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主 論 文 の 要 旨

論文題目　Study on Nitride-based Light Emitting Diodes Grown on Nano-structured Templates （ナノ構造テンプレートを用いる窒化物半導体LEDに関する研究）

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論 文 内 容 の 要 旨

III-nitride based light emitting diode is a very promising light source owing to its high energy conversion efficiency. However, there are still some fundamental issues that limit the LED efficiency. Due to the large lattice and thermal mismatch between nitrides and their foreign substrates such as sapphire or silicon, there is usually a high density dislocation density inside the epi-layers. The large refractive index of GaN leads to a very small total reflection angle. A large portion of emitted light is confined inside the chips. The spontaneous polarization and piezoelectric polarization induced quantum-confined Stark effect (QCSE), which separates the electron-hole wave function in the quantum wells, results in a degraded radiative recombination rate. Nanostructures offer a possibility to solve these issues. In this thesis, we designed and investigated several novel nanostructures of great potential to improve the LED performance.

The regular nanohole-patterned SiO_2 mask as well as the nanohole-patterned GaN template used in this research were processed by a thermal nanoimprint process followed by reactive ion etching and inductively coupled plasma etching. The nanoholes are regularly distributed in a hexagonal geometry. The hole diameter and pitch was 230 and 460 nm, respectively. The thickness of the SiO_2 layer was about 45 nm, and the depth of the GaN nanohole template was about 500 nm.

To fabricate large-scale nanoporous GaN templates, ICP dry etching and electrochemical etching have been reported previously. In this research, a novel thermal etching method in a metalorganic vapour phase epitaxy (MOVPE) reactor for the fabrication of nanoporous templates was studied. The nanoporous template etched with a pulsed NH_3 flow showed dense voids with a density of approximately 1×10^9 cm^{-2}. The thermal etching process was quite anisotropic and very likely to be dislocation sensitive. The etching rate of the c-plane could be more than 10 times higher than that of the side wall. The nanohole SiO_2 mask was found to be essential in this process. If there was no SiO_2 mask on the GaN template or if there was no nanohole windows on the SiO_2 mask, the nanoporous structure could not be formed. Compared to the pulsed etching process, the nanoholes fabricated with continuous NH_3 flow showed much smaller lateral diameter and the side wall of the voids became very smooth, implying that NH_3 could effectively protect the side wall while the hydrogen carrier gas promoted the isotropic etching. When continuous NH_3 flow was introduced, nanopyramids were spontaneously formed in every mask window even without introducing a TMG flow. The formation of nanopyramids was explained by the mass transport process. Under the SiO_2 mask, only spots where dislocations were located tended to decompose. When there was a nanohole-patterned mask, the products of decomposition could effectively escape through the short path at the interface of the GaN template and the SiO_2 mask to the nanohole window, then the decomposition process could continue. When the NH_3 flow was continuously introduced, the etching effect was reduced and GaN redeposition at the window region formed the nanopyramids. The fabricated nanoporous template showed almost full strain relaxation and a five-fold increase in photoluminescence (PL) intensity compared with a planar GaN template. By adjusting the regrowth parameters, the decomposition and deformation of the nanoporous templates during GaN regrowth was effectively suppressed. GaN films overgrown on the nanoporous templates fabricated from GaN templates with high dislocation density showed a five-fold increase in PL intensity compared with a planar sample. The dislocation density was reduced from 3×10^9 cm^{-2} to 4×10^8 cm^{-2} as measured by panchromatic cathodoluminescence (CL) measurement. The full widths at half maximum (FWHM) of the x-ray rocking curve data for the (002) and (102) planes were reduced from 326 and 882 arcsec to 203 and 464 arcsec, respectively. The surface flatness were also obviously improved. The surface morphology measured by AFM in 2×2 μm^2 area showed a lower root mean square (RMS) value of 0.16 nm compared with 0.23 nm of the planar sample. For nanoporous templates fabricated from high quality GaN template, however, the crystal quality was not obviously improved. The dislocation densities before and after overgrowth were both approximately 4×10^8 cm^{-2}. The twofold increase in PL intensity compared with the planar template was mainly attributed to the improved LEE. The dislocation reduction and embedded void formation were also confirmed by using a showerhead configuration. Three period $In_{0.1}Ga_{0.9}N$/GaN multiple quantum wells (MQWs) grown on the nanoporous templates that fabricated from high quality GaN templates showed a twofold increase in PL intensity, which was also mainly attributed to the improved LEE.

LEDs grown on the nanopyramids take the advantage of the semipolar (1-101) planes which could greatly reduce the QCSE in the quantum wells. In addition, nanopyramid based LEDs also have larger light emitting area. However, for GaN nanopyramid based LED, a large series resistance was usually reported due to a thick p-GaN layer required for surface planarization. By adjusting the growth condition, we successfully overcome the "self-limiting" phenomenon. Unlike previous reported nanopyramids which had sizes restricted by the mask window, the size of the GaN nanopyramids could be controlled by using lower growth temperatures and nitrogen as the carrier gas instead of hydrogen. The pyramid size was highly uniform and the abnormal growth of large islands was also successfully suppressed. It was found that the nanopyramids exhibited two types of shapes, one had a six-fold symmetry and the other showed a threefold symmetry. There were two PL emission peaks from MQWs grown on the nanopyramids. One peak with higher intensity centered at 2.76 eV has a FWHM of 294 meV, and another peak at 2.41 eV with lower intensity has a FWHM of 268 meV. By studying the monochromatic CL images, it was found that the longer-wavelength peak originated only from the apexes of pyramids with threefold symmetry. From the transmission electron microscopy (TEM) images, high-density stacking faults were observed at the apexes of the pyramids with threefold symmetry. The high-density stacking faults might be responsible for the higher indium incorporation at the triangular apexes of the pyramids by providing more atomic steps on the surface. In every nanopyramid an embedded void was observed. The formation mechanism was explained by a higher growth rate at the edge than at the center of the window region caused by selective area growth (SAG) and the small diffusion length. After 300 nm p-GaN growth following the growth of MQWs, fully coalesced flat p-GaN surface was achieved, confirming that the fast coalescence could be realized by this method. Besides a reduction of resistance, crystal quality improvement by lateral overgrowth and an even increased light emitting area were also expected. This method could be beneficial for realizing high efficiency nanopyramid based LEDs.

The nanopyramid based LED could effectively reduce the QCSE. However, the light extraction efficiency (LEE) was limited due to the formation of a flat p-GaN surface. The indium composition ratio distribution on the pyramids was also not uniform on the nanopyramids. A nanohole-patterned template was designed and fabricated for better LEE and indium composition uniformity. The nanohole template was measured to be almost completely strain relaxed which may reduce the QCSE in the quantum wells. It was found that compared with directly growing the InGaN/GaN MQWs on the nanohole template, initiating the regrowth process by growing high temperature GaN buffer layer of a certain thickness could improve the crystal quality of the MQWs. The integrated PL intensity of MQWs grown on a nanohole sample showed a four-fold increase compared with a planar sample. By measuring x-ray diffraction reciprocal space map of the asymmetric (105) reflection, it was conformed that the MQWs were coherently growth on the nanohole template. The slight red shift and the broader peak observed in the PL spectrum were attributed to a thicker InGaN well width and a larger indium inhomogeneity of the MQWs grown on the nanohole template. By measuring the monochromatic CL images of the MQWs on the nanohole template, it was found that the light emission mainly came from the MQWs grown on c-plane surface, and the spatial distribution of the emission intensity and wavelength were quite uniform. Two sets of regularly distributed nanostructures including inverted nanopyramid structure on the p-GaN surface and embedded void arrays were formed at the same time after growing the LED structure on a nanohole template. As high as 3.5 times increase in integrated PL intensity was observed for LED structure grown on a nanohole template compared with a planar sample owing to a much improved LEE. Although QCSE might be reduced by the strain relaxed nanohole template, the IQE of the nanohole based LED was measured to be lower than that of a planar LED, which probably resulted from a lower crystal quality of the MQWs grown on the nanohole template. This confirmed that the great increase of the PL intensity for the nanohole samples was owing to the great improvement of the LEE. By simulation of the LEE of the nanostructures with the finite difference time domain method, it was confirmed that the distinct nanostructures formed in the research consisting of both inverted pyramid structure on the surface and embedded voids exhibited the highest LEE compared with the conventional nanostructures with only embedded voids or only inverted nanopyramids on the surface.