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研究課題名（和文）Efficient Single Photon Emission From III-Nitride Quantum Dots

研究課題名（英文）Efficient Single Photon Emission From III-Nitride Quantum Dots

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研究成果の概要（和文）：短波長(300nm付近)で発光する窒化ガリウム量子ドットに適用するphotonic Bullseye構造のデザイン、作成、及び光学評価を行なった。実際に作成したbullseye構造を用いて、窒化ガリウム量子ドットからの光子取り出し効率を上昇させることに成功するができた。ベストデータでは、実験的に5%以上の取り出し効率(レート＝4.36MHz)を実現することができた（対物レンズの開口数＝0.4の場合）。無加工量子ドット(同じ波長で発光するもの)の理論的な最大光子取り出し効率値と比較すると、四倍程度の取り出し効率を実現することができたこと言える。

研究成果の学術的意義や社会的意義
量子ドット単一光子源は、量子コンピューターや量子鍵配送、真乱数生成という技術の基本素子として有望であるが、発光の取り出し効率は低いという問題がある。本研究では、Photonic Bullseye構造を作成することによって、窒化ガリウム半導体でできた量子ドットからの光子取り出し効率を上昇させることに成功した。窒化ガリウム量子ドットは、高温動作も可能・幅広い波長帯で発光が可能であるため、将来の量子技術の発展には注目されている。将来のデバイスを実現するため、取り出し効率の高いものが必要となる。

研究成果の概要（英文）：Photonic bullseye structures were designed and fabricated in GaN quantum dot samples. Analysis of the fabricated samples reveal that the single photon extraction efficiency was successfully increased by the bullseye structures. The highest extraction efficiency realized during in this study was measured to be >5% (for a lens with numerical aperture equal to 0.4, corresponding to a collection rate of 4.36MHz), which is much greater than the theoretical limit for an unprocessed GaN quantum dot emitting at the same wavelength.

研究分野：quantum materials　量子マテリアル

キーワード：量子ドット　半導体

１．研究開始当初の背景

Single photon emitters are being widely investigated around the world as they are fundamental elements for the realization of optical quantum computing and other quantum technologies such as quantum key distribution.[1] Semiconductor quantum dots (QDs) are ideal single photon emitters, and III-nitride QDs have been attracting particular attention in recent years because they can be used to generate single photons at high temperature[2-4] and over a wide range of wavelengths.[5] However, internal reflection at surface of the material surrounding the quantum dot leads to drastically inefficient photon extraction, and it can be shown that only a few percent of the photons emitted from such a III-nitride quantum dot will actually make it out of the material within a solid angle that can be collected using a microscope objective lens. It is therefore of great importance to investigate methods to increase the photon extraction efficiency from III-nitride quantum dots. One method to increase the photon extraction efficiency is to use a photonic bullseye, which consists of concentric annular regions with periodic variation in refractive index: a circular distributed Bragg reflector (DBR). The Bullseye structure therefore acts to inhibit in-plane propagation of light, and hence directs the luminescence from a quantum dot situated at the center into a narrow cone in the out-of-plane direction.[6] Although devices have been reported in the scientific literature for other materials that emit at longer wavelengths, there have been no reports of the successful realization of such a bullseye structure in the III-nitrides.

２．研究の目的

The aim of this project was to realize single photon emission with an enhanced extraction efficiency from a GaN quantum dot using a photonic bullseye structure.

３．研究の方法

The short wavelengths of GaN/AlN QDs, in particular, make the fabrication of photonic structures difficult as the size of the required features scale with the emission wavelength. Therefore our initial design was to use a $5\lambda/4$ period DBR, which would allow for a certain degree of relaxation in the fabrication conditions, and still provide sufficient reflectivity (albeit with a narrower stop-band). Finite Difference Time Domain (FDTD) simulations were then performed to optimize various structure parameters for operation at a wavelength of 310nm (a typical wavelength for GaN QDs grown at our institute). Figure 1 Shows some example simulations in which the enhancement from the bullseye structure is clear.
As a proof of principle study, an array of bullseye structures was fabricated into a sample of high-density self-assembled GaN/AlN QDs ($10^{14}cm^{-2}$), such that some structures contained quantum dots in the center. Fabrication was performed using electron beam lithography with calibrated dosage and reactive ion etching (RIE) in an Argon and Chlorine atmosphere.

Devices were characterized optically using micro-photoluminescence spectroscopy under cryogenic cooling conditions at a temperature of 9K in order to suppress the effects of phonon-induced linewidth broadening. A confocal microscope with a pinhole for spatial selection was used for these measurements (providing a measurement area of ~2μm). Single photon emission was characterized using a Hanbury Brown and Twiss type setup to measure the intensity autocorrelation, $g^{(2)}(\tau)$, of the emission. A combination of CW and pulsed (200fs pulses at 80MHz) lasers at 266nm were used for sample excitation.

４．研究成果

Device design shows that we can expect extraction efficiencies of up to around 12% for a perfectly fabricated device with 5 concentric trenches. Here we have chosen a numerical aperture of 0.4 for benchmarking, as that is the value of our objective lens in the experimental setup. However, theoretically efficiencies up to around 37% can be expected for lenses with NA=0.7).

Fabrication of the samples was performed as outlined above: A dummy run was performed to calibrate the processing steps, and structures with an optimized design were etched into a QD sample. An image of a fabricated device is shown in Figure 2. The QD emission was characterized to make sure that that the QDs themselves were typical GaN/AlN SK QDs with no special characteristics. The QDs were measured to emit in the wavelength range from 280nm~330nm, and time-resolved PL measurements (see Figure 3a inset) revealed lifetimes that where consistent with values reported in the literature for this wavelength range.

Initial evaluation of extraction efficiency enhancement was performed on ensemble emission properties. Ensemble emission from a bullseye structure and also from an unprocessed area immediately adjacent to the bullseye under the same excitation conditions (CW excitation at 266nm) were compared. The emission from the bullseye structures was clear, provides direct evidence of the extraction efficiency increase. Statistics on 108 bullseye structures measured this way show an average enhancement by a factor of round 6x. However, it must be noted that such a measured enhancement cannot be fully attributed to the extraction enhancement, as the bullseye structures may also be enhancing the absorption.

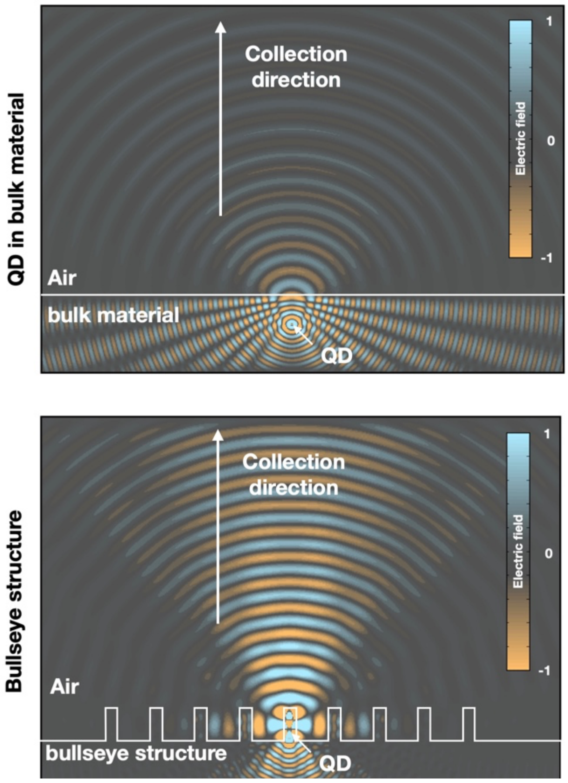

Figure 1: Simulations showing the emission pattern of a QD in bulk material and in a bullseye structure. There is a clear enhancement from the bullseye structure.

This issue can be experimentally overcome by measuring a single-photon-emitting QD under pulsed excitation at saturation conditions, and investigating the ratio of the photon count rate to the excitation rate (80MHz in the case of our Ti:Sapphire laser). Figure 3 shows the emission properties of a sufficiently isolated QD emission peak in a bullseye structure. The auto correlation data of the emission shows clear antibunching with a $g^{(2)}(0)$ value of 0.34: evidence of single photon emission from the QD. Indeed, the background level in the spectrum accounts for about 18% of the emission in the measurement window, and such a value of background would limit the $g^{(2)}(0)$ of an otherwise pure single photon emitter to a value of 0.33,[7] indicating that the quantum dot is indeed emitting single photons with high purity, and the main source of experimental degradation is the background emission.
The photon count rate from the QD, when calibrated for the detection efficiency of the experimental setup and the background counts have been removed, is equal to 2.55MHz. At a saturated excitation of 80MHz this corresponds to an extraction efficiency of 3.19%, which exceeds the limit of a single unprocessed QD under the same conditions (which at this wavelength would exhibit a maximum extraction efficiency of 1% into a lens of NA 0.4).

Table 1 shows the extraction efficiencies of a selection of measured QDs in bullseye structures, along with the measured $g^{(2)}(0)$ values. Note that in this study we have reduced the photon rate to account for the background emission. Therefore, the values shown in the figure represent the true extraction efficiencies of a single QD in the bullseye structure (under the assumption that the QD has an internal quantum efficiency of 100%: i.e., every laser pulse leads to the emission of a photon). Even with this correction, it is clear that the bullseye structures are successfully enhancing the single photon extraction from the QDs. These results represent the first example of top-down fabrication of extraction-enhancing structures for GaN QDs, and have been published in *ACS Photonics*.[8]

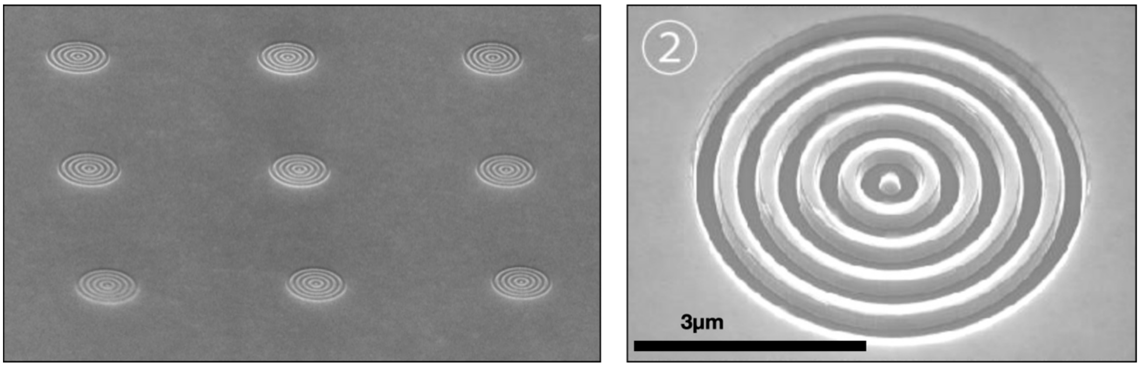

Figure 2. Electron microscope images of fabricated bullseye structure consisting of 5 concentric etched trenches. (a) Array view. (b) High-magnification image of a single bullseye structure.

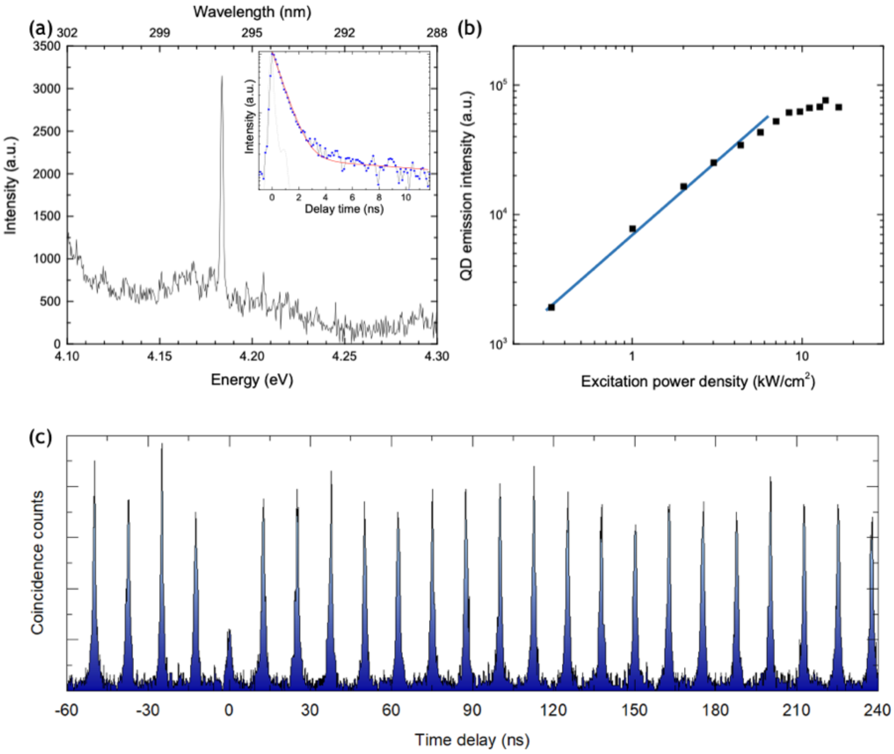

Figure 3 . Optical characterization of a GaN QD in a bullseye structure. (a) Emission spectrum and emission lifetime (inset) of the emitter. (b) Power dependence of the emission intensity, showing a linear increase and saturation. (c) Autocorrelation data with a $g^{(2)}(0)$ value of 0.34.

Sample number	Emission Wavelength	Emission purity [$g^{(2)}(0)$ value]	Emission rate	Extraction efficiency
QD1	296.3nm	0.34	2.55MHz	3.19%
QD2	293.6nm	0.44	4.36MHz	5.45%
QD3	278.2nm	0.29	2.66MHz	3.33%

Table 1. Wavelength, $g^{(2)}(0)$, and emission rate/efficiency data from several measured QDs.

The prototype devices discussed above clearly show the working principle of the structure, and any variations between the measured extraction efficiency and the calculated values can be ascribed to fabrication imperfections and variations from device to device, and also the exact location of the QDs in their respective bullseyes (any deviation from the device center will result in a decrease in efficiency). Such issues may be overcome in future by using a processing technique to pre-select candidate QDs prior to device fabrication, and then form the bullseye directly around these QDs.[6] Another possible method would be the direct fabrication of a site-controlled quantum dot in the central column of the bullseye structure. During this research project we also attempted the fabrication of such site controlled QDs via the etching of a quantum well into nanopillar structures. However, to date our fabricating process resulted in the formation of quantum disks with insufficient lateral quantum confinement. However, we believe that this approach will be successful in future, given further time and appropriate fabrication optimization.

In addition to the experiments outlined above, simulations were also performed to evaluate the theoretical photon extraction efficiency of InGaN/GaN quantum emitters formed in nano-pillar structures. Theoretical extraction efficiencies of ~8% (into a numerical aperture of 0.4) can be achieved from the nano-pillars. This work was published in *Light: Science and Applications.*[9]

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１．著者名 Xia Sijia、Aoki Tomoyuki、Gao Kang、Arita Munetaka、Arakawa Yasuhiko、Holmes Mark J.	４．巻 -
２．論文標題 Enhanced Single-Photon Emission from GaN Quantum Dots in Bullseye Structures	５．発行年 2021年
３．雑誌名 ACS Photonics	６．最初と最後の頁 -
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acsphotonics.1c00032	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 -

１．著者名 Sun Xiaoxiao、Wang Ping、Wang Tao、Chen Ling、Chen Zhaoying、Gao Kang、Aoki Tomoyuki、Li Mo、Zhang Jian、Schulz Tobias、Albrecht Martin、Ge Weikun、Arakawa Yasuhiko、Shen Bo、Holmes Mark、Wang Xinqiang	４．巻 9
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３．雑誌名 Light: Science & Applications	６．最初と最後の頁 1-7
掲載論文のDOI（デジタルオブジェクト識別子） 10.1038/s41377-020-00393-6	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

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〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考
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〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関			
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ドイツ	Leibniz-Institute for Crystal Growth			