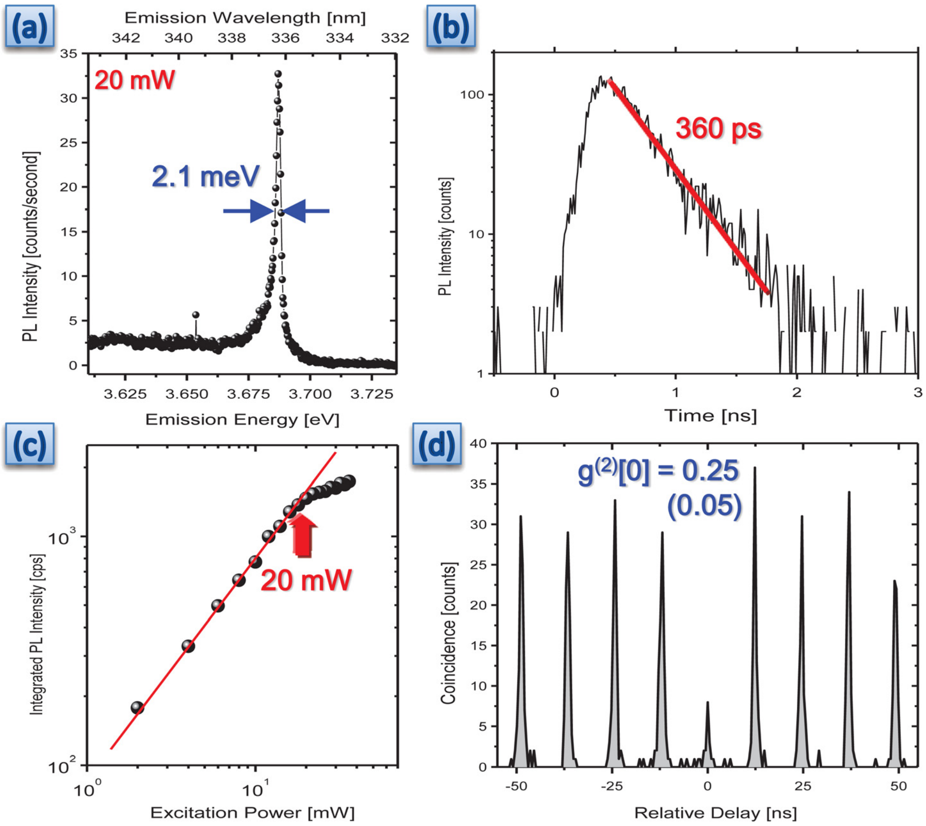

FIG. 1. (a) The emission spectrum from a single cubic GaN QD measured at 4 K with an excitation power of 20 mW. (b) The PL decay trace of this particular dot measured at 4 K. The PL decay time is about 360 ps (No deconvolution; Time resolution ∼230 ps). (c) Excitation power dependence of the QD at 4 K. The red line shows the linear dependence of the PL intensity at excitation powers < 20 mW. (d) Autocorrelation histogram of this particular QD at 4 K. The excitation power was 20 mW as indicated in Fig. 1(a). The number in the bracket is the background corrected value of $g^{(2)}[0]$.

is reasonably broad, most likely due to spectral diffusion, which may be enhanced by the presence of an internal field, and interaction between the exciton in the QD and surface defects on the mesa side walls. We note that the linewidths of cubic QDs exhibit a weak downward trend for increasing emission energies, i.e., decreasing size.[18] This size-dependent spectral diffusion suggests that there exists a residual permanent dipole in ZB GaN QDs, indicating the presence of internal built-in field.

TRPL studies were performed based on time-correlated single-photon counting, with 230 ps time resolution, by using one of the arms of the HBT setup, and a trigger signal from the excitation pulsed laser. Figure 1(b) shows the PL decay trace measured from the same QD shown in Fig. 1(a). The lifetime of this particular QD is ∼360 ps, which is about one order of magnitude shorter than those typically measured from polar GaN QDs emitting at the same energy.[19] The lifetime of 360 ps is in fair agreement with previous reports on cubic GaN QDs.[20] Indeed, measurements of the emission lifetime obtained by filtering various energies of the broad ensemble emission always revealed similar lifetimes (<390 ps),[18] consistent with an absence of a giant built-in electric field in such cubic QDs (i.e., in contrast to polar QDs,[19] the emission lifetime does not depend strongly on QD size).

To confirm the single-photon statistics of the emission from the QDs, we performed photon-autocorrelation measurement using the HBT setup. A histogram of the relative delay ($\tau = t_1 - t_2$) between a photon-detection event in one HBT arm at t_1 and in the other arm at t_2, is proportional to the second order coherence function $g^{(2)}(\tau)$. A measurement $g^{(2)}(0) < 0.5$ verifies the detection of a single photon (an n = 1 Fock state) from a single quantum emitter. Figure 1(d) shows such an autocorrelation histogram, measured at 4 K from the QD emission shown in Fig. 1(a). The suppression of counts at $\tau = 0$ in Fig. 1(d) is clear evidence for the single photon nature of the emission. The accumulation of the coincidence-counts histogram was carried out using an excitation power of 20 mW. The power dependence of the integrated QD emission intensity is shown in Fig. 1(c). The linear dependence suggests that this emission line originates from single exciton recombination. The intensity begins to saturate above an excitation power of 20 mW, whilst the background underlying the QD emission is still observed to increase (see Fig. 1(c)). Therefore, we chose 20 mW as an appropriate excitation power in order to optimize the count rate and signal-to-noise ratio. From the data shown in Fig. 1, we calculate the second order coherence function of the peak at zero time delay, $g^{(2)}[0]$, to be 0.25. This $g^{(2)}[0]$ is the coincidence counts (peak area) for $-T/2 < \tau < T/2$, normalized to the average counts of the surrounding peaks, where T is the repetition period of the pulsed excitation. The $g^{(2)}[0]$ value becomes 0.05 when corrected for background and detector dark counts.[21] The measurement of $g^{(2)}[0] < 0.5$ clearly demonstrates that the emission comes from single quantum state of the QD, even though the emission is relatively broad. The actual measured $g^{(2)}[0]$ value of 0.25 indicates a multi-photon probability suppression of 25% relative to an attenuated laser of the same power. This residual value of 0.25 probably originates from stray luminescence coming from an un-etched area of the sample in conjunction with imperfect spatial filtering.

Figure 2 shows the emission spectrum and the autocorrelation histogram from another cubic QD measured at 100 K. The excitation power was increased to 170 mW in order to compensate for the decrease in the count rate at higher temperatures. The actual measured $g^{(2)}[0]$ value is

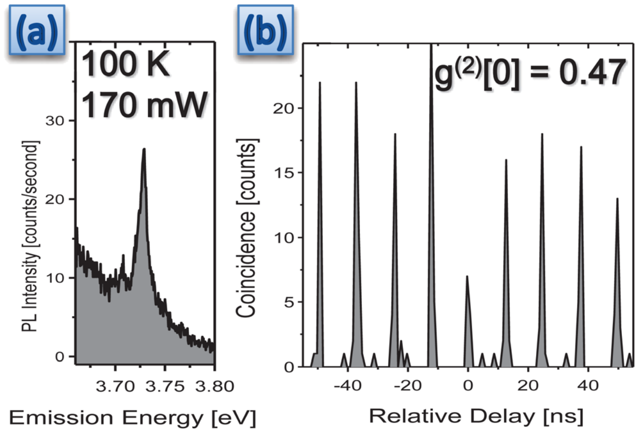

FIG. 2. (a) The emission spectrum and (b) autocorrelation histogram from the cubic QD measured at 100 K. The QD is different from the one shown in Fig. 1. The excitation power was 170 mW.

0.47, which might originate from stray luminescence. Taking account of the level of the background contamination, this $g^{(2)}[0]$ value seems to be an underestimate, probably due to the low count rate and finite integration time. Furthermore, an estimation of the background contamination in the spectrum is difficult, because we cannot appropriately evaluate the contribution of LA-phonon sidebands which typically appear at temperatures above 75 K.[18] However, the suppression of counts at $\tau = 0$ is clear evidence for the single photon nature of the QD emission. This result shows the potential of cubic GaN QDs for use as single-photon emitters that may operate at high temperature.

In summary, we have demonstrated single-photon emission from cubic GaN QDs. The $g^{(2)}[0]$ value is currently limited by background contamination that possibly comes from an un-etched region near to the mesa. The background corrected value of $g^{(2)}[0]$ is 0.05, indicating that a significant improvement of the $g^{(2)}[0]$ may be obtained by using a low QD-density sample with sparser mesa patterning. We have also shown a demonstration of single-photon emission at 100 K, indicating the potential for higher-temperature operation owing to the strong carrier confinement. The possibility of incorporating these QDs with photonic structures is another attractive aspect of the cubic GaN QDs on SiC/Si. Indeed, microdisks containing cubic GaN QDs have already been demonstrated,[22] and it is possible that photonic crystal nanocavities containing cubic GaN QDs may also be fabricated.[23] Using these photonic structures, we could improve the photon collection efficiency via the modification of the radiation pattern. A more interesting and advanced direction of study would be the realization of the Purcell effect, potentially improving the repetition rate and the coherence of the emitted photons.[1]

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