Electronic Supplementary Material (ESI) for Nanoscale.

Nanoscale

ELECTRONIC SUPPLEMENTARY INFORMATION

Polarisation-controlled single photon emission at high temperatures from InGaN quantum dots

T. Wang,[a,†] T. J. Puchtler,[a,†] T. Zhu,[b] J. C. Jarman,[b] L. P. Nuttall,[a] R. A. Oliver[b] and R. A. Taylor[a]

[a.] *Department of Physics, University of Oxford, Parks Road, Oxford, OX1 3PU, UK.*

[b.] *Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge, CB3 0FS, UK.*

† *E-mails: tong.wang@physics.ox.ac.uk; tim.puchtler@physics.ox.ac.uk*

QD PERFORMANCE AT 250 K

PL Characteristics and QD:Background Estimation

The strong confinement and exciton binding energy of nitrides allow us to observe QD emission at temperatures higher than 220 K. Fig. S1 displays the μ-PL spectrum of the studied dot at 250 K (−23 °C), a temperature even closer to ambient conditions, and easier to reach for on-chip thermoelectric cooling. This is the highest operation temperature of a-plane InGaN QDs ever reported. Despite significant carrier recombination through non-radiative pathways, the sharp emission feature of the QD emerges from the QW background with a peak intensity of around 50 cts/s in our μ-PL system. Under the same excitation power, the integrated intensity of the QD remains ~ 6% of that at 4.7 K, which again shows much slower thermal quenching of emission comparing to arsenide systems. At this temperature, the QD energy has redshifted to ~ 2.53 eV. Spectral diffusion induced broadening has also increased the QD linewidth to 24.8 ± 0.5 meV. Whilst a Voigt function that combines a Gaussian and a Lorentzian might provide a more accurate fitting, a single Gaussian profile, as shown in Fig. S1, does indeed model the experimental data well enough. This is because the true Lorentzian linewidth becomes negligible comparing to the actual linewidth at such high temperatures. The underlying QW emission has been fitted to another Gaussian with a much wider linewidth of ~ 80 meV.

As an example of the background estimation method used in the main manuscript, the integrated intensity of the QD and the background QW, within the spectral window indicated in Fig. S1, have been calculated. The ratio of the QD intensity to the total intensity, ρ, can hence be estimated accordingly. In the 250 K case, ρ has been estimated to be ~ 57%. However, the difficulty in ascertaining the exact spectral coverage to be included in the intensity integration is a limitation of this

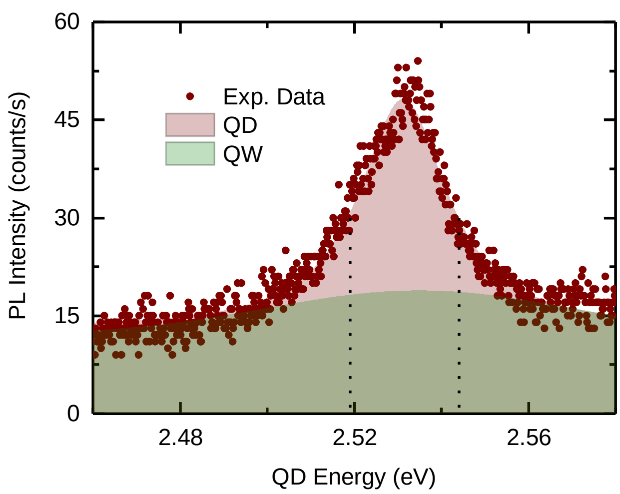

Fig. S1 μ-PL of the studied QD at 250 K. The sharp QD emission has been fitted to a Gaussian, and the QW to a different Gaussian. The dotted lines show the spectral window with which the integrated intensities of the QD and QW emission have been estimated.

estimation method. The band pass filters used in the spectral selection do not have perfect boxcar-like transmission profiles, adding uncertainty to the background estimation.

Photon Antibunching

The emission of the studied QD at 250 K has been passed to the HBT setup of the optical system, and the photon autocorrelation data recorded in Fig. S2. Despite the large amount of linewidth broadening and ~ 43% of background QW emission, the studied QD shows antibunching behaviour with a raw $g^{(2)}(0)$ value of 0.73. Although this uncorrected result is more than 0.5, and thus not sufficient to directly provide evidence for single-photon generation, the $g^{(2)}(0)$ value can be corrected with Eq. (4) in the main text. With a ρ of 57% as estimated earlier, the corrected $g^{(2)}(0)$ becomes 0.17, thereby indicating that the QD is indeed emitting as a single photon source at 250 K in the presence of underlying background QW emission. However, the uncorrected $g^{(2)}(0)$ should still be treated as the main performance indicator of the QD, and background reduction techniques would be needed for cleaner single photon emission at such elevated temperatures. 250 K is hence the highest temperature at which photon antibunching has been observed for *a*-plane InGaN/GaN QDs.

Radiative Lifetime

Time-resolved μ-PL has also been performed on the same QD to measure its radiative lifetime at 250 K. The fitting in Fig. S3 is performed using a modified Gaussian function, convolved with an exponential decay, as described in the main text. The exponential decay constant has been found to be 285 ± 11 ps, indicating similarly strong electron and hole wavefunction overlap even at 250 K, and thus ultrashort radiative lifetime. Again, the decrease in measured radiative lifetime is attributed to the even stronger non-radiative recombination at 250 K. The measured radiative lifetime agrees with expected values of *a*-plane InGaN/GaN QDs. As such, we have shown fast antibunched photon generation from the studied QD at 250 K.

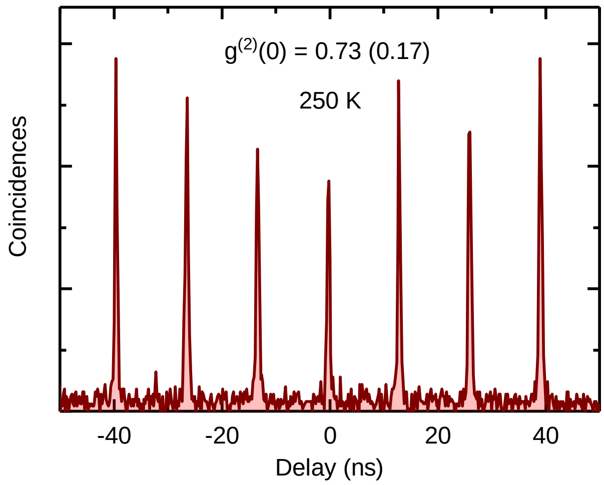

Fig. S2 Autocorrelation data from HBT experiments of the studied QD at 250 K. The value in the bracket is the background-corrected $g^{(2)}(0)$ value calculated using the method described in the main text. The raw data provides direct evidence for antibunching behaviour, while the corrected $g^{(2)}(0)$ shows that the QD is emitting as a single photon emitter in the presence of overlapping QW emission.

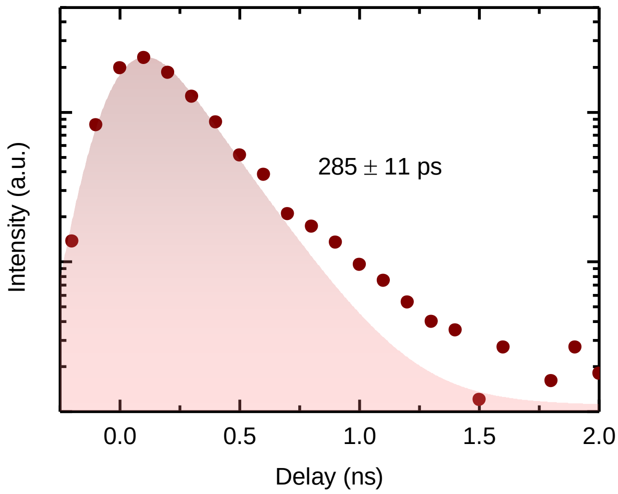

Fig. S3 Radiative lifetime measurements with time-resolved μ-PL at 250 K. Time-correlated single photon counting data have been fitted with a Gaussian function convolved with an exponential decay. The lifetime calculation shows fast photon emission at 250 K.