Photon correlation studies of single GaN quantum dots

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We present measurements of the second-order coherence function on emission from single GaN quantum dots. In some cases a large degree of photon antibunching is observed, demonstrating isolation of a single quantum system. For a selected quantum dot, we study the dependence of photon antibunching on excitation power and temperature. Using pulsed excitation, we demonstrate an ultraviolet triggered single-photon source operating at a wavelength of 358 nm.

Quantum dots[1] (QDs) in nitride semiconductors, potentially useful for improving the performance of short-wavelength devices such as light-emitting diodes and lasers[2], also offer exciting possibilities for optical quantum information applications. For example, optically addressed excitonic qubits could exhibit large two-qubit interactions due to the large built-in electric field found in nitride QDs[3]. In addition, with a wide range of possible emission wavelengths, nitride QDs are interesting as single-photon sources for applications such as free-space quantum cryptography[4], where a shorter wavelength could in principle allow for smaller transmitter and receiver telescopes. Another motivation is the potential for higher-temperature devices. Although II-VI compounds have been investigated for this purpose[5], nitride semiconductors are more resistant to degradation, and are suitable for electrically-contacted devices.

Recently, Stranski-Krastanov growth of high-quality GaN QDs on AlN has been demonstrated by metalorganic chemical vapor deposition[6]. Photoluminescence from single GaN QDs has been reported, and discrete spectral peaks have been identified that exhibit excitation power dependencies consistent with exciton and biexciton behavior[7]. Here, we report measurements of the second-order coherence function[8] $g^{(2)}(\tau) = \langle : I(0)I(\tau) : \rangle / \langle I \rangle^2$ on single GaN QDs. For some QDs, strong nonclassical behavior (photon antibunching) is observed, demonstrating isolation of a single anharmonic quantum system. Occasionally, we instead observe bunching on a nanosecond timescale. For a selected QD, we study the dependence of photon antibunching on excitation power and temperature, and demonstrate triggered single-photon generation.

For this study, hexagonal GaN/AlN QDs were grown on a (0001)-oriented 6H-SiC substrate. QDs with an average height of approximately 4 nm, an average diameter of 20 nm and a density of approximately 10^{10} cm^{-2} were grown on top of a 200 nm AlN buffer layer and covered with a 50 nm AlN cap layer. To isolate small numbers of QDs, mesas with diameters from $0.2 - 2\,\mu$m were fabricated by electron-beam lithography.

The sample was cooled to temperatures from 10-100K in a liquid-helium continuous-flow cryostat providing optical access through a thin window. The QD wetting layer was excited at a wavelength of 266 nm using a beam produced by second-harmonic generation with a frequency-doubled Nd:YVO$_4$ laser. This beam was focused from a steep angle to a $\sim 20\,\mu$m spot on the sample. The resulting photoluminescence was collected using an NA=0.6 microscope objective with cover-slip correction, and imaged onto a pinhole that selected a $2\,\mu$m region on the sample. The light was then sent to a cooled-CCD spectrometer or to a Hanbury Brown-Twiss (HBT) photon correlation setup. For the HBT setup, the light was first spectrally filtered using a prism-based monochromator configuration with a transmission band-

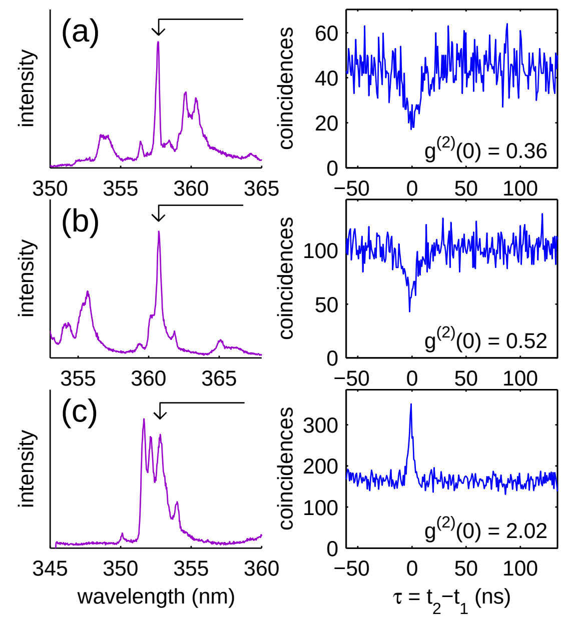

FIG. 1: (a)-(c) Left: Photoluminescence spectra of mesas A, B and C, respectively. The arrows indicate peaks selected for photon correlation measurements on right. Right: Number of measured coincidences vs. relative delay τ between two detectors. The time resolution (bin size) is 0.81 ns. Total count rates on two detectors: (a) 8200 s^{-1}, (b) 8000 s^{-1}, (c) 13300 s^{-1}; temperatures $<$ 20K.

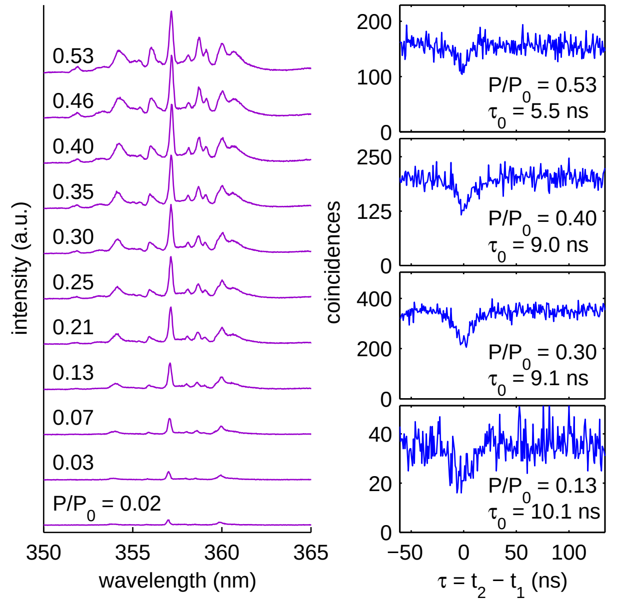

FIG. 2: Left: spectra measured at various excitation powers. Normalized powers P/P_0 are indicated. Right: photon correlation measurements at selected powers.

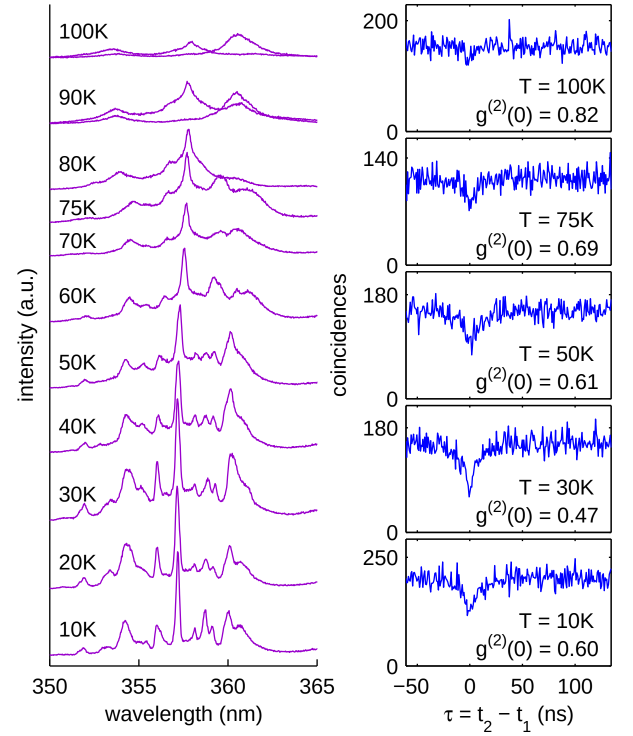

FIG. 3: Left: spectra measured at various temperatures as indicated. At 90K and 100K, blinking behavior was observed, and two representative spectra are plotted. Right: photon correlation measurements at selected temperatures.

width adjustable to $< 1\,\mathrm{nm}$. The light was then split into two paths by a beamsplitter, each path leading to a miniature photomultiplier-tube detector (200 ps time resolution, $50\,\mathrm{s}^{-1}$ dark counts, 20% efficiency). A time-to-amplitude converter followed by a multi-channel-analyzer computer card produced a histogram of the relative delay $\tau = t_2 - t_1$ between photon detections on counters 1 and 2. Integration times were typically at least one hour. A computer-controlled feedback system prevented the sample position from drifting during this time.

Spectroscopy of single mesas showed a wide variety of patterns of spectral peaks. Individual peaks typically showed a large degree of linear polarization along various directions. The spectral linewidths were highly variable. Most of the peaks showed at least a small amount of spectral diffusion on timescales of seconds or longer, and some peaks blinked. The results presented here are from QDs in $0.4\,\mu\mathrm{m}$ mesas selected for bright emission, narrow peaks and relatively stable spectra.

Figure 1 (left) shows photoluminescence spectra from three mesas labeled A, B, and C obtained using excitation powers well below saturation. The corresponding photon correlation data (right) were fitted with the function $G^{(2)}(\tau) = A\left[1 - (1 - g^{(2)}(0))\exp(-|\tau|/\tau_0)\right]$ to obtain the fitting parameters A, $g^{(2)}(0)$ and τ_0. When $g^{(2)}(0) = 0$, this describes a simple two-level system[9] with a spontaneous emission rate Γ, pump rate r, and $1/\tau_0 = \Gamma + r$. Mesa A produced a well-isolated sharp line (FWHM = 0.23 nm) at 357.6 nm. The correlation measurement shows a large degree of photon antibunching with fitting parameter $g^{(2)}(0) = 0.36$. For mesa B, shoulders appear connected to the main emission line at 360.7 nm. Antibunching was observed when the spectral filter was closed narrowly around the main peak, but the antibunching vanished when the filter was adjusted to include the shorter-wavelength emission, which is likely biexcitonic in origin. Mesa C produced a group of three bright spectral peaks. When the longest-wavelength peak was selected, strong photon bunching was observed with $g^{(2)}(0) \approx 2$. When the excitation power was increased, the correlation peak became shorter and narrower. Positive correlations with nanosecond timescales indicate a two-photon cascade process[10] that could occur, for example, if both detectors simultaneously measure exciton and biexciton luminescence.

The excitation power dependence for mesa A is shown in Fig. 2. The normalized excitation powers P/P_0 are indicated, with the parameter $P_0 \approx 10^3\,\mathrm{W\,cm}^{-2}$ obtained by fitting the intensity of the main emission peak with a Poisson distribution model for the single-exciton recombination intensity, $I/I_0 = (P/P_0)\exp(-P/P_0)$. A small peak at slightly shorter wavelength increases nonlinearly with power and may be a biexciton peak. On the right, photon correlation measurements obtained at selected excitation powers are shown, and the fitted antibunching timescale τ_0 is indicated in each case. The timescale changes little at weak excitation powers, but changes rapidly at higher powers. Qualitatively, this agrees with the two-level model above, where $\tau_0 = 1/(\Gamma + r)$ con-

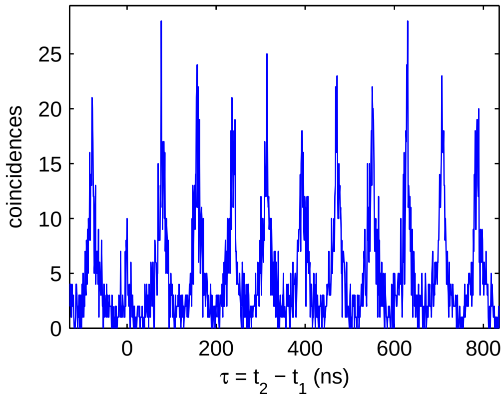

FIG. 4: Photon correlation measurement of mesa A under pulsed excitation. The total count rate was 1400 s^{-1}, and the time resolution (bin size) is 0.97 ns.

verges to $1/\Gamma$ in the weak-excitation limit. At weak excitation power, $\tau_0 \approx 9$ ns, in good agreement with the spontaneous emission lifetime $1/\Gamma = 8$ ns which we have measured independently using a pulsed excitation source.

The temperature dependence for mesa A from $10-100$K is shown in Fig. 3. As the temperature is increased, at first only a background appears to grow in the spectra, but by $50-60$K line broadening appears, possibly from acoustic phonon sidebands[11]. This broadening increases rapidly as the temperature increases further. At $90-100$K, the spectra began to blink with a timescale on the order of $1-10$ s. Two representative spectra from different blinking configurations are shown in these cases. In the photon correlation measurements, the spectral filter width was kept constant, but the center wavelength was adjusted as necessary to follow the main emission line. The antibunching decreases only slightly from $10-50$K, but fades rapidly from $50-100$K, reflecting the broadening in the spectra.

Finally, we used pulsed excitation of mesa A to demonstrate an ultraviolet triggered single-photon source. For such a source, the probability of measuring more than one photon in a given pulse is reduced compared to a Poisson distribution with the same mean photon number. Fig. 4 shows a photon correlation measurement at 10K of mesa A excited with 266 nm excitation pulses, obtained through third-harmonic generation with a Ti-sapphire laser. The pulses had a duration of ~ 2 ps, and the repetition period was reduced to 78.6 ns using a pulse picker. The measurement shows a series of peaks, with the peak at $\tau = 0$ corresponding to events where two photons were detected following the same excitation pulse. When detector dark counts are taken into account, the reduced area of the central peak corresponds to a reduction of the two-photon probability to 0.24 ± 0.03 times that for an equivalent Poisson-distributed source.

These results constitute the first demonstration of photon antibunching and triggered single-photon generation at ultraviolet wavelengths using GaN quantum dots. Devices capable of operating at higher temperatures should be possible if the exciton-biexciton spectral separation can be increased through modification of the quantum-dot size and composition[12]. Another goal will be to decrease the long spontaneous emission lifetimes observed in GaN quantum dots, which have been attributed to built-in electric fields that cause spatial separation of the electron and hole wavefunctions[13,14]. Elimination or reduction of the built-in field is therefore a priority for improving GaN-based single-photon sources. This may also help to decrease the spectral bandwidth, since the built-in field is thought to enhance the interaction with fluctuating electric fields[7].

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