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Top-down and Bottom-up Processes  
(トップダウン及びボトムアッププロセスを用いて作製した化合物半導体ナノ構造)  
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## 論文内容要約

Recently, the high information processing is required because the amount of information becomes larger and larger. To achieve the high speed information processing, the electronic devices become smaller and smaller. For such devices, optoelectronics devices which are combined electronics and optics devices are developed. However, the scale of semiconductor elements has reached the order of nanoscale, the problems appear due to a wave nature and a particulate property of electrons. Recently, the new concept semiconductor devices which positively utilize a wave nature and a particulate property of electrons are investigated, such as quantum dots (QDs) light emitting diodes (LEDs), laser diodes (LDs), and solar cells. Currently, top-down process and bottom-up process are used for the fabrication of semiconductor devices. However, the fabrication of high quality nanostructure is difficult by using conventional top-down and bottom-up processes. In case of top-down process, it is difficult to fabricate the photomask with the high accuracy in lithography process and to etch with high accuracy and without inducing the crystal defects in plasma etching. On the other hand, in case of bottom-up process, there are the limitation of growth materials and the internal stress of nanostructure.

In this work, the nanostructures of compound semiconductor were fabricated by using both top-down and bottom-up processes. In case of top-down process, GaAs QDs array was fabricated by combining a neutral beam (NB) and a bio-template processes. On the other hand, in case of bottom-up process, III-Nitride nanowall structures were fabricated by using molecular beam epitaxy (MBE) system and its characteristics were investigated.

At first, GaAs NDs array was fabricated by combining NB and bio-template processes. By using bio-template process, the uniform etching mask array was formed on GaAs surface. By using NB process, the sample was etched with defect-less. The fabricated nanostructure is called “nanodisks (NDs)” because of disk shape QDs. For the fabrication of the high in-plane density, uniformity, and separated NDs array, PEG ferritin molecules which are chemically modified with methoxy-polyethylene glycol succinimidyl esters were

used. The density of PEG ferritin and distance between PEG ferritins can be controlled by changing the ionic strength of an ammonium acetate buffer solution. As increasing the ionic strength of buffer solution, the density of PEG ferritin was increased and the distance between PEG ferritins was decreased. By using PEG ferritin and NB technology, the fabrication of distributed and sub-20-nm GaAs/AlGaAs nano-pillar which includes GaAs ND was achieved. The ND has two geometric parameters, thickness and diameter, and they are controlled independently. The thickness of ND can be controlled during the deposition of stacked layers. The diameter control of ND was investigated by tuning the etching processes. As increasing the hydrogen radical treatment time from 10 to 30 min, the diameter was decreased from 18 to 12 nm. During NB etching, not only iron cores but also GaAs-oxide worked as the etching masks due to the high etching selectivity of GaAs/GaAs-oxide. The photoluminescence (PL) emissions originated from GaAs NDs were observed after regrowth and these peak wavelengths of emission spectra were shifted corresponding to the diameter of GaAs ND.

Since QD LEDs and LDs require a thick active layer to obtain large optical gain, the fabrication process of three-dimensional (3D) GaAs NDs array was investigated. The surface oxidation state has an effect on the fabrication of 3D GaAs NDs array. By using the low temperature oxygen annealing in vacuum, hydrogen passivation, and native oxide, the 3D GaAs NDs array was fabricated with the high density ( $1 \times 10^{11} \text{ cm}^{-2}$ ) and uniformity. By utilizing 3D GaAs NDs array for LEDs, the LED which was operated at room temperature was fabricated.

Secondly, the GaN and AlN nanowall structures were grown on Si (111) substrate by using MBE system. The AlN nanowall structures were grown in the N/Al flux ratio from 200 to 550. Increasing the N/Al flux ratio, the width of AlN nanowall structures was increased from 60 to 120 nm, while the length of AlN nanowall structures was decreased from 470 to 190 nm. From XRD measurement, it was found that the AlN nanowall structures consist of the hexagonal AlN crystals grown along c-plane and the AlN nanowall structures were grown in the nitrogen-rich growth condition at the expense of the crystal quality. The GaN crystals in the various N/Ga flux ratios were also grown on Si (111) substrate. In the high N/Ga flux ratio of 350, the continuous and dense GaN nanowall structure was grown. As decreasing N/Ga flux ratio, the continuity of GaN nanowall was decreased. Finally, at the low N/Ga flux ratio of 50, GaN nanopillars were grown. The PL spectra of GaN nanowalls and InGaN multiple quantum wells (MQWs) on GaN nanowalls were measured. In case of GaN nanowall, decreasing N/Ga flux ratio, the emission originated from GaN near band edge became dominant. Moreover, InGaN MQWs were grown on GaN nanowall in the various N/Ga flux ratios. The difference of PL intensities at a low temperature and room temperature was decreased

while the surface morphology becomes close to the film structure as decreasing N/Ga flux ratios.

The GaN nanowall based LED was fabricated because the GaN based LEDs is widely investigated. Although the emission spectra could not be detected by current injection, the PL emission was observed. It is estimated that the injection carriers were lost by the stacking fault in the GaN nanowall layer and the Mg-doped GaN layer did not work as the p-type GaN such as too high Mg concentration.

In summary, the nanostructures of compound semiconductors were fabricated, GaAs NDs array by top-down process and III-Nitride nanowall structures by bottom-up process. The diameter of GaAs NDs was controlled, and the PL emission spectra corresponding to the size of GaAs NDs was observed. In addition, the highly dense and uniform 3D GaAs NDs array was fabricated and it was utilized for LEDs. The LED operated at room temperature. The GaN and AlN nanowall structures were grown on Si substrate and its crystal quality was investigated. Decreasing Nitrogen/III-element flux ratio, the qualities of nanowall structures were improved. Moreover, InGaN MQWs were grown on GaN nanowalls. As increasing N/Ga flux ratio during GaN nanowall growth, the difference of PL intensities at low temperature and room temperature became smaller. Furthermore, the fabrication of GaN nanowall based LEDs was achieved although the emission by current injection was not observed. The results indicated that the possibility of the fabrication GaN nanowall based LEDs.