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論 文 内 容 要 旨

InGaN alloy system has been widely used over a decade in visible optoelectronic devices because the band-gap of InGaN can cover the whole visible wavelength region. By using InGaN as an active layer, blue light-emitting-diodes (LEDs) with high output power have been commercially available. However, the light output power of InGaN-based green LEDs is still much lower than that of blue LEDs. In the case of a green laser diode (LD), its threshold current is much higher in comparison with a blue LD. These poor characteristics of devices are due to the poor crystalline quality of indium (In)-rich InGaN. The growth of In-rich InGaN with high crystalline quality has been still a challengeable issue. For growing In-rich InGaN with high crystalline quality, N-polar growth is expected because N-polarity has a benefit for capturing In-atoms, *i.e.*, one nitrogen atom is captured with three atoms of group-III while in group-III polar growth, one nitrogen atom is captured with only one atom of group-III [1]. Also, N-polar nitride-based devices have attracted attention because of their benefits of the reverse direction of the internal electric field compared with conventional Ga-polar GaN-based devices such as improvement in carrier confinement at high current density for N-polar LDs and LEDs [2].

Usually, the growth of N-polar GaN on sapphire substrates by metalorganic vapor phase epitaxy (MOVPE), which is widely used in the mass production of III-V semiconductors, is said to be difficult because its surface becomes rough with hexagonal hillocks due to the generation of inversion domain and the poor surface-migration of Ga adatoms on an N-face surface. Also, a high density of residual impurities generates residual electrons and it impedes the realization of p-type GaN [3]. In our previous study, it has been already succeeded in the growth of N-polar GaN of comparable crystalline quality with Ga-polar GaN by optimizing the initial nitridation of a sapphire surface and the other growth conditions to promote the lateral growth. However, the growth of N-polar InGaN with high crystalline quality has not been realized yet. The thick InGaN film is a highly attractive material for the applications of photovoltaic devices such as multi-junction solar cells. Even though the use of N-polar-InGaN-based devices has attract attention, there are few reports on optoelectronic devices fabricated on N-polar crystal plane and the device performances are still poor [4]. Moreover, the effect of growth conditions on the crystalline qualities of N-polar thick InGaN growth using MOVPE has not been researched yet. In addition, the previous research on a N-polar LED structure grown by MOVPE has still suffered from the low quality of crystalline and inhomogeneous emission characteristics [5]. Comprehensive researches on improving an N-polar InGaN/GaN quantum well (QW) are needed for application N-polar devices.

In this thesis, three objectives are mainly addressed in this thesis as follows.

The first objective is the realization of N-polar GaN films with high crystalline quality grown by MOVPE for device applications. The polarity control is treated by nitridation of a sapphire surface. Its polarity is confirmed by KOH wet etching. For optimizing the N-polar GaN growth so as to improve the surface morphology, crystalline quality, and residual donor concentration, the growth was performed under the various conditions such as growth temperature, growth rate, H_2/N_2 ratio and V/III ratio. At high growth-temperature, the sample had a rough surface due to step bunching. The crystalline quality evaluated with XRC measurements was improved by increasing growth temperature. Room-temperature photoluminescence showed strong yellow luminescence around 550 nm, which might be corresponding to the carbon impunity. The surface morphology was drastically changed with the growth rate. For the growth rate higher than 5 μm/h, many pits and cracks appeared on the surface. In the growth with large amount of H_2 flow rate, the rough surface also appeared because of the formation of step bunching. This rough surface resulted in a high concentration and low mobility of residual carriers in N-polar GaN films. High V/III ratio resulted in low residual carrier concentration and smooth surface morphology.

Si and Mg dopants were introduced for n-type and p-type conduction, respectively. In both cases, the surface morphology was roughened by increasing the dopant flow rate. From Raman spectroscopy of n-type N-polar GaN, LO phonon-plasmon coupled mode peaks shifted toward a lower wavenumber at high carrier concentration. N-type and p-type conduction were successfully demonstrated. The carrier concentration was from 2.75×10^{18} /cm^3 to 1.6×10^{18} /cm^3 for n-type conduction and 4×10^{17} /cm^3 for p-type conduction, respectively.

The second objective is the optimization of growth parameters for thick N-polar InGaN films grown by MOVPE. Effects of growth conditions such as the growth temperature, the group-III (In/Ga) source ratio, and the growth rate were experimentally investigated. The growth conditions strongly affected the surface morphology, the indium incorporation, and the crystalline quality of N-polar InGaN. The improper growth conditions resulted in the formation of irregularities such as large hillocks with hexagonal and triangular shapes. These hillocks could be successfully suppressed by adopting more thermal equilibrium conditions such as high temperature, slow growth rate, and low V/III ratio, because these conditions usually promote the surface migration of group-III adatoms during the growth. As a result, 200-nm-thick InGaN films with the low dislocation density of 4.5×10^9 cm^{-2} and the relatively high indium composition up to 15% could be obtained maintaining the smooth surface.

The third objective is the demonstration of N-polar LEDs. Both a single quantum well (SQW) and a multiple quantum well (MQW) on a N-polar GaN template are grown by MOVPE. The inclusion of zincblende phase and the appearance of hexagonal hillocks in a N-polar InGaN/GaN MQW are comprehensively investigated with respect to the growth temperature, partial pressure of H_2 in ambient, and V/III ratio. Based on the above results, N-polar InGaN-based blue and green LEDs were fabricated. By increasing the V/III ratio and reducing the QW growth period, the emission wavelength of MQW LEDs was changed from blue to green. The relatively clear interface in MQWs of a green LED showed as small ideality factor as 8.4 compared to 10.5 of blue LEDs. The electrically-injected emission with a wavelength of 517.4 nm at an injection current of 20 mA was obtained. The emission peak width was 59 nm at 143 A/cm^2, which was narrower than the reported value 63 nm of an N-polar green LED grown on a free standing GaN substrate. This lowering FWHM of EL spectrum compared to the previously reported green LED is originated from the optimization of the QW growth condition. The output power is 2.9 μW at 100 mA (145 A/cm^2).

In summary, the experimental results in this thesis show the high potential of the realization of In-rich N-polar InGaN and high efficiency N-polar GaN-based optoelectronic devices.

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論文審査結果の要旨

本論文は、青色発光ダイオード（LED）などで知られている窒化物半導体において、窒化物半導体の応用範囲の拡大を鑑み、発光層に用いられているInGaNについて、発光波長域の拡大に必要な混晶組成領域の拡張と結晶品質の改善に関する研究結果をまとめており、全7章からなる。

第1章は、序論であり、窒化物半導体InGaAlNの基本物性とその発光素子への応用を概観している。発光素子の発光波長域の拡大と、高効率化のために重要な薄膜エピタキシャル成長に関して、ウルツ鉱型結晶構造に起因する極性について述べている。すなわち、窒化物半導体における気相・固相間の窒素平衡蒸気圧は、従来からある化合物半導体の平衡蒸気圧より数桁高い。このため、混晶組成域が狭く、かつ、結晶の高品質化に必要な高温成長も実現できていなかった。この問題点を解決する方法として、通常用いられてきているIII族極性とは逆の窒素（N）極性成長を提案している。N極性では、結晶成長最表面での結合ボンド数の関係から窒素原子の捕獲が有利となるためである。

第2章は、窒化物半導体の代表であるGaNについて、サファイア基板上への平坦な表面を有する高品質結晶のエピタキシャル成長の最適化について述べている。成長手法は、化合物半導体のエピタキシャル成長において広く用いられている有機金属気相成長である。特に、キーテクノロジーであるサファイア基板表面の窒化による極性制御、成長温度やガス流量などの最適化について述べている。その結果、鏡面の高品質GaN薄膜の成長に成功している。さらに、素子応用において必須の伝導型制御についても、伝導型制御とキャリア濃度の制御を実現している。

第3章は、N極性InGaNの成長について述べている。成長温度と、N原料としてのアンモニアガスと金属原料との供給比（V/III比）の最適化によって、高品質InGaNを成長できることを示している。また、InGaNの組成制御できることも示している。

第4章は、III族極性とN極性のInGaNについて、それぞれの最適条件下で成長したInGaNの組成と結晶品質について、比較している。N極性では、III族極性に較べて、高温成長が可能で結晶品質を高められることと、In組成を高められ発光波長範囲の長波長化を実現している。

第5章は、発光素子の発光層として用いられているInGaN/GaN量子井戸構造のN極性成長を試みている。低温成長において発生しやすい立方晶の混入を高温成長で抑制できること、GaNバリア層成長時にInGaN井戸層成長時の窒素ガスに加えて水素ガスの導入と低V/III比すによって平坦な表面が得られることを示している。

第6章は、N極性窒化物LEDについて述べている。開発したN極性成長技術を用いて、実際に素子を作製し、電流注入による発光を実現している。

第7章は、本学位論文の総括であり、N極性におけるGaN成長、InGaN成長とInGaN/GaN量子井戸構造成長、および、N極性窒化物半導体発光ダイオードにわたっての結論を述べ、N極性成長による窒化物半導体の発展の可能性を示している。

本論文は、窒化物半導体において、六方晶系の結晶特有の極性に注目した研究である。従来からあるIII族極性成長は、成長しやすい性質を有するが、材料における組成や高品質化、および、素子設計におけるバンド構造と分極電界による制約があった。本研究は、N極性成長条件の最適化によって混晶組成域の拡大と高品質化を図り、LEDを作製してその有効性を示している。得られた結果は、窒化物半導体全般にわたるエピタキシャル成長技術として、高出力・高周波インバータなどへの応用も期待でき、応用物理学の発展に大きく貢献すると考えられる。

よって、本論文は博士（工学）の学位論文として合格と認める。