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Polarized single photon emission and photon bunching from an InGaN quantum dot on a GaN micropylramid

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We report on excitonic single photon emission and biexcitonic photon bunching from an InGaN quantum dot formed on the apex of a hexagonal GaN micropylramid. An approach to suppress uncorrelated emission from the pyramid base is proposed, a metal film is demonstrated to effectively screen background emission and thereby significantly enhance the signal-to-background ratio of the quantum dot emission. As a result, the second order coherence function at zero time delay $g^{(2)}(0)$ is significantly reduced (to $g^{(2)}(0) = 0.24$, raw value) for the excitonic autocorrelation at a temperature of 12 K under continuous wave excitation, and a dominating single photon emission is demonstrated to survive up to 50 K. The deterioration of the $g^{(2)}(0)$ -value at elevated temperatures is well understood as the combined effect of reduced signal-to-background ratio and limited time resolution of the setup. This result underlines the great potential of site controlled pyramidal dots as sources of fast polarized single photons.

1. INTRODUCTION

Generation of non-classical light is both of fundamental interest and a common condition for quantum information applications (QIA). Semiconductor quantum dots (QDs) are feasible photon emitters for QIA due to their atomic-like energy structure and their possibility to be integrated with other semiconductor devices on the same chip. Site-controlled QDs operating close to room temperature are demanded for widespread applications, and linearly polarized emitters are a prerequisite for certain QIA [1, 2].

III-nitride QDs have attractive properties in terms of deep confinement potentials needed for high temperature operation, whilst the small split-off energy of the nitrides causes a high degree of linear polarization already for weakly asymmetric quantum dots [3]. The demonstration of single photon emission at room temperature was recently reported for a GaN QD [4]. Also single photon biexcitonic emission from an InGaN QD in a nanowire at 200 K [5] and an electrically driven InGaN QD in a nanowire exhibiting single photon excitonic emission at 150 K [6] have been reported.

A promising approach for fabrication of site-controlled nitride-based QDs is to deposit a thin layer of InGaN on top of hexagonal GaN micropylramids [7]. QDs formed on the apex of the pyramids grown with this approach have been shown to exhibit single and sharp InGaN related emission lines with a high degree of linear polarization. A simple elongation of the pyramid base gives control of the polarization direction [8, 9]. The photon statistics of these pyramidal QDs have recently been shown to exhibit anti-bunching, but an spectrally overlapping background emission of uncorrelated photons will vanish high-performance single photon characteristics of the

emission [10]. Cathodoluminescence measurements have revealed that the broad background emission mainly originates from the bottom edges near the pyramid base [9].

A remedy to this problem of background emission is proposed by adding a post growth process to cover the lower parts of the pyramid sides as well as the area between the pyramids with a metal film. This reduces the background emission and largely improves the relative QD signal, $\rho = \frac{S}{S+B}$, where S (B) is the measured signal (uncorrelated background) intensity. As a result, significantly improved single photon characteristics were demonstrated.

2. EXPERIMENTAL DETAILS

The structure used in this investigation was grown by a horizontal low pressure hot-wall metal-organic chemical vapor deposition system. Selective area growth of GaN pyramids was performed on a lithographically patterned SiN masked GaN/AlN/SiC template [8]. The pyramids were covered by a thin layer of InGaN, from which nanoscopic 3D islands in a Stranski-Krastanov-like growth mode will be formed on the small (0001) top surface of slightly truncated pyramids. Finally, a capping layer of GaN was grown [7]. The resulting pyramids have a base diameter of 3 μm and are ordered in square 21×21 arrays with 5.5 μm pitch.

The process to cover the lower parts of the hexagonal GaN pyramids with metal was performed in four steps. First a layer of Al was sputtered on the pyramids with a nominal thickness of 200 nm, referring to the measured thickness on a planar sample, followed by spin coating of Hydrogen Silsesquioxane (HSQ) resist on top of the Al layer. Thermal annealing of HSQ at 470 °C under a protective N₂ gas flow during one hour transferred the HSQ resist into SiO₂. The thickness of HSQ was chosen not to completely cover the pyramids with SiO₂. Finally, the sample was immersed in an Al-etch solution

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for 6 minutes, removing the Al layer from the top part of the pyramids not covered with SiO_2 . In this simple process, which does not rely on conventional photo- or e-beam lithography and crucial alignments, all pyramids were completed with the metal mask simultaneously. The insert of figure 1b shows a scanning electron microscopy (SEM) image of a pyramid after these processing steps.

A 355 nm continuous wave diode pumped solid state laser was used for the measurements. The excitation laser was led through a refractive microscope objective (NA = 0.42) and focused down to a spot size of about $1\text{ }\mu\text{m}$ on the sample, allowing excitation of one single pyramid. The sample was mounted on the cold finger of a continuous flow helium cryostat, with the ability to reach temperatures down to 4 K. The laser power used was in the range of 10 to 160 μW , as measured before the objective. The photons emitted from the sample were collected by the same objective and guided through a monochromator with the focal length of 550 mm, allowing a spectral resolution of 0.28 meV. Micro-photoluminescence (μPL) spectra were recorded, either by a charge coupled device (CCD) or by a photo multiplier tube (PMT) [11] operating in the photon counting mode. Time correlated single photon spectroscopy (TCSPS) measurements were performed in the same setup converted to a Hanbury-Brown and Twiss interferometer (HBT) by a beam splitter in the signal path, guiding half of the signal through another monochromator to a second PMT [12]. Both monochromators allowed filtering of the μPL -signal, before it reached the PMTs, with a band pass of 0.11 nm for the relevant wavelengths. The time differences between the arrival of subsequent photons onto the PMTs were recorded by a TCSPC module with the overall instrument time constant of $\tau_i = 0.12\text{ ns}$. The statistical occurrence of measured time differences were compiled into histograms with the bin width set to 128 ps for all experiments except the one performed at 80 K, for which a larger bin width, 256 ps, was used. All TCSPS measurements were performed after the metallization process. It should be noted that we here have a factor 2.5 higher spectral resolution and six times faster detectors than in our previous work [10].

3. RESULTS AND DISCUSSION

Before the metallization process, the PL-signal from the investigated QD was spectrally overlapping with a strong background emission corresponding to $\rho \lesssim 0.57$. The intensity of this background emission was reduced by a factor of 20 after the metallization, causing an increase of relative signal intensity up to $\rho \approx 0.92$ (see figure 1a). The main reasons for this are that the metal film screens the excitation of the bottom pyramid edges and blocks any remaining background emission from this region. Despite the fact that the absolute intensity of the QD was reduced by a factor 2 after the metallization, possibly due to incomplete etching with remnants of the Al layer,

the QD signal remains relatively strong with ~ 40000 registered counts per second for the dominating emission line. The predominant peak X at low excitation powers is attributed to the single exciton. For higher powers, a second peak XX, downshifted by 4.5 meV vs. X, interpreted as the biexciton, gains intensity and becomes comparable to that of the exciton. A power dependence of X and XX (figure 1b) shows the typical linear and superlinear behaviors expected for the exciton and biexciton, respectively. However, it should be noted that a superlinear power dependence alone cannot be used to strictly link XX to the biexciton, since also trions may exhibit a nearly quadratic power dependence at low temperatures [13]. Both the X and XX peaks do exhibit a high degree of linear polarization (see inserts of figure 2a) along the same polarization direction, indicating that both emission lines originate from the same QD [14]. Besides the X and XX peaks, a third high-energy peak is observed at $\sim 3.246\text{ eV}$. This peak is concluded to originate from another QD as its emission is uncorrelated with X. There is also a weak low energy shoulder on XX but characterized by a polarization direction different from that of X and XX and, therefore, concluded to originate from another QD. Under optimized growth conditions, most pyramids accommodate only one QD each [8] but this particular pyramid reveals spectral features of more than one QD. A small pyramid-to-pyramid variation of the top surface may be the reason for this, as truncated pyramids with intentionally larger top surface are known to accommodate more QDs [9].

The μPL spectrum shown in figure 2a is acquired with a PMT in the photon counting mode, at the same experimental conditions as for the autocorrelation measurements of X shown in figure 2b. The experiment reveals a small value of the second order coherence function $g_{exp}^{(2)}(0)=0.24$, which implies that the emission is dominated by single photons from the investigated QD.

Autocorrelation histograms of X measured at 50 K and 80 K (Figs. 3a and b) demonstrate higher $g_{exp}^{(2)}(0)$ values, but single photon emission still dominates at 50 K with $g_{exp}^{(2)}(0)=0.45$, and clear anti-bunching is observed at 80 K with $g_{exp}^{(2)}(0) = 0.63$. The background level in the spectra measured at elevated temperatures (figures 3c and d) are similar to the low-temperature measurement at 12 K (figure 2a) despite the fact that the excitation power is higher. However, the peak intensity of the QD signal drops significantly as the temperature is elevated while the emission lines broaden.

It was earlier argued that the exciton and biexciton emission lines from the same QD do exhibit almost identical polarization properties [14]. We have here monitored a polarization dependence (inserts of figure 2a) which is in consistence with the nearly identical polarization behavior for the exciton and biexciton. Furthermore, the XX-X cross-correlation histogram (figure 2c) which exhibits the typical asymmetric bunching pattern, that proves biexciton emission, is a strong and direct experimental support for the arguments given in Ref. 14.

FIG. 1. (Color online) a) Two μ PL spectra of the QD recorded with a CCD before (dotted red line) and after (solid blue line) the metallization process. The exciton and biexciton emissions are labeled X and XX in the figure. b) Power dependence of the integrated intensity of X (black circles) and XX (red squares) performed at 25 K. The lines represent the best fit to the low power data with the exponents 1.0 and 1.8, respectively. The insert shows a SEM picture of a hexagonal GaN pyramid with the lower part of its sides covered by Al.

Further analysis of the experimental data requires a mathematical formulation of the autocorrelation second order coherence function $g^{(2)}(\tau)$, as provided in Ref. 15, with correction for the superimposed uncorrelated background emission,

$$g^{(2)}(\tau) = 1 - \rho^2 e^{-|\tau|/\tau_c}, \quad (1)$$

where τ is the time difference between subsequent photons and τ_c is the characteristic anti-bunching time constant given by

$$1/\tau_c = 1/\tau_x + W, \quad (2)$$

with the exciton lifetime τ_x and the effective pump rate W [16, 17]. Convoluting 1 with the temporal impulse response function of the TCSPS apparatus, involving the instrument time constant τ_i , the measured second order autocorrelation function $g_m^{(2)}(\tau)$ can be calculated given the three parameters ρ , τ_x and τ_i by,

$$g_m^{(2)}(\tau) = \frac{1}{2\tau_i} \int_{-\infty}^{+\infty} g^{(2)}(\tau) e^{-|\tau-t|/\tau_i} dt. \quad (3)$$

Finally, for non-zero histogram bin width τ_{BW} , the measured value $g_m^{(2)}[0]$ at zero time difference is obtained by integrating $g_m^{(2)}(\tau)$ symmetrically around zero [10],

$$g_m^{(2)}[0] = \frac{1}{\tau_{BW}} \int_{-\tau_{BW}/2}^{+\tau_{BW}/2} g_m^{(2)}(\tau) d\tau. \quad (4)$$

FIG. 2. (color online) a) μ PL spectrum recorded with a PMT and a photon counter ($\rho = 0.94$ for X). b) Autocorrelation histogram of X with $g_{exp}^{(2)}(0) = 0.24$. c) Cross-correlation histogram of XX and X. The two inserts show the polarization resolved data for X (black squares) corresponding to 84% degree of polarization and XX (red circles) corresponding to 85% degree of polarization. The fitted curves are of the form $I(\theta) = I_{min} \cdot \sin^2(\theta - \varphi) + I_{max} \cdot \cos^2(\theta - \varphi)$, where $I_{min}^X = 0.20$, $I_{max}^X = 0.93$, $\varphi^X = 3.9^\circ$, $I_{min}^{XX} = 0.25$, $I_{max}^{XX} = 0.88$ and $\varphi^{XX} = 1.2^\circ$. The concentric circles have the radii of 0.5.

Here, square brackets are used to distinguish the value $g_m^{(2)}[0]$ obtained by integration over a finite bin width from the corresponding value $g_m^{(2)}(0)$ for an infinitesimal bin width.

The exciton lifetime (τ_x) ultimately sets the upper frequency of the single photon emitter. It is clear from 2 that τ_x can be determined as the extrapolated value of τ_c at zero laser excitation power, corresponding to vanishing pump rate W [17]. For this purpose, TCSPS autocorrelation histograms of X were measured for different excitation powers and τ_c was extracted by fitting the data with 3 using τ_c as the only fitting parameter. The other parameters involved in 3, i.e. the instrument time constant τ_i and the relative exciton signal ρ , are known from independent measurements or instrument settings. Figure 4 shows the linear dependence of the anti-bunching rate ($1/\tau_c$) with increasing laser power, corresponding to an exciton life time of $\tau_x = 0.7$ ns.

A high relative signal ($\rho \approx 1$) is essential for high quality single photon emission, and our results show that ρ is reduced from 0.92 to 0.83, when the temperature is elevated from 12 K to 80 K. However, as already discussed, the value of $g_m^{(2)}[0]$ is also affected by the TCSPS instrument time constant τ_i and the histogram bin width τ_{BW} in ratio with the characteristic anti-bunching time τ_c [10]. The non-vanishing values obtained for $g_{exp}^{(2)}(0)$ are

FIG. 3. (color online) a and b) Autocorrelation histograms of X at 50 K and 80 K, respectively. c and d) μ PL spectra recorded by a PMT and a photon counter at 50 K and 80 K, respectively.

indeed fully explained by 4 when including the contributions from the non-zero background emission (ρ) and the limited instrumental time resolution (τ_i and τ_{BW}). This implies that the investigated QD itself behaves as an essentially ideal single photon emitter up to 80 K, but the influence from the above mentioned parameters radically increases the value of $g_m^{(2)}[0]$ at elevated temperatures.

The main causes for the remaining finite value of $g_m^{(2)}[0]$ will now be discussed for the exciton autocorrelation measurements at different temperatures (figures 2a, 3a and 3b). At 12 K (figure 2a), a curve fit of $g_m^{(2)}(\tau)$ to the experimental data yields $\tau_c = 0.45$ ns, with a corresponding value of $g_m^{(2)}[0] = 0.31$. In this case, the dominating factor for non-zero $g_m^{(2)}[0]$ is the finite instrument time constant. A deconvolution of $g_m^{(2)}(\tau)$, corresponding to $\tau_i = 0$, yields the significantly lower value $g_m^{(2)}[0] = 0.17$, but a reduction of the background emission ($\rho=1$) or reduction of the bin width ($\tau_{BW} = 0$) alone has negligible effect. Thus, faster detectors are needed to further improve the $g_m^{(2)}[0]$ value measured at 12 K.

The corresponding analysis of the data at elevated temperatures yields $\tau_c = 0.27$ ns and $g_m^{(2)}[0] = 0.46$ for 50 K (figure 3a) and $\tau_c = 0.12$ ns and $g_m^{(2)}[0] = 0.69$ for 80 K. For these temperatures, both the uncorrelated background and the instrumental time constant contribute almost equally to the finite value of $g_m^{(2)}[0]$. It should be noted that the influence from the instrumental time constant as well as the bin width becomes successively more significant at higher temperatures when τ_c becomes small.

FIG. 4. (color online) The anti-bunching rate ($1/\tau_c$) extracted from the TCSPS histograms and plotted against the excitation laser power. The error bars define 95% confidence intervals for $1/\tau_c$ of the best curve fit. The exciton lifetime determined as the extrapolated anti-bunching time at zero power is $\tau_x = 0.7^{+0.55}_{-0.28}$ ns, where the error refers to the maximum and minimum gradients of a line within the error bars. The histograms for the first and last data points are shown as inserts, with the fitted curve $g_m^{(2)}(\tau)$ shown as a red solid line. The characteristic anti-bunching times with 95% confidence intervals are $\tau_c = 0.53 \pm 0.17$ ns, $\tau_c = 0.45 \pm 0.09$ ns, and $\tau_c = 0.23 \pm 0.03$ ns, for the successively increased powers.

4. CONCLUSIONS

In summary, we have presented an approach to screen the background emission from GaN micropylramids by a metal film which dramatically improved the single photon characteristics (to $g_{exp}^{(2)}(0) = 0.24$) of the linearly polarized excitonic emission from an InGaN QD located at the apex of a pyramid. Furthermore, we provide the first evidence of a biexciton in this QD system (with 4.5 meV binding energy), as characterized by a typical bunching pattern in the cross correlation between the exciton and biexciton emission lines. Single photon characteristics remain up to 50 K, but the measured second order coherence function at zero delay time significantly increases at elevated temperature, mainly due to a reduced peak intensity of the QD emission that lowers the signal-to-background ratio and the reduced anti-bunching time that makes the limited time resolution more significant.

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