Probing the nature of carrier localization in GaInNAs epilayers by optical methods

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(Received 14 May 2013; accepted 21 June 2013; published online 3 July 2013)

Photoluminescence (PL), optical pumping, and reflectance studies of nominally undoped and p-type GaInNAs epilayers are presented. The PL peak energy of the *nominally* undoped sample exhibits an S-shaped dependence on temperature for $T < 50$ K. This is attributed to recombination of bound excitons localized on traps. The energy of the PL circular-polarization maximum coincides with the energy of the free-exciton related reflectance feature at all temperatures. In heavily p-type samples the S-shaped temperature-dependence of the PL energy disappears, and the PL peak and circular polarization maximum coincide with the reflectance feature at all temperatures, indicating that the PL is free-exciton-like.
[http://dx.doi.org/10.1063/1.4813388]

The III–V material GaInNAs has recently stimulated much interest due to the possibility to grow it lattice matched to GaAs with an absorption edge at 1 eV.[1,2] Such properties offer potential for implementation as a fourth-layer component in the next generation of multi-junction solar cells, with predicted efficiencies greater than 44%.[3] These efficiencies are beginning to be realized;[4] issues remain concerning such dilute nitride materials, specifically with respect to yield and lifetime. The main problem in this system relates to the low solubility of nitrogen in GaInAs and the tendency for phase segregation during MBE growth.[5–7] To incorporate reasonable amounts of nitrogen into the alloys (1%–3% for solar applications), lower temperatures than are optimum for III–V materials growth must be utilized. This reduces the material quality,[3] typically leading to low minority carrier diffusion lengths.[1–3,8] A partial remedy is post-growth annealing (RTA), which has been shown to improve the quality of the material significantly.[9–11] Despite the improvements in the quality of these alloys and their prospect for use in the next generation of multi-junction solar cells,[4] impurities, alloy fluctuations, and defect formation remain as serious problems.[5,9–12]

When GaInNAs material is grown without any specific dopants, one generally observes it to be *p*-type. However, n-type material has also been demonstrated.[3] The carrier sign in nominally undoped material has been attributed to the presence of vacancies and interstitials that arise during the low-temperature growth.[3,5,12] In the present work, we investigate GaInNAs epilayers which are either nominally undoped or doped with beryllium acceptors. We show that at sufficiently high levels of beryllium doping the band edge photoluminescence is due to free excitons at low temperatures, rather than to excitons localized at isoelectronic centers, as is the case in undoped GaInNAs.[3,6] The dominance of free excitons in heavily doped p-type samples, *at all temperatures*, has interesting implications for photovoltaics, because operation in the radiative limit is required for optimum solar cell performance. The results presented below show that optical pumping experiments are very useful for differentiating between free and localized exciton recombination in GaInNAs.

In this paper we present data from two samples. Both are GaInNAs epilayers grown by MBE on GaAs substrates. The atomic composition of both samples is $n_{Ga} = 91\%$, $n_{In} = 9\%$, $n_{As} = 97.2\%$, and $n_N = 2.8\%$. Sample 1 is a nominally undoped 1.6 μm GaInNAs layer grown at a temperature of 450 °C; sample 2 is doped with beryllium acceptors at $n_{Be} = 2 \times 10^{18}\ \mathrm{cm}^{-3}$, with a thickness of 1 μm, and is grown at 420 °C. Both samples are terminated with a 75 nm GaAs cap and are subjected to RTA at 800 °C in nitrogen-rich conditions. Thermo-power measurements show that the background carrier polarity in sample 1 is *n*-type. The dominance of donors in this sample is attributed to out-diffusion of Si from the *n*-type GaAs substrate during annealing and/or growth.

The samples are placed in a variable temperature closed-cycle optical cryostat. Photoluminescence is excited using the 1064 nm line of a Nd:YAG laser. The laser photon energy is 1165 meV and lies between the fundamental E_0 gap at 1100 meV and the split-off $E_0 + \Delta_0$ gap at 1400 meV[13] as required for the observation of optical pumping.[14,15] The laser beam is polarized in the σ_+ (left circular) or σ_- (right circular) sense using a Babinet-Soleil compensator. The emitted light is collected and focused onto the entrance slit of a single monochromator equipped with a cooled InGaAs array detector operating in the 900–1700 nm wavelength range. A liquid crystal quarter-wave plate/linear analyzer combination is placed just before the spectrometer entrance slit and used to separate the σ_+ and the σ_- components of the emitted light, which are recorded separately. The circular polarization P of the luminescence at each photon energy is calculated using the equation: $P = (I_+ - I_-)/(I_+ + I_-)$. Here $I_+(I_-)$ is the intensity of the $\sigma_+(\sigma_-)$ PL

