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Citation: *Journal of Applied Physics* **103**, 063520 (2008); doi: 10.1063/1.2896638

View online: <http://dx.doi.org/10.1063/1.2896638>

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Room temperature photoluminescence at 4.5 μm from InAsN

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(Received 27 November 2007; accepted 26 January 2008; published online 26 March 2008)

Nitrogen incorporation in InAsN epilayers grown by radio-frequency plasma-assisted molecular beam epitaxy was investigated as a function of growth conditions. Reduced growth rate, growth temperature, and arsenic flux significantly enhance the nitrogen incorporation. Optimal growth conditions allowed us to obtain high quality InAsN with nitrogen composition of up to 2.5%. The epilayers exhibit intense 4 K photoluminescence (PL) with double-peak features, which were attributed to free carrier recombination and localized carrier recombination. Strong room temperature PL emission up to a wavelength of 4.5 μm is obtained. © 2008 American Institute of Physics. [DOI: 10.1063/1.2896638]

Dilute nitride alloys have attracted increasing attention over the last decade due to the large bandgap reduction with the incorporation of a small amount of nitrogen into the host III-V compounds.¹ They are recognized to be promising materials for a wide variety of device applications in the near-infrared (IR) spectral range.² The narrower bandgap dilute nitrides, such as InAsN, were recently realized to be suitable for optoelectronic devices operating in the mid-IR spectral range of 3–5 μm .³ InAsN epilayers grown on GaAs (Refs. 4–6) and InP (Ref. 7) have been reported. Although lasers containing highly strained InAsN/InGaAs quantum wells grown on InP by radio-frequency (rf) plasma-assisted molecular beam epitaxy (MBE) operating at 2.38 μm have been reported,⁸ the study of InAsN growth and properties is still in its infancy. It is acknowledged that the quality of InAsN severely degrades with the introduction of nitrogen because of the difficulty in growing phase-segregation-free epilayers⁹ and the formation of dinitrogen In–N complexes.¹⁰ Veal *et al.*¹¹ reported the observation of a broad and weak photoluminescence (PL) emission at a wavelength of 3.5 μm at 77 K for the InAsN epilayer containing nitrogen of up to 2.2%. The spectral features showed the severe degradation of the epilayers and to the best of our knowledge, no PL emission was reported for higher nitrogen composition. Additionally, it has been reported that nitrogen is more difficult to incorporate into InAs than into GaAs.¹²

In this paper, we investigate the incorporation of nitrogen into InAs as a function of growth conditions. We found that the nitrogen composition is strongly dependent on the growth parameters: growth rate, growth temperature, and As flux. Based on these results, we successfully synthesized high quality InAsN containing nitrogen of up to 2.5%. Intense 4 K PL was observed for all the samples with a particular feature of a double peak in the emission. Strong room temperature PL emission at a wavelength of 4.5 μm was obtained. The origins of the double-peak emission were identified by temperature dependent PL and laser excitation power dependent PL measurements.

A set of samples consisting of 200-nm-thick InAsN were grown on InAs substrate by MBE using a Veeco rf plasma nitrogen source, at fixed plasma powers of 160 and 210 W with N_2 flux of 5×10^{-7} mbar. To investigate the nitrogen incorporation, growth rate, growth temperature, and As flux were separately varied in the range of 0.2–1.0 $\mu\text{m}/\text{h}$, 360–460 °C, and 0.6–10 times of minimum As flux (referred to the critical As flux for As-terminated InAs growth), respectively. Double crystal x-ray diffraction (DCXRD) was employed to determine the nitrogen composition and crystalline quality of the epilayers. 1- μm -thick InAsN epilayers were grown for an enhanced PL study using an Ar^+ ion laser (514 nm) which produced excitation power of up to 1.6 W with a spot diameter of 1 mm on the sample. Temperature dependent PL measurements over the range of 4–300 K and power dependent PL measurements at 4 K in the range of 0.2–1.6 W were carried out using conventional lock-in techniques. Additional measurements were made with a Bruker IFS 66/S Fourier transform IR spectrometer (FTIR) and a 77 K InSb photodiode detector using a step-scan and a lock-in technique to achieve better resolution at high temperature.

Nitrogen composition determined by DCXRD as a function of growth parameters is shown in Fig. 1. The nitrogen incorporation is strongly dependent on the growth conditions. Figure 1(a) shows that the nitrogen composition is roughly inversely proportional to the growth rate for the samples grown at a temperature of 420 °C and using minimum As flux. Reducing the growth rate from 1.0 to 0.2 $\mu\text{m}/\text{h}$, the nitrogen composition increases from 0.25% to 1.25%. The incorporation rate (defined as nitrogen composition multiplied by growth rate) is almost constant over the growth rate range of 0.2–1.0 $\mu\text{m}/\text{h}$. This behavior indicates that the nitrogen radicals incorporate into InAs like dopant atoms and is consistent with the observations in MBE grown GaAsN and InGaAsN.^{13–15} The effect of growth temperature on nitrogen incorporation is shown in Fig. 1(b). The optical properties of the samples were monitored by PL measurements after growth. At temperatures of 460 °C and above, no nitrogen is incorporated into InAs. Reducing the temperature, the nitrogen composition increases and satu-

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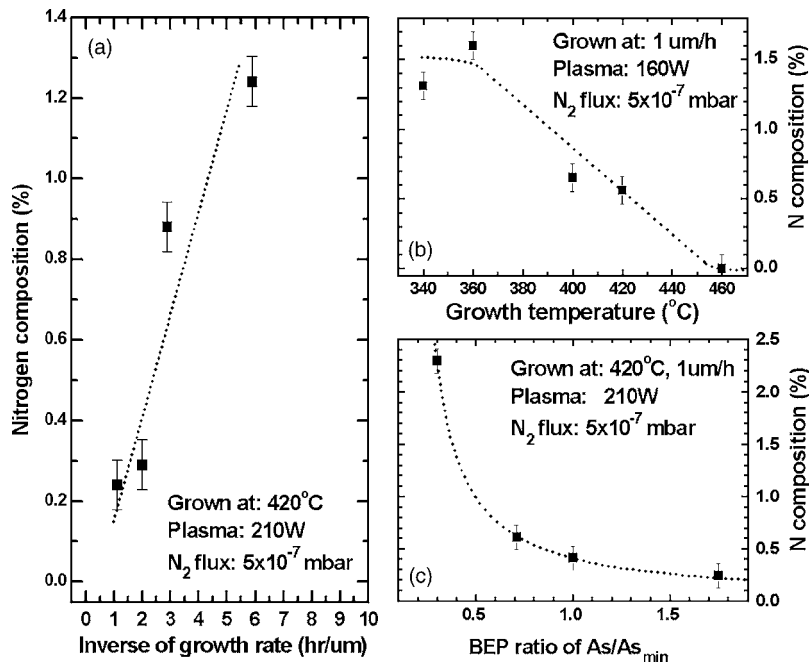


FIG. 1. N incorporation as a function of growth rate (a); growth temperature (b) and ratio of As flux to minimum As flux (c). Lines are guide to the eye.

rates at temperatures around 380 °C while maintaining PL emission. However, no InAsN related PL emission was observed for the samples grown at temperatures ≤ 340 °C. This dependence of nitrogen composition on the growth temperature in InAsN is similar to the dependence observed in GaAsN, which was attributed to the reduced nitrogen segregation on the surface¹⁶ and is in a good agreement with a thermodynamic model.¹⁷

The As flux supply has a significant effect on the nitrogen incorporation, as shown in Fig. 1(c). Excess As flux reduces the nitrogen incorporation because the adatom sites are more favorable for As.⁹ Reducing the As flux could enhance nitrogen incorporation by approximately three times without degradation of the optical properties of the alloy; whereas insufficient As flux causes poor PL emission due to the formation of nonradiative centers of As vacancies. This investigation pointed out the key growth conditions to en-

hance the nitrogen incorporation: low growth rate, reduced growth temperature, and low As flux. Additionally, to maintain good optical properties, we stress that a growth temperature above 380 °C and an As supply above 0.7 times the minimum As flux are required.

A set of 1- μm -thick InAsN epilayers with different nitrogen compositions was successfully obtained for PL study. All the samples were grown at growth temperature of 380–420 °C, growth rate of 0.3–1.0 $\mu\text{m}/\text{h}$, and at close to the minimum As flux. Figure 2 shows the DCXRD rocking curves from the epilayers. Table I gives a summary of the growth conditions, nitrogen composition, and DCXRD full width at half maximum (FWHM). All the samples exhibited a narrow epilayer peak which reflects the high crystalline quality of the material. Particularly for the InAsN epilayer containing nitrogen of 2.52%, a FWHM as narrow as 47 arc sec indicates a good crystalline quality of the material compared with previously reported InAsN containing nitrogen of 2.2% (FWHM ~ 145 arc sec).¹

The 4 K PL spectra from the samples are shown in Fig. 3(a). All the samples demonstrate intense PL emission and the wavelength is redshifted for higher nitrogen composition. This observation is consistent with a previous report which indicates a clear bandgap reduction with nitrogen incorporation.¹¹ Additionally, all the spectra exhibit the fea-

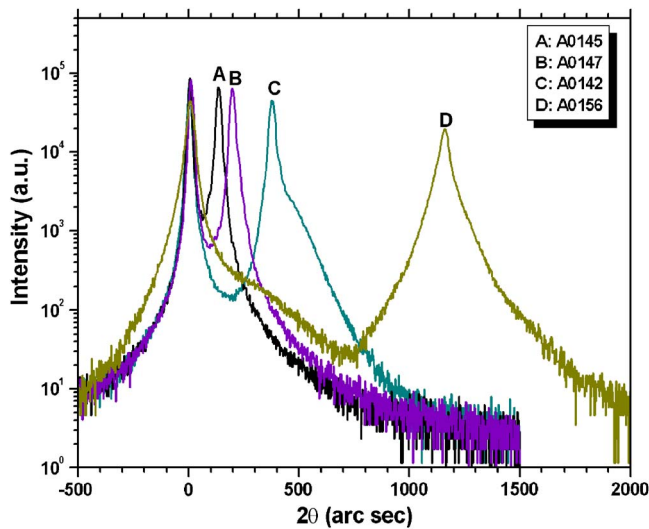


FIG. 2. (Color online) DCXRD rocking curves from a series of InAsN epilayers.

TABLE I. Summary of growth conditions of 1- μm -thick InAsN epilayers (growth temperature T_g and growth rate G/R), the nitrogen composition and DCXRD epilayer peak width (FWHM).

Sample	Growth conditions			FWHM (arc sec)
	T_g (°C)	G/R ($\mu\text{m}/\text{h}$)	N composition (%)	
A0145	420	0.5	0.29	25
A0147	420	1.0	0.41	29
A0142	420	0.3	0.83	32
A0156	380	0.3	2.52	47

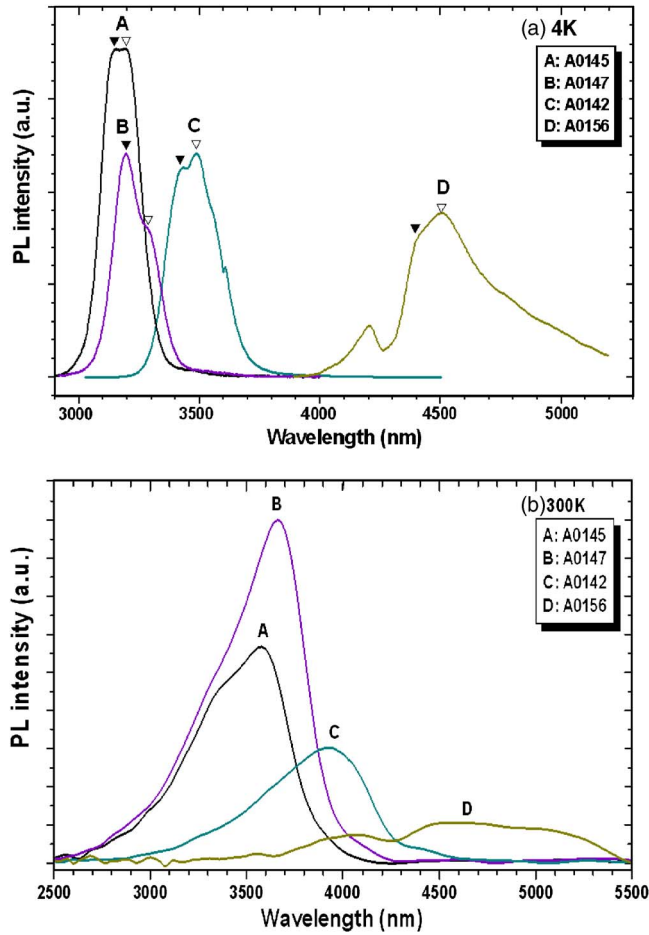


FIG. 3. (Color online) PL spectra from a series of InAsN epilayers at 4 K (a) and at room temperature (b) (measured by FTIR). Solid and open symbols indicate peak positions of short wavelength and long wavelength emission, respectively.

ture of double-peak emission (the dip around $4.3 \mu\text{m}$ in sample D is due to atmospheric CO_2 absorption in the optical path). It is worth noting that strong room temperature PL emission is evident for all the samples, as shown in Fig. 3(b) and a long wavelength of $4.5 \mu\text{m}$ is obtained. This observation reflects the high quality of the alloy with nitrogen composition of up to 2.5%, which is in agreement with DCXR measurements. The feature of double-peak emission at 4 K is different from previously reported PL spectra on GaAsN (Ref. 18 and 19) and GaInAsN.^{20,21} Dilute nitride materials generally exhibit a *single, asymmetric* emission peak with a long low-energy tail at low temperature. This single peak PL emission is generally associated with the formation of localized states in the material due to the inhomogeneity of the nitrogen distribution and always exhibits a strong *blueshift* with increasing laser excitation power or temperature.^{18–21} Veal *et al.*¹¹ observed a broad single peak emission from InAsN at 4 K and assigned it to the contribution from localized excitons combined with a contribution from free excitons. In our case, we suppose that the short wavelength emission and the long wavelength emission originate from free carrier recombination and localized carrier recombination, respectively. The high quality of the alloy certainly improves the homogeneity of the nitrogen distribution so that the localized states are shallower. The photogenerated carriers are less trapped by the localized states and the contribution from free carrier recombination was clearly visible at 4 K.

To further identify the origins of the double-peak emission at 4 K, PL measurements with increasing laser excitation power and at varying temperatures were performed. As shown in Fig. 4(a), with increasing excitation power, the short wavelength emission develops more rapidly than the long wavelength emission. This is indicative of carrier detrapping from localized states. This detrapping leads to a

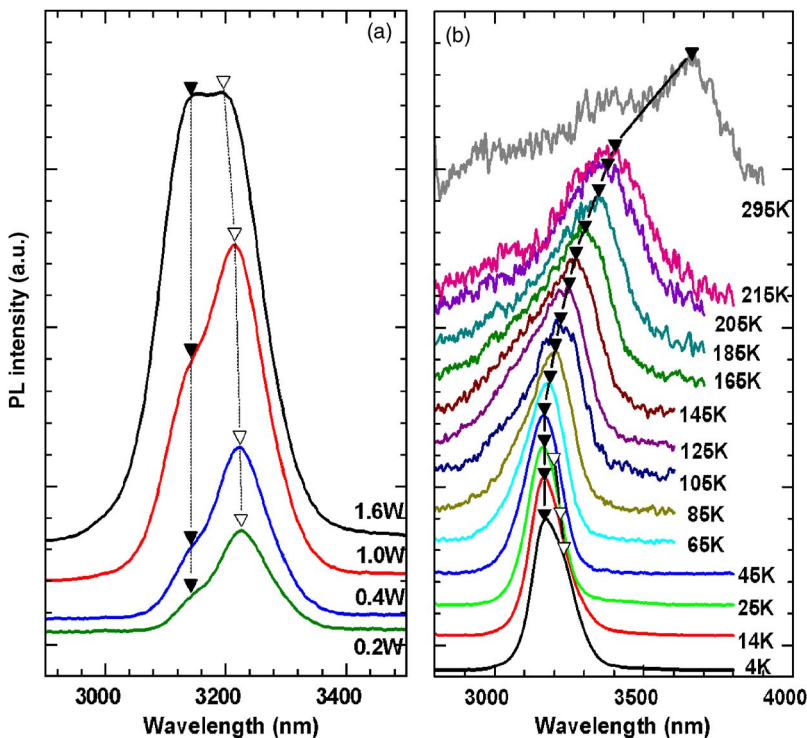


FIG. 4. (Color online) Laser excitation power dependent PL spectra at 4 K (a) and temperature dependent PL spectra (b) for sample A0145. Solid and open symbols indicate the peak positions of short wavelength and long wavelength emission, respectively.

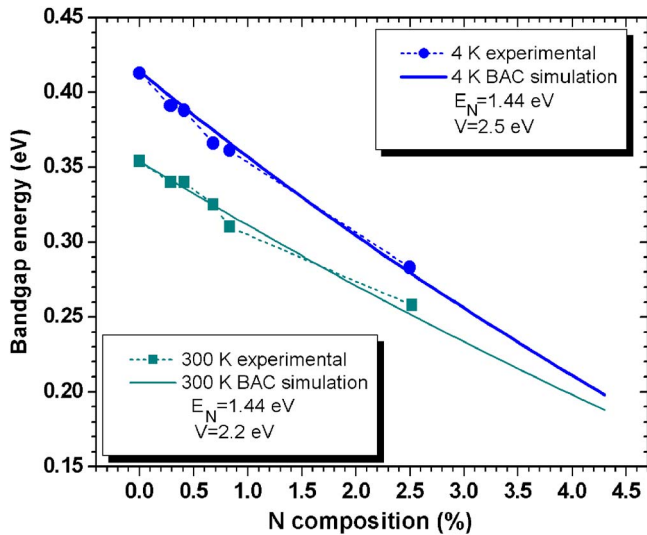


FIG. 5. (Color online) The dependence of InAsN bandgap energy at 4 and 300 K on nitrogen composition determined by PL measurements. The solid lines are the simulation based on the BAC model and the parameters of E_N and V , the best fitting are given

slight blueshift of peak energy of the long wavelength emission with increasing excitation power. A similar behavior was observed in temperature dependent PL spectra. With increasing temperature, the short wavelength emission intensity dramatically increases relative to the long wavelength emission whereas the long wavelength emission quenches rapidly and is suppressed completely at a temperature of ~ 45 K. With increasing temperature, the localized carriers are detrapped to the extended states and contribute to the free carrier recombination. The PL peak wavelength of the short wavelength emission is constant up to 45 K and then shows the usual redshift due to bandgap reduction with increasing temperature. The long wavelength emission exhibits a slight blueshift with increasing temperature, which is a typical behavior of PL emission associated with localized carrier recombination. This observation further clarifies the interpretation of the double-peak emission.

The dependence of bandgap energy of InAsN at 4 and 300 K on nitrogen composition determined by the energy of short wavelength peak in PL spectrum is shown in Fig. 5. The bandgap reduction is well described by band anticrossing model (BAC) for a range of dilute nitrides including GaAsN, GaInAsN, and InPN.²² In the model, the interaction of N states with the extended states of the semiconductor matrix yields the formation of two conduction subband E_- and E_+ given by

$$E_{\pm} = 0.5[(E_N + E_C) \pm \sqrt{(E_N - E_C)^2 + 4V^2x}],$$

where E_C is the bandgap of host semiconductor, E_N is the energy of the N state, V is the interaction potential between the two subbands, and x is the N fraction. Simulations result in 1.44 eV of E_N for 4 and 300 K dependence, 2.5 and 2.2 eV of V for 4 K, and 300 K dependence, respectively. These values agree with the reported values.²³ The higher value of E_N for the dependence at 4 K indicates stronger interaction between nitrogen states and host material's con-

duction band, which gives larger bandgap reduction at 4 K than at 300 K. The bandgap reduction observed at 300 K in our experiment agrees well with the previously reported values which were deduced from optical absorption.^{24,25}

In summary, we report the effect of growth conditions on nitrogen incorporation and present the key growth parameters needed to increase the nitrogen composition. High quality InAsN epilayers with nitrogen composition up to 2.5% were obtained. Intense double-peak PL emission was observed at 4 K and the origins of the emission were interpreted. A record strong room temperature PL emission at a wavelength of 4.5 μm was observed.

The authors wish to thank EPSRC for supporting this research project and for providing the award of a Ph.D. studentship.

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