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Photoluminescence of $InAs_{0.926}Sb_{0.063}N_{0.011}$/InAs Multi-Quantum Wells in the Mid-Infrared Spectral Range

M. de la Mare,*[1] P.J. Carrington,[1] R. Wheatley,[1] Q. Zhuang,[1] R. Beanland,[2] A.M. Sanchez[2] and A. Krier[1]

[1]*Physics Department, Lancaster University, Lancaster, LA1 4YB, UK*

[2]*Department of Physics, University of Warwick, Coventry, CV4 7AL, UK*

Abstract:

We report on the epitaxial growth and photoluminescence of $InAs_{0.926}Sb_{0.063}N_{0.011}$/InAs multi-quantum wells grown using plasma-assisted molecular beam epitaxy. These dilute nitride quantum wells exhibit bright photoluminescence in the mid-infrared spectral range up to a temperature of 250 K without any post-growth annealing. Consideration of the power dependent photoluminescence behaviour are consistent with a type I band line-up in these quantum wells, arising from a strong lowering of the conduction band edge due to N-induced band anti-crossing effects.

Dilute nitride alloys containing a small amount of nitrogen are of particular interest due to the large band-gap reduction,[1] the interesting physics,[2] and the potential for a wide variety of device applications.[3,4] Furthermore, the incorporation of nitrogen

into narrow band-gap semiconductors such as InAs is attracting increasing attention because it can facilitate emission in the technologically important mid-infrared (2-5 μm) spectral range[5]. Dilute nitrides also have some potential for reduction of Auger recombination[6] and inter valence band absorption processes which currently limit the quantum efficiency of bipolar devices at longer wavelengths. However, although InAsN/InGaAs quantum well lasers operating at 2.38 μm have been demonstrated,[7] no mid-infrared light emitters utilizing dilute nitrides have yet been realized, due to the difficulty in growing device quality epitaxial material.[8,9] Photoluminescence emission from bulk InAsN has been reported[10] and associated with strong carrier localization effects arising from disorder.[11] More recently, following further optimization of growth parameters, InAsN with bright PL has been obtained.[12,13] In this respect the growth of InAsNSb alloys is of particular interest due to the potential for producing strain balanced alloys on InAs to further improve material quality. Adding Sb to InAs introduces compressive strain into the quantum well, whereas N results in tensile strain, so that high quality lattice matched material can be envisaged as required for practical devices. Our previous studies of bulk InAsNSb have shown that the addition of Sb to InAsN increased the incorporated nitrogen concentration and also improved the optical quality.[14] The incorporation of nitrogen may also be used to change the quantum well band alignment from type II to type I. To date the only published information on this material is from single quantum wells.[15] In this work, we report the growth and photoluminescence of InAsNSb quantum wells and confirm the band line-up to be type I.

InAsNSb multiple quantum wells (MQWs) were grown on n-InAs (001) substrates using a VG-V80H MBE reactor. Thermal effusion Knudsen cells were used to supply the In flux, two (Veeco) valved cracker cells were used to provide As_2 and Sb_2 and a radio frequency plasma nitrogen source supplied the atomic nitrogen. The preparation and oxide desorption in the growth chamber was carried out in the conventional manner.[16] *In situ* reflection high-energy electron diffraction was employed to control surface reconstruction. The substrate temperature was measured using an infrared pyrometer calibrated using surface reconstruction transitions at a fixed As flux. The growth of the MQWs was calibrated using the same growth method of previously grown InAsSbN bulk layers.[14] Optimization of the growth conditions for this material led to high crystalline quality epitaxial material, with weak localisation effects and a low residual carrier concentration of 2×10^{17} cm^{-3}.[17]

In the present structures, a 200 nm thick un-doped InAs buffer layer was first grown at 480 °C, the temperature was then lowered to 420 °C for the growth of the ten period $InAs_{0.926}Sb_{0.063}N_{0.011}$/InAs MQW region using a growth rate of 0.5 μm/hr. The thickness of the InAsSbN quantum wells and InAs barriers was 12 and 24 nm respectively, so that the strain in the QW was approximately 0.3%. Finally, a 100 nm thick un-doped InAs capping layer was grown. For the active region a 6×10^{-6} mbar N flux was used at a plasma power of 160 W. The As flux was kept to a minimum for the growth of the InAs layers, because excess As flux has been shown to reduce the N incorporation in the epilayer since the adatom sites are more favourable for As than they are for N.[18] The structural quality of the resulting epilayers was characterised by high resolution x-ray diffraction (HRXRD) measurements using a Bede QC200 diffractometer. Photoluminescence (PL) measurements were

performed on all samples over the temperature range 4 to 300 K using an Oxford Instruments variable temperature continuous flow He cryostat. An Ar^+ ion laser (514nm, with maximum power density of 20 Wcm^{-2} at the sample) was used for excitation. The emitted radiation was collected using CaF_2 lenses and focused into a 0.3m Bentham M300 grating monochromator. The radiation was detected using a cooled (77 K) InSb photodiode detector and a Stanford Research (SR850) digital lock-in amplifier.

Figure 1 shows a comparison of the (004) x-ray diffraction spectra for $InAs_{0.987}N_{0.013}$, $InAs_{0.924}Sb_{0.076}$ and $InAs_{0.926}Sb_{0.063}N_{0.011}$ MQWs grown on InAs substrates. The addition of N to InAs layers introduces tensile strain into the layer while the addition of Sb introduces compressive strain, these two strain offsets can be seen from the central peak position in the XRD spectra of fig. 1. When N is added to an InAsSb layer the compressive strain of the material is reduced, this can be seen from the blue (lower) rocking curve in fig. 1, this may allow for the production of strain balanced material with high enough N concentration. The sample containing only nitrogen has a small number of very weak, broad satellite peaks. By comparison the InAsSb sample shows a large number of intense satellite peaks characteristic of abrupt interfaces and good crystalline quality. The InAsSbN sample shows a few distinct satellite peaks which are narrower with a half-width of approximately 65 arc-seconds compared to 128 arc-seconds for the InAsN suggesting that the crystalline quality of the InAsSbN MQWs has been improved compared to that of the InAsN MQWs, although they are not as abrupt as the InAsSb QW. This is consistent with the number of satellite peaks present for each sample as shown in fig. 1 and is in agreement with published data relating to strain balancing effects[14,19]. The InAsSbN alloy composition was determined from energy dispersive x-ray microanalysis measurements on a 1µm-thick InAsSbN epilayer grown under identical conditions. The results agreed closely with simulation of the x-ray diffraction data. The inset to figure 1 shows a TEM image where the 10 InAsSbN QW are clearly evident. However, the interfaces of these wells exhibit a diffuseness of up to 3 nm It is quite likely that thenitrogen is the cause of this reduction in interface quality, since we have previously reported InAsSb/InAs MQW with high quality interfaces.[20] It is possible that further increase in the Sb content could improve the interface quality and clearly further work is needed to optimise the abruptness of the QW interfaces and the overall quality of the InAsSbN.

The dependence of the 4K PL on laser excitation power for the InAsSbN MQW is shown in fig. 2. The peak intensity is observed at a wavelength of 3.62 µm with a shoulder at3.48 µm. In the bulk alloy the peak was observed at 4.38 µm, which is a good indication of strong quantization. Increasing laser excitation power induces a significant blue-shift of the emission wavelength for a type II system.[19]

However, we observed only a modest blue-shift which indicates that the band line-up in our MQW is type I. The inset of fig. 2 shows the spectrum of the PL emission peak. This was de-convoluted as two Gaussian peaks associated with e_1-hh_1 and e_1-lh_1

transitions which are each subject to inhomogeneous broadening. Small variations in either the composition associated with N inhomogeneities, or well width associated ith structural disorder, (as indicated in the inset to fig. 1), could result in such broadening causing an increase in linewidth of the spectral peak[21]. The linewidth of the InAsSbN e_1-hh_1 transition measured at 4 K was found to be 21 meV while the same transition for InAsSb was found to be 11 meV. (No PL was observed from the InAsN/InAs MQWs). The increase in linewidth is therefore most likely to be due to the N incorporation although as mentioned above the MBE growth parameters are not yet fully optimised.

Fig. 3 shows the temperature dependent PL spectra obtained from our InAsSbN MQW. A blue-shift in the emission wavelength of dilute nitrides with increasing temperature is associated with strong localisation caused by the filling of the energy states in the band tails[18]. Only a very small blue-shift was observed in our material, which is indicative of type I QW and is consistent with quite weak localisation (~ 4 meV) associated with N inhomogeneities in our QWs as shown in fig. 4.[22] Above about 80 K electrons have sufficient energy to be excited into the conduction band continuum so that the primary recombination mechanism becomes band-band. Returning to fig.3, three peaks can be de-convoluted in the spectra of above 60 K; one near 3.2 μm associated with the InAs barrier emission and two peaks originating from recombination of electrons and holes in the InAsSbN MQW. At 4 K the two peaks at 3.48 μm (356 meV) and 3.67 μm (338 meV) correspond to e_1-hh_1 and e_1-lh_1 transitions respectively (-see fig. 5). Although the PL does not persist up to room temperature, we observed emission up to 250 K which is a significant improvement in temperature quenching compared with that previously reported for InAsN MQWs.[23,24]

Figure 5 shows a schematic diagram of the $InAs_{0.926}Sb_{0.063}N_{0.011}$/InAs quantum well. This was obtained by firstly calculating the InAsSb/InAs band alignment and then modifying it due to the effects of nitrogen. The $InAs_{0.937}Sb_{0.063}$/InAs band alignment was calculated using the Krijn model[25,26] with a band bowing parameter of 0.67 while the material parameters such as hole effective masses were obtained by linear interpolation between bulk InAs and InSb values using Vegard's law. $InAs_{0.937}Sb_{0.063}$/InAs exhibits a type II band alignment with conduction and valence band offsets of ΔE_c = 39 and ΔE_v = 82 meV respectively with 0.5% compressive strain in the QW. The addition of 1.2% nitrogen reduces the strain to 0.3% which reduces the conduction and valence and band offsets to ΔE_c = 29 meV and ΔE_v = 76 meV respectively. The conduction band is then further lowered by 63 meV as estimated from the band anti-crossing model for InAsN[13,27,28] resulting in a type I QW with a conduction band offset of ΔE_c = 34 meV. The eigenvalues of the confined electron and hole levels were calculated using a single band Schrödinger solver for a symmetrical rectangular quantum well. The hole masses used were $0.366m_e$ and $0.024m_e$ and an electron effective mass of $0.06m_e$ was used based on previously published data.[17,29] The transition energies for e_1-hh_1 (368 meV) and e_1-lh_1 (329 meV) which we obtained are in quite good agreement with the experimentally measured values obtained from the PL spectra in fig 3.

In summary, $InAs_{0.926}Sb_{0.063}N_{0.011}$/InAs MQWs were successfully grown by MBE and found to exhibit PL emission in the mid infrared spectral range around 3.6 µm at 4 K. The absence of any significant blue-shift in the power dependence of the spectra and the good agreement with calculation confirms the type I QW band alignment. Improvements to the structural quality to create more abrupt QW interfaces could be realized by optimizing the As and Sb flux switching sequence[30]. The conduction band offset in this QW is insufficient to enable it to be directly implemented as the basis of the active region for the development of a diode laser and further work is needed to address this point. Nevertheless, this material system shows promise since it provides a valuable method of tailoring the band structure and an different means of achieving a type I QW system which could subsequently be used to access the technologically important mid-infrared spectral range.

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Figure Captions

Fig 1. (Colour online) High resolution x-ray diffraction spectra of InAsN (black), InAsSb (red) and InAsSbN (blue) MQWs. Insert (a) shows a TEM image of the InAsSbN MQW while (b) shows an enlarged image having interface diffuseness~ 3 nm The green arrows show the position of the zero order MQW peak.

Fig 2. (Colour online) Power dependence of the PL from InAsSbN MQW at 4 K. The arrows are guides to the eye and represent the e_1-hh_1 and e_1- lh_1 transitions (see fig.5). The inset shows the corresponding de-convolution of the spectrum taken at 1.8W laser power.

Fig 3 (colour online) Temperature dependent PL spectra of $InAs_{0.926}Sb_{0.063}N_{0.011}$ /InAs MQW. Dotted lines are guides to the eye.

Fig 4. (Colour Online) *T*-dependence of the PL peak energy for e_1-lh_1 (Black squares) and e_1-hh_1 (Red circles). The continuous lines are the calculated *T*-dependence of the transition energy according to the empirical Varshni equation.

Fig 5. Calculated transitions in the $InAs_{0.926}Sb_{0.063}N_{0.011}$/InAs quantum well at 4K

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