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Effect of annealing on phase separation in ternary III–N alloys

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Major developments in wide band gap III–N semiconductors have led to commercial production of high brightness light-emitting diodes and to demonstration of room temperature laser light emission under pulsed and continuous-wave operation in violet [1], and, recently, in blue spectral region [2]. However, the internal structure of InGa_N and AlGa_N alloys indispensable for fabrication of high-quality light emitting devices has not been properly investigated. There exist many reports on optical properties of thick ternary alloys and GaN/InGa_N and GaN/AlGa_N heterostructures. Most of papers are deduced to determination of compositional dependence of band gap of "bulk" AlGa_N and InGa_N alloys. Values reported for bowing parameters of AlGa_N range from $b = 0$ [3] to $b = 1.0$ eV and $b = 1.3$ eV [4, 5]. For InGa_N alloys reported range of bowing parameter is much wider: from $b = 1$ eV to $b = 4.8$ eV [6, 7]. Nearly all authors use approximation of strained, but homogenous layer determining In composition from X-ray diffractometry. However, it was shown recently that at least for InGa_N ternary alloys compositional fluctuations (phase separation) can play a very important role [8] and must be taken into account calculating In composition from optical transitions energy.

In this paper we report investigation of phase separation in ternary III–N alloys (AlGa_N and InGa_N) and influence of annealing on optical and structural properties of these samples.

The samples studied in this work were grown by low pressure metalorganic chemical vapor deposition (MOCVD) employing an AlGa_N nucleation layer deposited at 530°C on (0001) sapphire substrates. Ammonia, trimethylindium (TMI), trimethylgallium (TMG) and trimethylaluminum (TMA) were applied as component precursors. Purified hydrogen and/or argon [10] were used as carrier gases. Three types of samples were grown: thick (2–3 μm) AlGa_N layers grown directly on sapphire substrate (sample A); structures with thin (50 nm) InGa_N layer sandwiched between 3 μm Ga_N buffer layer and 100 nm thick Ga_N cap layer (sample B); structures with multiple ultrathin (3–5 nm) InGa_N insertions in Ga_N matrix sandwiched between thick Ga_N buffer layer and 100 nm Ga_N cap layer (sample C). The details of growth were reported elsewhere [9, 10].

The photoluminescence (PL) study was performed in the temperature range 4–300 K using a continuous wave He–Cd laser (excitation density 25 W/cm²) or a pulsed excimer laser for excitation.

Temperature dependencies of photoluminescence peak position for sample A and the same sample after rapid thermal annealing (RTA) are shown in Fig. 1. Temperature-dependent PL study of as-grown sample revealed so-called "S-shaped" temperature-dependent emission shift. This feature indicates presence of some kind of localization centers in the layer. After RTA at 1100°C for 120 sec this feature practically disappears and the near-band-edge PL peak position shifts to the high-energy side more than 20 meV. This high-energy

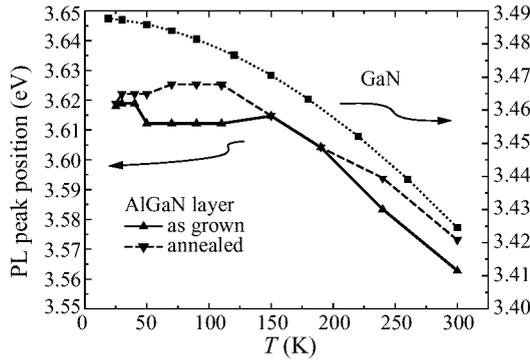


Fig. 1. Photoluminescence peak position vs temperature for as grown and annealed AlGaIn layer. GaN PL peak position is shown for reference.

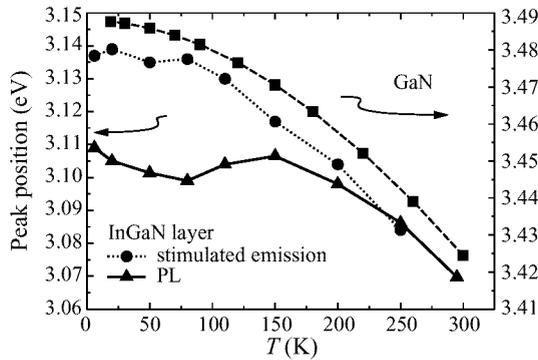


Fig. 2. Photoluminescence and stimulated emission (at threshold) peak position vs temperature for InGaIn layer. GaN PL peak position is shown for reference.

shift of near-band-edge emission after RTA was also observed in PL spectra of thin AlGaIn layers grown on GaN epilayers. RTA also strongly affects electronic properties of the structure. Room temperature Hall mobility after the RTA increased to $200 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ from $30\text{--}40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in the as-grown structure indicating efficient decrease in compositional fluctuations for annealed structure.

Figure 2 shows temperature dependence of PL peak for sample B at low excitation density (25 W/cm^2) (Fig. 2, solid curve) and energies of stimulated emission at edge geometry (Fig. 2, dashed curve). (For stimulated emission energies were taken at threshold excitation density). It is clearly seen that stimulated emission follows the band gap of GaN, while PL at low excitation density reveals “S-shaped” emission shift. This behavior points to low density of localized states formed by phase separation of InGaIn layer.

In our previous work [10] it was shown that growth of multiple ultrathin (3–5 nm) InGaIn insertions in GaN matrix (structure C) leads to formation of dense array of nanoislands with high In composition (quantum dots). The density of these localization centers is so high that at low temperatures these quantum dots (QD) can produce gain necessary even for lasing in vertical direction. Temperature dependencies of photoluminescence peak position for as-grown sample C and the same sample after rapid thermal annealing (RTA) are shown in Fig. 3. After RTA at 1300°C for 30 sec QD-related PL peak shifts to low-energy side

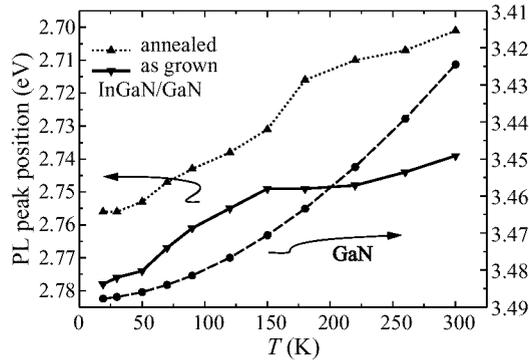


Fig. 3. Photoluminescence peak position vs temperature for as grown and annealed InGaN/GaN multilayer samples. GaN PL peak position is shown for reference.

more than 20 meV. This shift is accompanied by increasing width of PL spectra from 100 to 130 meV indicating increase both in maximum depth of localization potential and in its nonuniformity.

In conclusion, we investigated temperature dependence of photoluminescence for different types of structures with ternary III–N alloys. It is shown that optical properties can be strongly affected by composition fluctuations in AlGaIn or InGaIn alloys. For AlGaIn layers rapid thermal annealing leads to effective decrease in nonuniformity, while for InGaIn multilayer structure with dense arrays of quantum dots RTA leads to increase in phase separation.

Acknowledgements

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