Enhanced luminescence in GaInNAsSb quantum wells through variation of the arsenic and antimony fluxes

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Photoluminescence efficiency was enhanced in molecular-beam-epitaxial-grown 1.55-μm GaInNAsSb single quantum wells through modulation of the arsenic and antimony fluxes. The arsenic-to-antimony flux ratio was found to be a key consideration at reduced group-V fluxes in maintaining the beneficial effects of antimony while reducing the number of point defects, most likely arsenic antisites. Samples were also characterized by high-resolution x-ray diffraction, secondary ion mass spectrometry, and low-temperature photoluminescence. These findings offer a means to substantially reduce dilute-nitride laser threshold current densities. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2213176]

Dilute-nitride materials have received much attention recently for their promise in covering the entire 1.2–1.6-μm communication range with high-performance GaAs-based vertical-cavity surface-emitting lasers, high-power edge-emitting lasers, modulators, and detectors.[1,2] It is well established that low growth temperatures and high arsenic fluxes are required for single phase molecular beam epitaxial (MBE) growth of these materials.[3,4] Despite improvements through the use of antimony as a surfactant[5] and reduction in nitrogen plasma-related damage,[2] these growth conditions result in many defects[6–8] that degrade laser performance. Indeed, even the lowest-threshold 1.5-μm GaInNAsSb lasers[9] still show significant monomolecular recombination (30-50%) of the threshold current) in *Z*-parameter measurements.[10]

It is of great interest to reduce the quantities of arsenic antisites and interstitials and gallium vacancies in dilute-nitride active regions to improve laser performance.[6–8] Increasing growth temperature helps to reduce point defect formation but is limited by phase segregation and/or roughening to $\lesssim$460 °C. This is especially true for 1.55-μm range material that contains ~40% indium and several percent nitrogen (see, for example, Refs. 11 and 12). Reduction of the arsenic flux during growth has been reported by several groups for GaInNAs with mixed results.[11,13,14] Pavelescu *et al.* found a broad range of arsenic fluxes produced virtually identical 1.3-μm range material quality, but substantial degradation occurred at a critical minimum flux.[14] Reduced arsenic flux significantly improved the optical properties of 1.55-μm range GaInNAs, but laser thresholds still remain higher than antimony-containing devices.[11] Studies of dilute-nitride antimonides have not examined the reduced arsenic and antimony flux regimes; the typical approach has been to study the influence of increasing antimony flux under constant growth conditions (see, for example, Refs. 5 and 15–19). We present two studies of 1.55-μm range GaInNAsSb single quantum wells (QWs) that examine the roles of the arsenic and antimony fluxes on the room-temperature optical properties studied via photoluminescence (PL). Structural properties are also quite important and were examined with high-resolution x-ray diffraction (HRXRD) and low-temperature PL. Compositional analysis was performed with secondary ion mass spectrometry (SIMS). While growth under reduced arsenic and antimony fluxes slightly degraded the structural quality, the optical properties were significantly improved. This growth regime will enable significantly reduced laser thresholds and may even improve the stability of device performance with operating temperature.

Samples were grown by solid-source MBE on semi-insulating (100) GaAs. Indium and gallium were supplied with thermal evaporation cells. Dimeric arsenic and monomeric[20] antimony were supplied with cracking cells. Reactive nitrogen was supplied by a SVT Associates rf plasma cell operated with 0.5 SCCM (SCCM denotes cubic centimeter per minute at STP) of ultrapure nitrogen gas and 300 W of input power at 13.56 MHz. Ionized species were removed from the molecular beam by deflection plates at the exit aperture of the plasma cell, one dc biased to −40 V and the other grounded.[2] The active region was nominally a single 75 Å $Ga_{0.59}In_{0.41}N_{0.028}As_{0.942}Sb_{0.03}$ QW surrounded on either side by 220 Å $GaN_{0.03}As_{0.97}$ barriers grown at 410 °C. The active region was grown upon a 3000 Å GaAs buffer and capped with 500 Å of GaAs, grown at 580 °C. Growth temperature was monitored by both band-edge absorption and pyrometry. The measured beam equivalent pressure (BEP) ratios were converted into flux ratios in the usual manner.[21] Only flux ratios are reported here.

In the first experiment, the QW arsenic-to-group-III flux ratio (As/III) was varied with all other growth parameters held constant. The QW antimony BEP was held constant at $\sim 1.2\times 10^{-7}$ torr. Samples were grown out of order to rule out flux drift as a cause for any observed behavior. Figure 1 plots the peak room-temperature PL intensity and emission wavelength, as a function of the As/III flux ratio for as-grown samples. Typical growth conditions[9] of $-10\times$As/III

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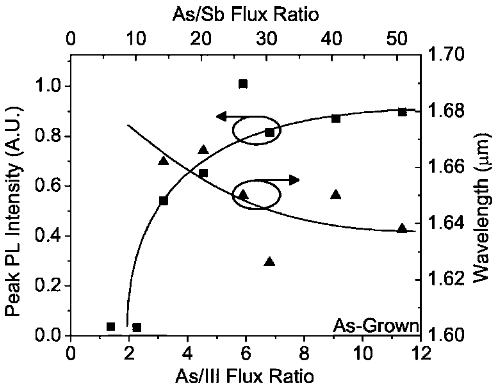

FIG. 1. As-grown peak PL intensity (squares) and wavelength (triangles) for GaInNAsSb QWs grown under various As/III flux ratios, with all other conditions held constant. The arsenic-to-antimony flux ratio (As/Sb) is also shown.

flux ratio produced excellent PL efficiency. Moreover, a wide window of stable emission efficiency and wavelength was observed, similar to observations for 1.3-μm GaInNAs.[14] Upon postgrowth annealing, the PL intensity improved and the emission wavelength blueshifted but remained qualitatively similar to the results of Fig. 1. The optical properties degraded rapidly at reduced As/III, in agreement with theoretical predictions[3] and experimental work on GaInNAs emitting at ~1.3 (Ref. 14) and ~1.55 μm.[11] SIMS measurements showed a substantial increase in antimony content, a moderate increase in nitrogen content, and a moderate decrease in indium content at reduced arsenic fluxes. This is consistent with previous study of the effects of antimony on indium and nitrogen incorporation in (Ga, In)NAs.[15,22]

Figure 2 plots (002) $\omega/2\theta$ HRXRD scans of the samples grown at various values of As/III. Minimal change in strain and structural quality was observed at high As/III flux ratios; however, structural quality degraded significantly at low As/III. (224) reciprocal space mapping showed substantial structural issues at low As/III, with extremely weak QW- and barrier-related diffraction intensities. A broad peak near the GaAs peak was also evident, indicating diffraction from smaller in-plane and larger out-of-plane lattice constants. This may be related to roughening or segregation effects and merits further study. This degradation was also evident in low-temperature PL. Spectra were collected at 25 K under 10 W/cm^2 of excitation. Following the work of Buyanova *et al.*,[23] the low-energy tail was fitted to an exponential decay allowing an estimate of the localization potential from compositional inhomogeneity. This method agrees qualitatively with temperature-dependent PL measurements of the divergence of the PL peak energy from the Varshni equation (for example, Ref. 12). Figure 3 plots the localization potential with the As/III flux ratio. A significant increase in localization effects is evident at reduced As/III, in agreement with theory.[3]

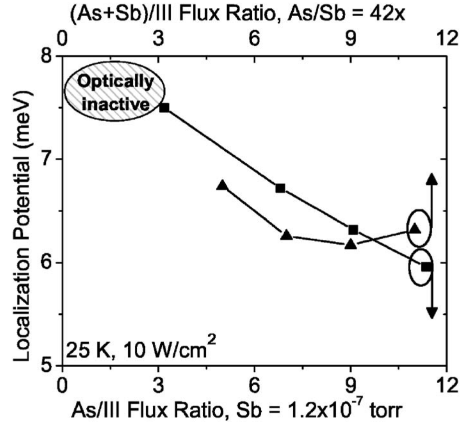

FIG. 3. Localization potentials measured from the PL line shape at 25 K and 10 W/cm^2 excitation. Increased localization is evident at reduced As/III (squares). Localization is not a strong function of the (As+Sb)/III, under constant As/Sb flux ratio of 42 times (triangles).

In the second experiment, the arsenic-and-antimony-to-group-III flux ratio [(As+Sb)/III] was varied, with the arsenic-to-antimony flux ratio (As/Sb) held constant at ~42×, typical of high-quality long-wavelength GaInNAsSb.[9,24] Figure 4 plots the as-grown peak PL intensity and wavelength, as a function of the (As+Sb)/III. The luminescence improved markedly with decreasing (As+Sb)/III; the improvement persisted upon anneal. This likely provides a method to substantially reduce dilute-nitride-antimonide laser thresholds at 1.55 μm.

Little wavelength shift was observed between the samples, consistent with the (004) $\omega/2\theta$ HRXRD scans shown in Fig. 5. Some slight degradation in the QW-related fringes for the 5×(As+Sb)/III sample may indicate minor structural degradation. This was confirmed by localization measurements performed on these samples, shown in Fig. 3.

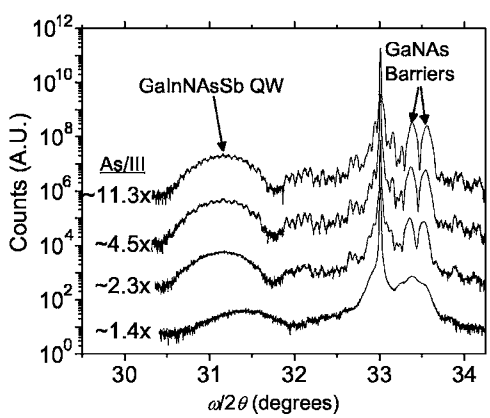

FIG. 2. (004) $\omega/2\theta$ HRXRD scans of representative samples from the As/III study of Fig. 1. Substantial structural degradation was observed at reduced As/III.

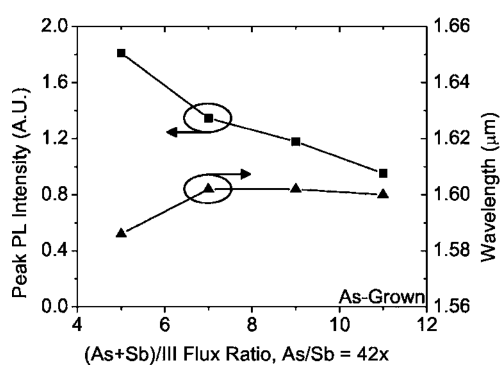

FIG. 4. As-grown peak PL intensity (squares) and wavelength (triangles) for GaInNAsSb QWs grown under various (As+Sb)/III flux ratios, with all other conditions held constant. The As/Sb flux ratio was maintained at 42 ×.

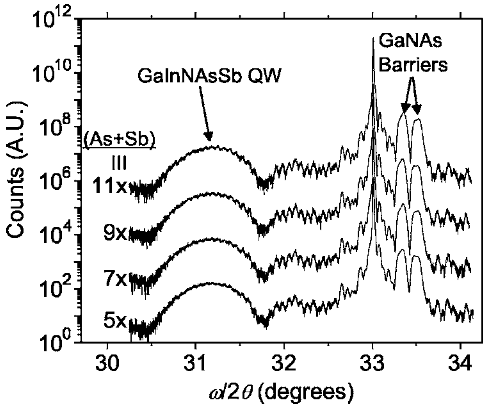

FIG. 5. (004) $\omega/2\theta$ HRXRD scans of samples from the (As+Sb)/III study of Fig. 4. Little degradation was observed at reduced values of (As+Sb)/III.

Comparable localization was observed at elevated (As+Sb)/III but increased moderately for the 5× sample.

The improved luminescence at reduced (As+Sb)/III is consistent with the observations of Jaschke *et al.* for GaInNAs,[11] but is interesting given the substantial degradation observed in the first study at a fixed antimony flux. The antimony flux is only a small fraction of the total group-V flux but has a substantial effect on the optical properties. While further study is required, the cause for the improvement likely lies in the antimony surface concentration and the resulting surface diffusion length (SDL).[25,26] In the first study, the antimony surface coverage increased at reduced As/III, consistent with increased antimony incorporation observed with SIMS. Due to the reactive surfactant properties of antimony, the SDL was likely reduced beyond the optimal point, resulting in decreased structural quality and increased defects. By contrast, a fixed As/Sb flux ratio helped maintain an approximately constant antimony surface concentration, and hence, an appropriate SDL. The reduced arsenic flux then resulted in fewer point defects and increased luminescence, similar to the findings of Jaschke *et al.*,[11] and in keeping with separate studies on the nature of antimony in GaInNAs.[27] These conclusions are reinforced by SIMS measurements that showed no change in composition for samples grown under a fixed As/Sb flux ratio. Because the incorporation kinetics are strong functions of the antimony surface concentration, we may conclude that the antimony surface coverage, and hence the SDL, were nearly constant amongst these samples.

The interrelation between the arsenic and antimony fluxes has been investigated to produce 1.55-μm range GaInNAsSb quantum wells of improved optical quality. The luminescence improved markedly with decreasing (As+Sb)/III ratio, under a constant As/Sb ratio, in contrast to simply modulating the arsenic flux. Further study of the growth kinetics involved would be beneficial; however, the significance to device applications is clear. This technique provides a method to further reduce 1.55-μm laser thresholds from the 450–600 A/cm^2 regime (Refs. 9 and 28) to the 200–300 A/cm^2 range typical of 1.3-μm devices.[29]

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