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Structural and optical studies of nitrogen incorporation into GaSb-based GaInSb quantum wells

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We investigate the incorporation of nitrogen into (GaIn)Sb grown on GaSb and report room temperature photoluminescence from GaInSb(N) quantum wells. X-ray diffraction and channeling nuclear reaction analysis, together with Rutherford backscattering, were employed to identify the optimal molecular beam epitaxial growth conditions that minimized the incorporation of non-substitutional nitrogen into GaNSb. Consistent with this hypothesis, GaInSb(N) quantum wells grown under the conditions that minimized non-substitutional nitrogen exhibited room temperature photoluminescence, indicative of significantly improved radiative efficiency. Further development of this material system could enable type-I laser diodes emitting throughout the (3-5 μm) wavelength range. © 2012 American Institute of Physics. [doi:10.1063/1.3675618]

Mid-infrared (mid-IR) lasers are important for a wide variety of applications including trace gas sensing, infrared countermeasures, and free space optical communications. GaSb-based diode lasers with type-I quantum well (QW) active regions are attractive for lasers operating at mid-IR wavelengths (3-5 μm), due to their relatively simple design as compared to interband cascade1 and intersubband2 cascade devices. Type-I diode lasers have been demonstrated for wavelengths up to 3 μm, utilizing active regions based on GaInAsSb/AlGaAsSb QWs (Ref. 3) and GaInSb/GaSb QWs.4 To achieve longer emission wavelengths without strain relaxation, the arsenic content in the QWs must be increased, however. This in turn decreases hole confinement and degrades laser performance.3–5 Active regions based on the quinary AlGaInAsSb barriers6,7 have enabled lasers operating out to 3.6 μm by improving hole confinement.8 However, laser performance still degrades rapidly with further increase in QW arsenic content.7,8 An attractive approach to extending the emission wavelength further is to incorporate dilute quantities of nitrogen into the QW.9–11 It is well-established that incorporating dilute quantities of substitutional nitrogen leads to a strong reduction in the bandgap, due to band anticrossing12 between the localized level introduced by nitrogen and the host conduction band.13 The advantage for the case of GaSb-based lasers is that the bandgap reduction occurs almost exclusively in the conduction band, leaving the valence band virtually unperturbed, which would be quiet beneficial for maintaining adequate hole confinement in the active region.

While dilute-nitride antimonides grown on GaSb have been previously studied by several groups, the optical quality has proven to be a significant challenge. There are a few reports of photoluminescence (PL) from dilute-nitride antimonides, but only at low temperatures9,10,13 or via sophisticated ultra-fast up-conversion techniques.11 The difficulty in achieving material of sufficient optical quality to observe room temperature luminescence is likely rooted in the mismatch in properties such as atomic size and electronegativity between the nitrogen and antimony atoms, which leads to a significant fraction of nitrogen atoms being incorporated into non-substitutional sites.14 Nitrogen-related non-radiative centers have been observed in the GaAs-based dilute-nitrides by many groups and are typically attributed to non-substitutional nitrogen atoms in the form of N-As and N-N split interstitials.15–17 This issue is expected to be even more challenging in the case of dilute-nitride antimonides, because the mismatch in properties between antimony and nitrogen is greater than that between arsenic and nitrogen. Indeed, Pham et al. suggested that the dominant nitrogen defect in InSb1−xNx is interstitial N–Sb.14 Minimizing the fraction of non-substitutional nitrogen is of paramount importance for improving the optical quality of dilute-nitride antimonides and is the primary motivation for this study. To this end, we have investigated the growth of GaNSb to determine the key parameters that minimize the concentration of non-substitutional nitrogen.

Samples were grown in a Varian Gen. II solid-source molecular beam epitaxy (MBE) system equipped with a Veeco Mark-IV valved-cracker for antimony, Veeco SUMO effusion cells for gallium and indium, and a SVTA rf plasma source for nitrogen. The plasma source was equipped with dc-biased deflection plates at the aperture to electrostatically deflect energetic ions away from the substrate.18 The substrate temperature was measured using a pyrometer. For initial studies of the growth space, 100 nm thick GaNSb films were grown at a variety of growth rates (GRs) and substrate temperatures on undoped (100) GaSb substrates. The antimony to gallium beam equivalent pressure ratio was fixed at 2.3 for GaSb and GaNSb layers. Reflection high-energy electron diffraction (RHEED) indicated that the growing surface exhibited a (1 x 3) antimony-stabilized surface reconstruction. The nitrogen concentration in these films was quantified with high-resolution x-ray diffraction (HR-XRD) and nuclear reaction analysis Rutherford back-scattering (NRA-RBS).

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Figure 1 plots a representative HR-XRD $\omega/2\theta$ scan about the (004) reflection of GaSb, for a 100 nm GaNSb film. The nitrogen content was determined to be 1.8% using dynamical simulation.

Figure 1

Figure 2. (Color online) Variation of apparent nitrogen content measured, as determined from HR-XRD, with growth temperature for two different growth rates. For the 1 $\mu$m/h GR, at low growth temperatures, the nitrogen incorporation transitions to a supply limited regime.

$^{14}$N($^{2}$H, $^{17}$O) reaction with a 3.72 MeV $^{4}$He$^{2+}$ beam$^{24}$ was used for detection of nitrogen. A 150 mm$^{2}$ passivated implanted planar silicon (PIPS) detector with a $3 \times 12$ mm slit was used to detect the emitted protons at 135$^{\circ}$ with respect to the incident beam. A 25 $\mu$m thick mylar foil was placed in front of the detector to absorb the backscattered alpha particles. RBS spectra were also obtained simultaneously at 168$^{\circ}$ with another PIPS detector. Both RBS and NRA measurements were carried out in random and $h_{100}$, $h_{110}$, and $h_{111}$ axial channeling directions. The fraction of substitutional nitrogen atoms in the films was obtained by comparing the random and channeling yields of the RBS (antimony signal from the GaNSb) and the NRA measurements.$^{25}$ Channeling NRA-RBS (c-NRA-RBS) has been applied to investigate the fraction of substitutional nitrogen atoms in GaAs-based dilute-nitrides.$^{23,26}$ Figure 3 shows the total and substitutional nitrogen in our samples as determined by NRA-RBS in the random and channeling directions ($h_{100}$, $h_{110}$, and $h_{111}$). Our results show that the total nitrogen concentration remained constant with increasing growth temperature, as further confirmed by secondary ion mass spectrometry (SIMS) measurements (not shown).

However, the substitutional nitrogen concentration was
found to decrease monotonically with increasing growth temperature. This indicates that the incorporation of nitrogen atoms into non-substitutional sites increases substantially at elevated temperatures. Due to the flux peaking effect in ion channeling, isolated nitrogen interstitials in either the hexagonal or tetrahedral sites can be revealed in the (110) channeling direction in the c-NRA-RBS measurement.24 However, c-NRA-RBS measurements of the GaNSb films in all axial directions showed similar channeling yields, suggesting that the non-substitutional nitrogen atoms are not in the form of isolated interstitials. Non-substitutional nitrogen atoms in these films are most likely present in the form of small random clusters and/or N-Sb and N-N split interstitials.

Based upon these findings, 10 nm thick Ga0.7In0.3Sb(N) QWs with GaSb barriers (Fig. 4) were grown to study optical quality. The QWs were grown at a temperature of 310°C to minimize the fraction of non-substitutional nitrogen-related non-radiative recombination centers. An AlSb carrier blocking layer was inserted between the QW and the sample surface to prevent surface recombination of photo-produced carriers. Samples were characterized using room-temperature PL with a 532 nm diode pumped solid-state laser as the excitation source. The PL was dispersed through a 0.5 m spectrometer and detected with an InSb photodetector. Samples were characterized using room-temperature PL with a 532 nm diode pumped solid-state laser as the excitation source. The PL was dispersed through a 0.5 m spectrometer and detected with an InSb photodetector.

Through studies of the substitutional nitrogen incorporation into GaNSb, we were able to identify the growth space that minimized the incorporation of non-substitutional nitrogen. This has led to a significant improvement in the optical-quality of dilute-nitride antimonide material system, as evidenced by the first demonstration of room temperature PL spectra from this material system. Through further optimization of the growth conditions and extension to other alloy compositions, the emission wavelength of dilute-nitride antimonide QWs can likely be extended to longer wavelengths (>3 μm), with significantly enhanced optical properties.

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