論文の内容の要旨

論文題目　MOCVD Growth and Optical Characterization of Site-Controlled III-Nitride Semiconductor Quantum Dots in Nanowires
（位置制御したIII族窒化物半導体ナノワイヤ中量子ドットのMOCVD選択成長とその光学評価）

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III-nitride semiconductor quantum dots (QDs) are attractive building blocks for efficient light emitters in blue/ultraviolet wavelength regions. They have the potential to realize single photon emitters (SPEs) operating at room-temperature, due to the strong electron confinement and large binding energy of excitons. Generally, semiconductor QDs including III-nitride semiconductor have been epitaxially grown via the so called Stranski-Krastanov (SK) growth process in which the strain is the driving force for the QD formation. Such technical development of the crystal growth has prompted the extensive development of QD-based devices.

However, the spontaneous formation nature of SK growth mode causes random distribution both in position and size, and seriously inhibits full exploration of their potential applications. In order to overcome such difficulties, various methods for control of QD positions have been extensively pursued particularly in III-V QDs: for instance embedding QDs in three dimensional nanostructures, growth of hetero structures in patterned openings, and top-down etching from two dimensional single quantum wells have been demonstrated. These are obviously advantageous for device integration into optical resonators, and fabrication of single QD-based photon emitters, photovoltaic devices.

Semiconductor nanowires have emerged as a promising platform to embed single QD. These are highly beneficial for the following reasons. First, the location of QD as well as the size can be predetermined by that of the nanowire. Second, the three dimensional geometry offers efficient light extraction. Moreover, small-diameter nanowires offer efficient strain relaxation in axial heterostructures and epitaxial structures grown on non-lattice-matched substrates. Also, the growth of a disk-like quantum dot in a nanowire allows the QD height as small as a few nanometers or less, which drastically reduces the influence of quantum confined stark effect (QCSE) in III-nitride heterostructures.

This thesis presents original research works on the crystal growth and optical characterization of site-controlled single GaN QDs in GaN/Al(Ga)N nanowires. Chapter 1 is the introduction of this thesis, in which an overview of research field on III-nitride QD and QD in nanowire structures is introduced.

In chapter 2, the general properties of III-nitride compound semiconductor are presented. After the introduction of structural properties, the electronic properties and the consequence optical properties will be given. These are extended to GaN/Al(Ga)N heterostructures in terms of the effect of built-in polarization.

In chapter 3, the experimental details on the crystal growth and optical characterization are described. In this work, the metalorganic chemical vapor deposition (MOCVD) system was used for the crystal growth. After the general description, the details of the MOCVD which the author has operated will be introduced. Next, the detailed information of the micro-photoluminescence (PL) measurement system for the single QD spectroscopy is given.

Chapter 4 describes the selective area growth of thin GaN nanowires by MOCVD. The growth of GaN nanowire having small diameter less than 50 nm is crucial for embedding GaN QDs. By exploring the growth conditions including material flow rates and pattern fill-factor in detail, the selective are growth of Ga-polar GaN nanowires with a diameter of 50 nm has been demonstrated. The diameter of GaN nanowire is thinnest ever reported in selective area growth to the best of our knowledge. It has been shown that an appropriate shell layer is effective to considerably improve the optical properties, suggesting the importance of controlling surface states.

Chapter 5 is devoted to the growth of site-controlled single GaN QDs in GaN/Al(Ga)N nanowires. The design of the sample structure such as substrates, crystal polarity of nanowires, surface morphology of the barriers was optimized aiming at maximizing the luminescence intensities of GaN QDs. More importantly, in order to nucleate a single GaN QD in a nanowire precise control of the GaN deposition and growth interruption which induces re-evaporation of GaN has been found to be crucial parameter. The optimized structures were analyzed by high-resolution transmission electron microscope (HRTEM), and the formation of a very tiny GaN QD with the thickness of around 1 nm in nanowire tip has been identified.

In chapter 6, the optical properties of the single GaN QDs in nanowires have been studied. The narrow PL peaks corresponding discrete density of states of QD have been observed. The measured values of linewidth are almost comparable or much less compared to those of conventional SK GaN QDs. From the excitation power dependent PL measurements, the excitonic and biexcitonic emissions have been observed for the first time in the selectively grown GaN QDs. The biexciton binding energy reaches up to +52 meV, which is believed to be the largest value ever reported in the III-V semiconductor QD system. The origin of the large binding energy might be originated from the large overlap between electron and hole wavefunctions in the small QDs. The temperature dependent PL measurements have been performed to verify the carrier confinement in the QDs. Surprisingly, the emissions from the single QD is observed even at room-temperature, revealing strong thermal stability. The activation energies and the exciton-phonon coupling coefficients are discussed. Further collaborative works on optical characterizations including photoluminescence excitation (PLE) measurements, photon correlation measurements are presented. The results in this chapter evidence that the GaN QDs exhibit intrinsic properties of QDs. The excellent optical properties suggest that the site-controlled GaN QDs in nanowires constitute a promising platform for not only the investigation of fundamental physics, but also the development of quantum optical devices based on nitride based nanostructures.

Finally, in chapter 7, conclusions of this thesis are presented. Implications of the results presented in this thesis are discussed. An outlook for the future research and development is also given.