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研究課題名（英文）Device Physics of High Temperature Operational Single Photon Emitters

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研究成果の概要（和文）：位置制御されたナノワイヤGaN量子ドットにおいて350K（摂氏７７度）における単一光子発生を世界で初めて実現した。発光自体は400Kまで確認されたが、発光強度の温度依存性を調べた結果、温度が３００Ｋを超えると、発光強度が急に下がることが分かった。更に、ドット内のエキシトンとフォノンとの相互作用の影響で起きる発光線幅の増大も具体的に研究し、他の材料の既存文献データと比較した。ＧａＮには、変形ポテンシャルだけではなく、ピエゾの相互作用メカニズムが存在するため、フォノン相互作用によって起きる発光線幅増大の程度が大きい。

研究成果の概要（英文）：The main results of this research project were the measurement of single photons from a site controlled GaN nanowire quantum dot at an ambient temperature of 350K (77C). The emission itself was measured up to temperatures of 400K, but the intensity was too low to perform autocorrelation measurements. This was the result of a sharp decrease in emission intensity at temperatures above 300K (the origin of which is not fully clear at present). The phonon-induced linewidth broadening was also investigated from 4K to 400K (and the rate of broadening was compared to other materials). At temperatures above 200K phonon wings begin to dominate the emission spectrum, and the linewidth rapidly broadens to values of ~40meV. The phonon interactions are strong in GaN due to the existence of both deformation potential and piezoelectric coupling mechanisms.

研究分野：ナノ量子情報

キーワード：単一光子発生　窒化物ガリウム　量子ドット

１．研究開始当初の背景

The prospect of using single photons to encode information for the secure distribution of cryptographic keys and for quantum computation protocols has led to intensive research into the development of devices that can emit single photons on demand. To date there have been several materials and nanostructures used for the generation of single photons. In particular, III-nitride quantum dots have shown great promise due to the fact that the material system can be used for single photon emission over a range of wavelengths (from the ultraviolet to the infrared), and also operation at relatively high temperatures has been realized (300K at UV, 280K in the red). High temperature operation will remove the necessity of a cryogenic cooling system, thus making a system cheaper to operate. The utilization of such devices in computing racks will require device operation at temperatures of around 350K, and some degree of resilience to fluctuations in temperature.

２．研究の目的

The aim of this project was to elucidate the physics of III-nitride quantum dots towards realizing single photon sources that operate at even higher temperatures (T>300K). High-temperature operating devices (T~350K) would be suitable for use in traditional computer racks, and their realization would be a milestone towards the development of useable quantum information technology. However, as such high temperature operating devices would be a new frontier, it is necessary to fully characterize the temperature dependent emission properties of the III-nitride QDs, such as their linewidths and intensities. Understanding such properties will be crucial to the development of these devices.

３．研究の方法

Experiments were performed on III-nitride based nanowire quantum dots. The main experimental method used for this study was micro-photoluminescence spectroscopy. Nanowire quantum dots (GaN/Al$_{0.8}$Ga$_{0.2}$N, and also InGaN/GaN) were excited using either a continuous wave (λ = 266nm), or pulsed laser (wavelength tunable, 80MHz). Samples were held under vacuum in a helium cryostat so that the temperature could be controlled from temperatures of ~4K to ~450K. Spectroscopy was performed using a 30cm grating spectrometer equipped with a nitrogen-cooled charge coupled device (CCD). For the GaN/AlGaN nanowire quantum dots, the linewidths and emission energy/intensity were characterized over a wide range of temperatures to evaluate the degree of phonon-induced broadening. This was compared to other material systems. Single photon emission statistics were measured using a Hanbury-Brown & Twiss setup consisting of two photomultiplier tube detectors and a beamsplitter (the spectrometer was used as a tunable spectral filter in order to isolate the emission during this measurement).

４．研究成果

The main result attained from this research project was the measurement of single photons from a solid state nanostructure at an ambient temperature of 350K. This proof-of-principle study was the first time that single photon emission was measured at such a high temperature, showing that III-nitride semiconductor nanostructures can be used as single photon emitters even under such extreme conditions. One of the key experimental methods that made this measurement possible was the tuning of the pulsed excitation laser to a wavelength of 247nm, which led to an improved signal to noise ratio, facilitating the single photon collection. This work was published in ACS Photonics. Figure 1 shows an emission spectrum measured at a temperature of 350K from an

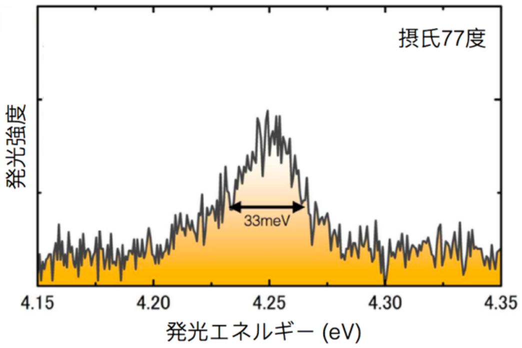

Fig1: Emission spectrum from a GaN nanowire quantum dot measured at 350K.

individual, site-controlled and spatially isolated, nanowire quantum dot. The emission is in the UV at an energy of 4.25eV, which is typical for small GaN quantum dots. The emission linewidth is measured to be ~33meV, a fact which will be discussed later. A lower intensity shoulder peak is present in the low energy side of the emission spectrum. This is related to the biexciton-exciton transition (verified by power dependent spectroscopy as a part of this study) of the quantum dot. This transtion occurs in these quantum dots with a biexciton binding energy (peak separation) of ~40meV.

The measurement of single photons from such a structure requires the isolation of emission related to a single transition in the quantum dot. To that end, it is important to use a spectral filter to remove the low energy shoulder emission in the spectrum. Filtering the center of the emission spectrum using the exit slit of spectrometer provided the required isolation, and enabled the measurement of the intensity autocorrelation shown in figure 2.

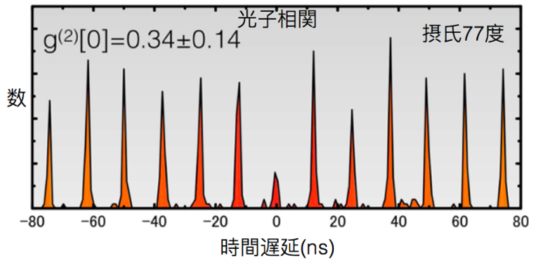

Fig 2. Autocorrelation measurement performed on the peak shown in figure 1. The measured $g^{(2)}[0]$ value of 0.34 is clear evidence of a single photon emission process.

The suppression of coincidence counts at time delay 0 is evidence of the quantum nature of the emission. Indeed, a $g^{(2)}[0]$ value of less than 0.5 is evidence of the single photon nature of the emission. Pure single photon emission would result in a $g^{(2)}[0]$ measurement of zero. In the experiment performed here, the non-zero value of $g^{(2)}[0]$ is due to unfiltered background emission that contaminates the photon purity measured from the quantum dot. It is possible to calculate the effect of the background contamination- leading to a corrected $g^{(2)}[0]$ value of ~0.06, indicating that the quantum dot itself is acting as an almost perfectly pure single photon emitter. However, it is currently not experimentally possible to completely isolate the emission from the background contamination- and it is the measured $g^{(2)}[0]$ value of 0.34 that characterizes the quality of the device. Regardless, this result represents the first time that single photon emission has been measured from a device at an ambient temperature higher than room temperature. Indeed, we note that eggs can be hard boiled at temperatures of 350K (77C).

In addition to the single photon emission measurement, the emission linewidth of the emission from a single quantum dot was investigated over a temperature range of almost 2 orders of magnitude in order to elucidate the broadening mechanism. The basic data is shown in figure 3. It is clear that the lineshape of the emission changes as the temperature is increased, due to the emergence, of phonon wings in the emission spectrum. This broadening has been measured before in other material systems, so a comparison is possible. At comparatively low temperatures (4K ~ 100K) the linewidth grows linearly with increasing temperature: the well-known broadening due to interactions with acoustic phonons (the phonon number at a given energy grows linearly with temperature). We measure the broadening rate in this case to be ~7 μeVK^{-1}. However, at temperatures above 200K, where the phonon wings begin to dominate the spectrum, we can evaluate a linear broadening with a much increased

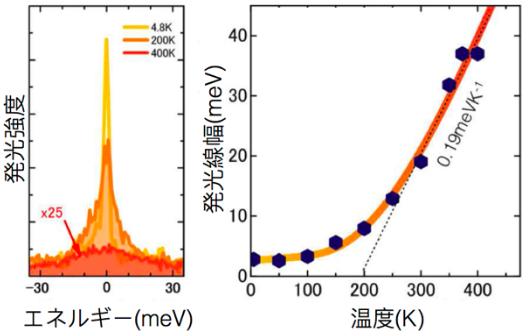

Fig 3. (Left) Emission spectra of the quantum dot at several representative temperatures. (right) The emission linewidth (FWHM) of the emission over two orders of magnitude of temperature.

gradient of ~0.19meVK^{-1}. This value is indeed much larger than the values reported for other material systems (InAs/GaAs QDs: 0.02meVK^{-1}, CdTe/ZnTe QDs: 0.05meVK^{-1}) and is most likely a direct consequence of the piezoelectric nature of the nitride material system: phonons can couple to the electronic states via both a deformation potential mediation (present in all materials) and also a piezoelectric mechanism. This helps us to understand the linewidth of these quantum dots at high temperature (up to approximately 40meV at high temperature).

The temperature dependence of the integrated emission intensity was also investigated over the same temperature range. The data is shown here in figure 4, along with previous data from the literature on GaN quantum dots and quantum wells. It is apparent that QDs maintain a higher relative emission intensity than quantum wells at higher temperatures (as expected due to the 3D confinement provided by the dots), and this has now been clarified to higher temperature ranges. We measured a sharp decrease in the emission intensity for temperatures above 300K, noting that no emission was observable at temperatures higher than 400K. 400K is the highest temperature to date at which photon emission has been observed from a single quantum dot. The exact mechanism of the sharp decrease in emission intensity at high temperature is not understood at present, and it is still unclear if this is a property of this particular dot, or if it is something more general, that may act as a limit to the temperature range over which GaN QDs can operate. Nevertheless, the work performed as a part of this project was able to extend the knowledge in the literature on the temperature dependence of GaN QD photoluminescence.

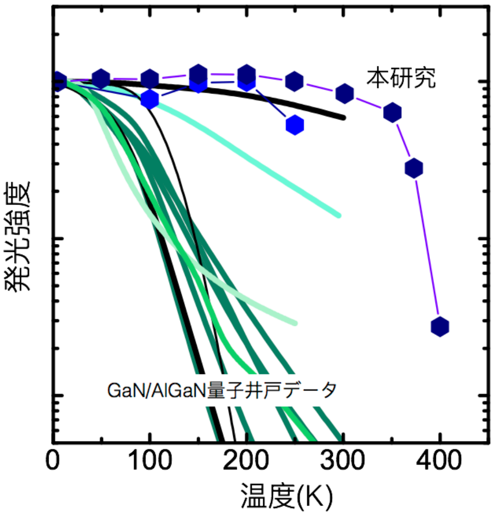

Fig 4. Temperature dependence of the emission intensity of an isolated GaN nanowire quantum dot (hexagons with purple line).

It is important to make special acknowledgement of the work that the crystal growers have made towards the results presented in this work. It is through their effort that material of a suitable quality could be fabricated for the measurements described here.

In addition to the work outlined above, results were obtained in the observation of linearly polarized single photon emission from InGaN/GaN quantum dots formed in non-polar and semi-polar regions of nanowires. Single photon emission was observed from the localization centers in all three material regions of the same type of structure.

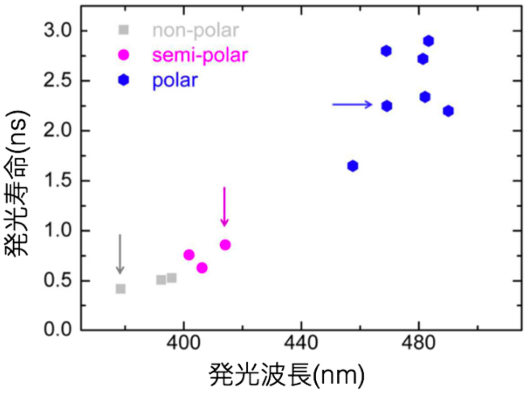

Fig 5. Emission lifetimes measured from various InGaN/GaN nanowire quantum dots in regions of different crystal polarity. The lower internal electric field in the semi-polar / non-polar quantum dots leads to faster emission lifetimes than those measured from polar-material quantum dots.

Due to the lower value of electric field across the semi-polar and non-polar quantum dots, and hence a less intense quantum confined Stark effect (QCSE) the emission lifetimes were measured to be much quicker for these structures, when compared to the polar quantum dots from the same structures. Results are shown in figure 5. This work was performed in collaboration with research groups in Spain and Germany, and was also published in ACS Photonics.

５．主な発表論文等

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６．研究組織

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