Optical properties of GaN pyramids

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Picosecond time-resolved photoluminescence (PL) spectroscopy has been used to investigate the optical properties of GaN pyramids overgrown on hexagonal-patterned GaN(0001) epilayers on sapphire and silicon substrates with AlN buffer layers. We found that: (i) the release of the biaxial compressive strain in GaN pyramids on GaN/AlN/sapphire substrate led to a 7 meV redshift of the spectral peak position with respect to the strained GaN epilayer grown under identical conditions; (ii) in the GaN pyramids on GaN/AlN/sapphire substrate, strong band edge transitions with much narrower linewidths than those in the GaN epilayer have been observed, indicating the improved crystalline quality of the overgrown pyramids; (iii) PL spectra taken from different parts of the pyramids revealed that the top of the pyramid had the highest crystalline quality; and (iv) the presence of strong band-to-impurity transitions in the pyramids were primarily due to the incorporation of the oxygen and silicon impurities from the SiO_2 mask. © *1999 American Institute of Physics.* [S0003-6951(99)02609-1]

Wide band gap III–nitrides have recently attracted considerable interest due to their applications for optical devices which are active in the blue and ultraviolet (UV) wavelength regions and electronic devices capable of operation at high temperature/power and in harsh environments.[1] The recent success of the edge-emission lasers[2] based on III–nitrides is encouraging for the study of other laser geometries such as vertical cavity and microdisk cavity lasers. These alternative laser geometries offer several benefits resulting from confinement of the optical mode to a microcavity, including enhanced quantum efficiency and a greatly reduced lasing threshold. Additionally, the compatibility to two-dimensional array fabrication is an inherent attribute of these lasers, which are of much interest for optical display, imaging, scanning, optical parallel interconnects, and ultraparallel optoelectronics applications. A dry etching technique has been applied previously to fabricate GaN microdisk cavities.[3,4] A large enhancement of the intrinsic transition quantum efficiency has been observed in GaN/AlGaN multiple quantum well microdisk cavities.[3] Furthermore, when individual disks were optically pumped, optical modes corresponding to the radial and the Whispering Gallery modes were observed.[3]

In this work, we have studied the optical properties of an array of self-organized GaN hexagonal pyramids fabricated by selective epitaxial metalorganic chemical vapor deposition (MOCVD) growth. It has been shown previously that self-organized GaN microcavities produced by selective epitaxy are of either hexagonal prisms or hexagonal pyramids due to the nature of the crystal structures.[5–7] Furthermore, it has been demonstrated that the threading dislocation density in GaN can be significantly reduced by employing lateral epitaxial overgrowth.[8–11] Thus it is of great importance to study and understand the optical properties of these novel structures produced by selective epitaxy.

GaN pyramids were grown by selective epitaxy on the GaN epilayers on GaN/AlN sapphire or GaN/AlN/silicon substrates as depicted in Fig. 1(a). Before the pyramidal overgrowth, a 1-μm-thick GaN epilayer was grown on a (0001) sapphire or silicon substrate with a thin AlN buffer layer. A 0.2-μm-thick SiO_2 mask was coated on the GaN epilayer. Hexagonal windows with 3.5 μm per side and 20 μm apart were prepared by photolithography together with dry etching, followed by the GaN pyramidal overgrowth. Scanning electronic microscopy (SEM) was employed to

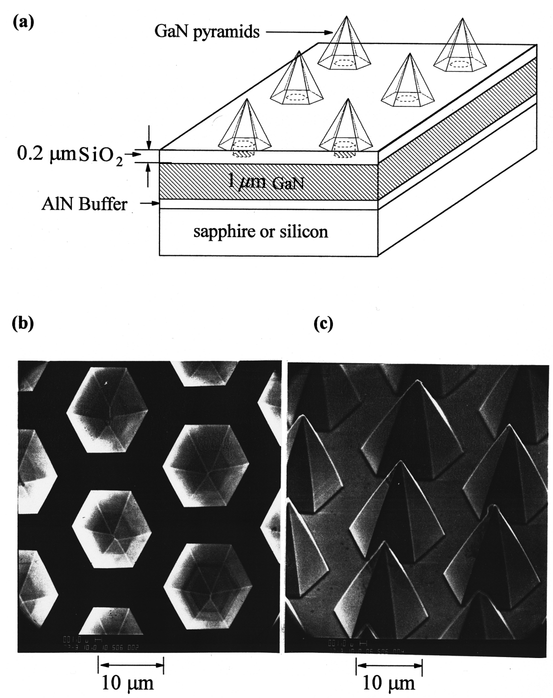

FIG. 1. (a) Schematic diagram showing GaN pyramids fabricated by selective epitaxial overgrowth on the GaN/AlN/silicon or GaN/AlN/sapphire substrates; (b) top view, and (c) side view of SEM images of a pyramid array.

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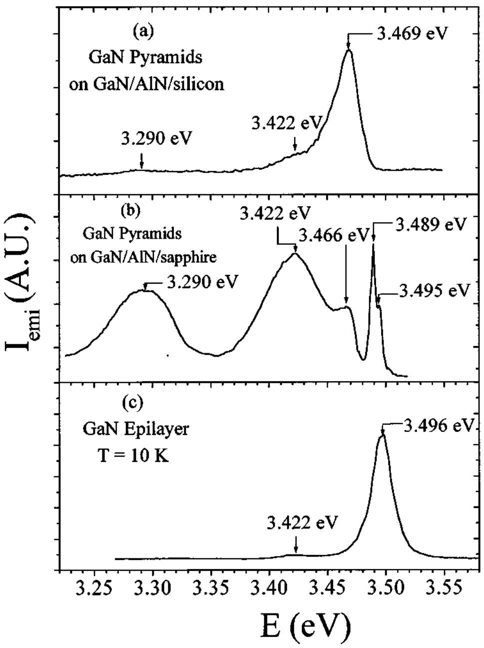

FIG. 2. Low-temperature ($T = 10$ K) PL spectra of the GaN pyramids overgrown on (a) the GaN/AlN/silicon substrate and (b) the GaN/AlN/sapphire substrate. PL spectrum of a GaN epilayer grown under identical conditions is plotted in (c).

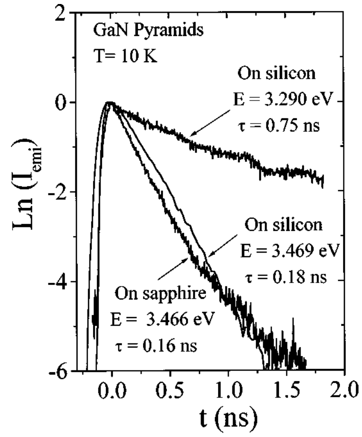

FIG. 3. PL temporal responses measured at 10 K and at emission energies $E = 3.294$ and 3.469 eV for the GaN pyramids overgrown on the GaN/AlN/silicon substrate, and at $E = 3.466$ eV for the GaN pyramids overgrown on the GaN/AlN/sapphire substrate.

study the morphology of the GaN pyramids, which revealed that all six surfaces were extremely smooth with very good morphology as shown in Figs. 1(b) and 1(c). These GaN pyramids formed a two-dimensional (2D) array. The length of each side of the base of the self-organized pyramids was about 7μm and the height of the pyramid was about 14 μm.

Low-temperature PL spectra were measured by a picosecond laser spectroscopy system with an average output power of about 30 mW at $\lambda = 292$ nm and a spectral resolution of about 0.2 meV. The laser beam was focused onto a spot of about 20 μm in diameter. A single photon counting system and a streak camera were used to collect time-resolved PL data. The time resolution of the single photon counting system and the streak camera were 20 and 2 ps, respectively. Detailed information on the time-resolved PL system can be found elsewhere.[12]

The low-temperature PL spectra of the GaN pyramids on the GaN/AlN/silicon and GaN/AlN/sapphire substrates are presented in Figs. 2(a) and 2(b), respectively. For comparison, the PL spectrum of a GaN epilayer on sapphire substrate with AlN buffer layer grown under identical conditions as the GaN pyramids on GaN/AlN/sapphire substrate is also included in Fig. 2(c). For the GaN pyramids on GaN/AlN/silicon, the main emission band at 3.469 eV is attributed to either a neutral donor bound exciton or band-to-impurity transition. This assignment is based on its spectral peak position and its decay lifetime (~0.18 ns) (Fig. 3). Two other emission lines at 3.422 and 3.290 eV are also evident. The emission line at 3.422 eV is very close to the emission line associated with the presence of oxygen impurities in GaN epilayers reported previously.[13,14] We thus assign the emission line at 3.422 eV as the recombination between the electrons bound to substitutional oxygen donor impurities and free holes, or the (D^0,h^+) transition. The emission line at 3.290 eV is not a donor–acceptor pair transition, since its lifetime is only about 0.75 ns as shown in Fig. 3. For the GaN pyramids on the GaN/AlN/sapphire substrate [Fig. 2(b)], besides the transition line at 3.466 eV (with a decay lifetime ~0.16 ns as shown by Fig. 3) and the impurity related transition lines at 3.422 and 3.290 eV, there also exist transition lines at 3.489 and 3.495 eV. The mechanisms of these transition lines will be published in another paper.[15]

For the GaN pyramids grown on GaN/AlN/sapphire substrate, the emission line at 3.489 eV is about 7 meV below the corresponding peak at 3.496 eV for the GaN epilayer. This 7 meV spectral redshift can be explained by the release of the biaxial compressive strain in the overgrown GaN pyramids. A 1.5 meV blueshift of the band edge transitions in the laterally overgrown GaN stripes on GaN/AlN/6H–SiC(0001) substrate with respect to that of the underlying GaN epilayer has been previously reported.[11] This is expected since the GaN epilayer on 6H–SiC(0001) substrates is subject to a biaxial tensile strain.[16] However, in our case with a sapphire substrate, it corresponds to a biaxial compressive strain. Its release in the GaN pyramids leads to a redshift of the spectral peak. The magnitude of the strain in the GaN epilayer can also be calculated. The 7 meV redshift corresponds to a release of the ϵ_{zz} value of about 0.05% (denoting the magnitude of the uniaxial strain along the c axis)[17] in the GaN pyramids with respect to that of the GaN epilayer.

The impurity related transitions at 3.422 and 3.290 eV are found in both pyramid samples grown on GaN/AlN/silicon or GaN/AlN/sapphire substrates as shown in Figs. 2(a) and 2(b). However, in the GaN epilayer, besides the transition line at 3.496 eV only a weak transition at 3.422 eV is observed. It thus suggests that these two emission lines at 3.290 and 3.422 eV are associated with Si and O impurities due to the use of a SiO_2 mask in the overgrowth of pyramids. Since the emission line at 3.422 eV is observable in the two pyramid samples as well as in the GaN epilayer, it further confirms the previous assignment that this transition may be related to an oxygen but not a silicon impurity. The emission line at 3.290 eV, which is about 220 meV below the band gap, is observable in both pyramid samples but not may be the GaN epilayer. It may be related to an acceptor level

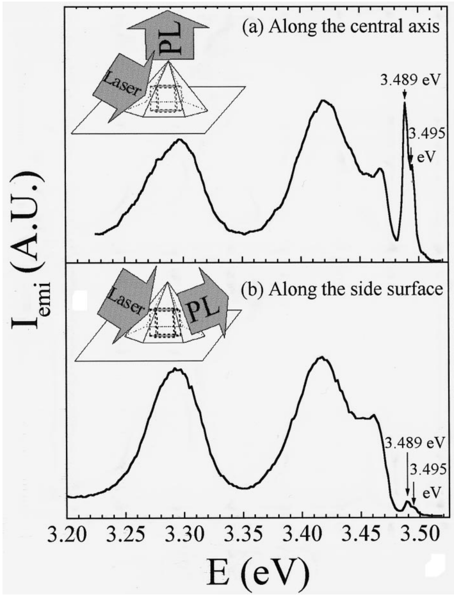

FIG. 4. Low-temperature PL spectra of the GaN pyramid overgrown on the GaN/AlN/sapphire substrate collected along (a) the central axis and (b) one of the six surfaces of the pyramid.

induced by silicon impurity as calculated[18] and observed previously.[19] In the GaN pyramids on GaN/AlN/silicon substrate, the emission lines at 3.489 and 3.495 eV are absent. This is consistent with the fact that it is much harder to grow high quality GaN on silicon than on sapphire substrate partly due to the larger lattice mismatch between GaN and Si than that between GaN and sapphire.

Diffusion of Si and O impurities from the SiO_2 mask during the pyramidal overgrowth should leave an impurity distribution in the pyramids with fewer Si and O impurities close to the top of the pyramids. In order to check the crystalline quality and purity in different parts of the pyramids, we have employed two different configurations to collect PL from the overgrown pyramids on GaN/AlN/sapphire as illustrated in the insets of Fig. 4. In both configurations, the incident laser beam is perpendicular to one of the six surfaces of the pyramid. PL is collected along the central axis (or one of the surfaces) of the pyramid as shown in Fig. 4(a) [Fig. 4(b)] in such a way that the PL from the top (or base) part of the pyramids dominates. Comparing the PL results shown in Fig. 4, the intrinsic transitions lines relative to the band-to-impurity transitions are significantly enhanced in the top of the pyramids [configuration Fig. 4(a)]. The absolute emission intensity of the transition line at 3.489 eV in the top part of the pyramid [Fig. 4(a)] is also much higher than that in the base part. These results imply that the crystalline quality and purity of the top part of the pyramids is higher than that of the base part. Our results are consistent with that reported in Ref. 9. It was shown there that dislocation diminishes above approximately one third of the pyramid height within a pyramidal volume.

In summary, our results show that: (i) the release of the biaxial compressive strain in the GaN pyramids overgrown on sapphire substrate leads to a 7 meV redshift of the spectral peak position with respect to that of the strained GaN epilayer grown under identical conditions; (ii) in the GaN pyramids on GaN/AlN/sapphire substrate, strong band-edge transitions involving both the *A* and *B* valence-edge bands with much narrower linewidths than those in the GaN epilayer are observed, indicating the improved crystalline quality of the overgrown pyramids; (iii) the top portion of the pyramid has a much higher crystalline quality and purity than the base part; and (iv) both oxygen and silicon impurities have been incorporated into the overgrown pyramids due to the use of SiO_2 mask and high growth temperature. Our results suggest that self-organized microcavities formed by selective epitaxy can be further developed for the realization of GaN microcavity lasers with minimum parasitic optical losses as well as a simplified device process that completely eliminates the need for etching the crystal. Indeed, room temperature laser action in GaN pyramids grown on silicon substrate by selective lateral overgrowth has been demonstrated most recently.[20]

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