

REPORT DOCUMENTATION PAGE			Form Approved OMB NO. 0704-0188	
Public Reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comment regarding this burden estimates or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.				
1. AGENCY USE ONLY (Leave Blank)		2. REPORT DATE 07/03/04		3. REPORT TYPE AND DATES COVERED Final 11/01/01-10/31/03
4. TITLE AND SUBTITLE OPTICAL STUDIES OF GaAsSbN ALLOYS AND THEIR QUANTUM WELL HETEROSTRUCTURES			5. FUNDING NUMBERS <del>DAAD19-02-0004</del> DAAD19-02-1-0004	
6. AUTHOR(S) DR. SHANTHI IYER				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) DEPARTMENT OF ELECTRICAL ENGINEERING, NORTH CAROLINA A & T STATE UNIVERSITY, 1601 EAST MARKET STREET, GREENSBORO, NC -27411			8. PERFORMING ORGANIZATION REPORT NUMBER 3	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) U. S. Army Research Office P. O. Box 12211 Research Triangle Park, NC 27709-2211			10. SPONSORING / MONITORING AGENCY REPORT NUMBER 42995.1-EL-H	
11. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.				
12 a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.			12 b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) In this work, the growth and characterization of GaAsSbN quantum wells (QWs) grown in an elemental solid source molecular beam epitaxy (MBE) system with a RF plasma nitrogen source is presented. A systematic study has been carried out to determine the influence of various growth conditions such as the source shutter opening sequence, substrate temperature, various nitrogen (N) pressures and annealing in various ambient on the optical and structural properties of the QWs. N and Sb incorporations were found to depend strongly upon both the substrate temperature and source shutter opening sequence. The substrate temperature in the range of 430-470°C was found to be optimum. The presence of Pendullusong and high frequency fringes due to the cap layer were observed in high-resolution x-ray diffraction (HRXRD) spectra of QWs, for N composition variation up to 2.3%, attesting to the excellent structural quality of the grown layers and interfaces. With increase in N concentration up to 0.7% the energy gap decreases at a rate of 270meV/% change in N, while with further increase in N concentration the change in PL peak energy reduces to 30meV/% change in N. Narrow PL FWHM of 20 meV at PL peak energy of 0.82 eV at 4K has been achieved on annealed samples. The PL temperature dependence studies carried out on these samples indicate evidence of "inverted s-shaped" behavior in the low temperature regime, being more pronounced in samples annealed in a N ambient. The samples annealed <i>in situ</i> under an As overpressure exhibit less carrier localization, which is attributed to a reduction in the nonradiative recombination centers.				
14. SUBJECT TERMS Quantum well, GaAsSbN, Molecular Beam Epitaxy, photoluminescence, x ray diffraction, dilute nitrides, antimonide nitrides			15. NUMBER OF PAGES 65	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OR REPORT UNCLASSIFIED		18. SECURITY CLASSIFICATION ON THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL

NSN 7540-01-280-5500

Standard Form 298 (Rev.2-89)

Prescribed by ANSI Std. Z39-18

298-102



*FINAL REPORT*  
*to*  
*ARMY RESEARCH OFFICE*

**OPTICAL STUDIES OF GAASSBN ALLOYS  
AND THEIR QUANTUM WELL  
HETEROSTRUCTURES**

Grant No: DAAD19-02-0004

Period: 11/01/01-10/31/03

Program Manager: Michael Gerhold

PI:

Shanthi Iyer

Department of Electrical Engineering  
North Carolina A&T State University  
Greensboro NC 27411

## Table Of Contents

<u>Title:</u>	Page
Memorandum of transmittal .....	1
Report and Documentation.....	2
I. Introduction.....	4
II. Objective and Background.....	5
III. Research Activities and Major Accomplishments.....	6
A. Comprehensive Study Of GaSb epilayers grown on InSb.....	10
B. Annealing Effects on the Temperature Dependence of the Photoluminescence of GaAsSbN SQWs .....	35
C. Effect of Nitrogen on GaAsSbN/GaAs SQWs .....	53
IV. Publications & Thesis arising from ARO .....	63
V. Participating Scientific Personnel and Reports Submitted .....	65

**Grant# DAAD19-02-0004, funded for \$260,000 from  
11/01/01-10/31/03**

**Project Title: OPTICAL STUDIES OF GaAsSbN ALLOYS AND THEIR  
QUANTUM WELL HETEROSTRUCTURES**

This is a final technical report describing the research activities of the above ARO grant. The two-year grant period began on November 1, 2001 and was completed on a timely manner on October 31, 2003. This program involved the effort of one faculty member, five graduate students and one undergraduate student. Amongst these is an Air Force veteran and one female graduate student. During this period, there has also been a collaborative effort with other institutions. Discussions and suggestions of Dr. Ralph Dawson from University of New Mexico on Molecular Beam Epitaxy (MBE) growth were extremely helpful. Transmission electron microscopy of selected samples was carried out by Dr. Gerd Duscher from NC State University, x-ray diffraction measurements by Kevin Matney from Bede Inc., and Dr. K.K. Bajaj assisted in the theoretical interpretation of the experimental data. These collaborations were very fruitful and permitted the rapid advances outlined in this report.

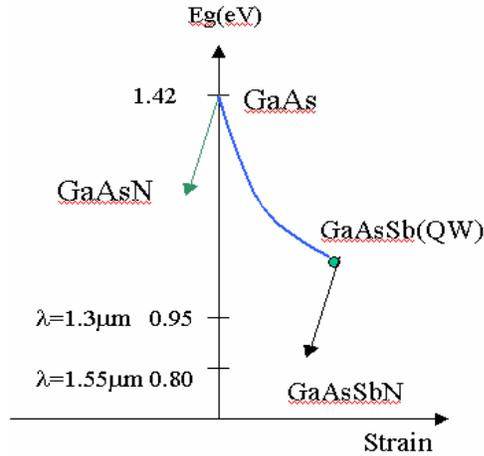
Funding from this grant also allowed leveraging additional funding for the MBE lab from NASA by my colleague, Dr. Ward Collis, on nitride-based solar cells.

**I. INTRODUCTION**

Dilute nitrides are an exciting new class of semiconductors based on combining GaN (wurtzite crystal) with GaAs (Sb) (zincblende crystal). In addition to differences in crystal structure, there is a tremendous bowing in the band gap energy as a function of composition as shown in Figure 1. [1]. This behavior has been explained using a two level band anti crossing (BAC) model for the variation of band gap energy as function of composition. However, most of the work has been concentrated on III-V arsenide-nitrides, namely GaAsN and InGaAsN alloy systems. The latter is a low band gap material and corresponds to the device operating in the near infrared regime. However to achieve wavelengths close to 1.55  $\mu\text{m}$  requires 5% nitrogen [2], which exceeds the solid solubility limit. The more recent approach on the growth has been on the pentanary alloy InGaAsSbN where Sb is used as a surfactant. This system has been somewhat successful and lasers with emission wavelengths as long as 1.49  $\mu\text{m}$  have been reported. [3].

However, Sb based systems such as GaAsSbN remain still attractive as they have definite advantages over In based systems largely due to the following factors, namely:

1. In GaAsSbN there is only one group III element and therefore N can only bond to Ga; a major difference compared with InGaAsN. This leads one to expect that the level of incorporation of N in GaAsSbN may be very similar to that in GaAsN. In addition, there is no disorder on the cation lattice but only on the anion lattices, in contrast to InGaAsN where both the cation and anion lattices have disorder.
2. The N incorporation seems to be relatively insensitive to the Sb mole fraction in the system [4]. This behavior is in contrast with the strong decrease in N incorporation in the presence of In as described earlier.



**Figure.1** Reduction of the band gap and lattice parameter in III-V system on introduction of N. [1]

3. If the strained layers of InGaAs and GaAsSb are compared, they have very similar band gaps and lattice parameter dependency corresponding to an equivalent composition of In or Sb in the usable range (0-40%) with the bowing coefficient in the case of GaAsSb being somewhat higher. However, the band-offset configurations are completely different. Strained InGaAs/GaAs has a large conduction band offset, while GaAsSb/GaAs exhibits a large valence band (VB) offset. The cumulative effect of above characteristics results in lower transition energies (~150 meV lower) and longer wavelengths for GaAsSb/GaAs quantum wells (QW) for the same compressive strain. Hence for the same N composition, GaAsSb exhibits a higher wavelength emission than InGaAs system.
4. In the GaAsSbN/GaAs system, variations in the Sb composition affects the VB offset while N composition primarily affects the conduction band offset. Hence one can tune the conduction band and valence band offsets independently making this system very amenable to band gap engineering.

The features mentioned above make GaAsSbN alloys and their heterostructures very attractive for investigation for efficient light emission applications in the long wavelength region.

The work on this system has been carried out primarily by Ungaro and Harmand [5, 6], a French group. N incorporation (as much as 1.6%) has been successfully accomplished in this material system by Ungaro et al. [6] using MBE technique and the band gap can be reduced by as much as 0.4 eV in GaAsSbN systems. The N incorporation rate is found to be somewhat higher in this system than in arsenide-nitride systems [7].

## II. OBJECTIVE AND BACKGROUND

This program involved a comprehensive study of GaAsSb/GaAs and GaAsSbN/GaAs quantum well heterostructure, ranging from MBE growth study to characterization, with a view to implement 1.55  $\mu\text{m}$  and longer wavelength laser sources on GaAs. During the initial period of the grant, efforts were directed towards installation of the EPI UNI-Bulb RF source in the EPI 930 MBE system and setting up the necessary characterization, namely, low temperature photoluminescence (PL), x-ray simulation and

an annealing station. The experimental configuration for the nitrogen supply is as shown in Fig.1. This grant provided only partial funding towards the equipment, and the rest came from the Department.

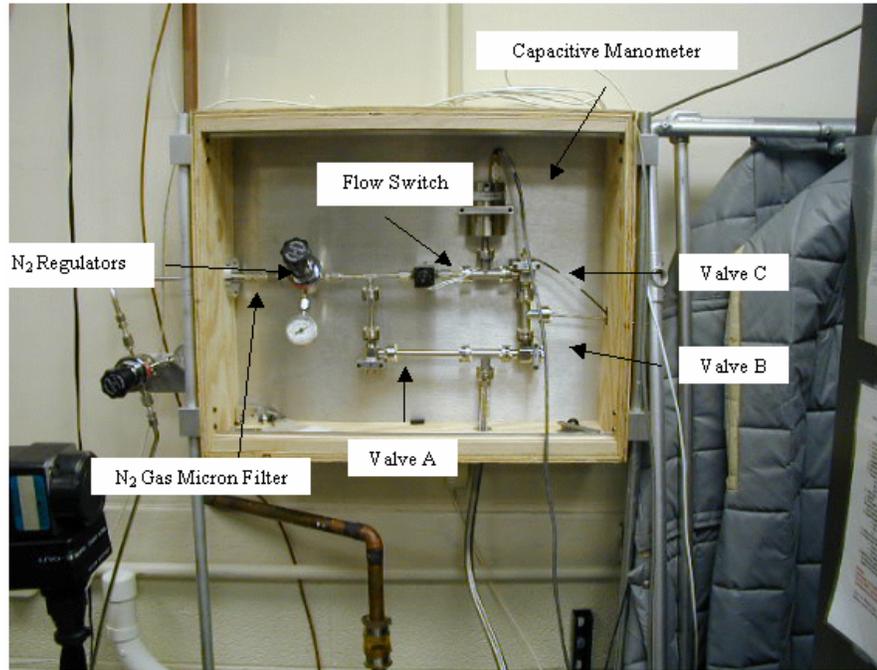


Figure 1: Nitrogen setup in the MBE Lab

In this work, a systematic study has been carried out to determine the influence of different MBE growth parameters on the optical and structural properties of the GaAsSbN/GaAs single quantum wells (SQW). Effects of annealing on the PL characteristics under different ambient and annealing temperature were also carried out. Temperature dependence of the PL properties was also a subject of study.

### III. RESEARCH ACTIVITIES AND MAJOR ACCOMPLISHMENTS

The research activities during this period have been broadly classified into the following categories:

#### 1. Comprehensive study of GaAsSbN/GaAs QW Structures grown by MBE

Detailed study on the variation in the optical properties of the as-grown and the effect of annealing on GaAsSbN/GaAs QW heterostructures with growth temperature have been carried out. Incorporation of N and Sb at different growth temperatures has been determined using SIMS.

- In-situ RHEED evaluation of the growth

- Growth temperature optimization and N/Sb incorporation dependence with growth temperature.
- Source shutter opening sequences investigation of interface/layers quality and elements incorporation.
- Annealing effect studies on ex-situ N ambient and in-situ As overpressure ambient, as well as annealing temperatures.

## 2. Annealing Effects on the Temperature Dependence of the Photoluminescence of GaAsSbN SQWs

Detailed study on the effect of annealing on PL has been carried out. PL temperature dependence for SQW has been studied. Recombination mechanism has been investigated. TEM measurement is carried out to verify the analysis.

- Temperature dependence of the PL spectra in as-grown and ex-situ annealed sample.
- Temperature dependence of the PL spectra in in-situ annealed sample.
- The quantitative investigation of the recombination mechanisms.
- TEM z-contrast image verifies the nonradiative recombination due to the composition modulation at the interface for the nitride layers.

## 3. Nitrogen effects on GaAsSbN/GaAs SQWs

Detailed study on the variation in the optical properties of the GaAsSbN /GaAs QW heterostructures with N concentration has been carried out, with all the other growth parameters kept invariant.

- HRXRD scans reveal sharp interfaces for all nitrogen concentrations in range of 0-2.5%.
- PL peak energy exhibits red shift with increase in nitrogen composition in the layer.
- Effect of laser power intensity on the layers with different nitrogen concentrations is investigated.

Major accomplishments in each of above research activities are summarized below

- Comprehensive study of GaAsSbN/GaAs SQW Structures grown by MBE
  - All layers grown at temperature above 430 °C exhibit smooth surface morphology.
  - The substrate temperature in the range of 430-470 °C is found to be optimum.
  - Simultaneously opening source shutters yield sharper layer interfaces and incorporates higher nitrogen.
  - A significant increase in PL intensity (about 10 times) with a narrowing of 10K PL line shape (about 30-45 meV) and blue shift in emission energy (about 50-90 meV) are observed on ex-situ annealing samples. Better

result of 10K PL FWHM (about 20-30 meV) with simultaneously opening the source shutters has been obtained.

- In-situ annealing under As overpressure reveals better results.
- PL emission wavelength as long as 1.52  $\mu\text{m}$  at room temperature has been obtained on annealed sample with GaAlAs barrier layer.

*Initial results of this work were published in MRS Proc. 03 as is included in the report. A rough draft of the manuscript which details the work is also attached herewith and will be submitted to J. Appl. Phys.*

● Annealing Effects on the Temperature Dependence of the Photoluminescence of GaAsSbN SQWs

- Asymmetric PL line shape is observed due to N-induced nonradiative recombination centers
- The temperature dependent PL characteristics of GaAsSbN/GaAs SQWs exhibit an “inverted S-curve” at low temperatures indicative of carrier localization.
- The carrier localization is shallow in as-grown samples and becomes pronounced on annealing in a N ambient, which is attributed to the enhanced defect-induced diffusion at the interface, thereby leading to composition modulation and/or rearrangement of bonding at the interface.
- In-situ annealed samples exhibit less carrier localization with a more symmetric PL line shape, which is attributed to the reduction in the concentration of nonradiative recombination centers.
- The values of the activation energies and the thermal efficiencies of two recombination channels have been determined in the range of 24-32 meV and 7-11 meV respectively.
- SQWs with room temperature PL energy as low as 0.81 eV with FWHM of 54 meV have been achieved in these samples.

*The manuscript is attached herewith and has been submitted to J. Appl. Phys.*

● Effect of nitrogen on GaAsSbN/GaAs SQWs

- HRXRD  $\theta$ - $2\theta$  scan reveals sharp interfaces for the nitrogen concentration range investigated in the range of 0-2.5%.
- PL peak energy decreases at a rate of 270meV/ % change in N for a nitrogen concentration varying upto 0.7% N there after it decreases at a rate of 30meV/% N.
- PL peak energy exhibits an increase in energy at a N concentration of 2.3%.

*The manuscript is under preparation and is attached herewith.*

Detailed description of the above research work is as follows.

## BIBLIOGRAPHY

- [1] F. Bousbih, S. Ben Bouzid, R. Chtourou, F. F. Charfi, J. C. Harmand, G. Ungaro, *Mat. Sci & Engg C* **827** (2002)
- [2] Fischer M, Reinhardt M and Forchel A 2000 *Electron Lett.* **36** 1208.
- [3] W. Ha, V. Gambin, M. Wistey, S. Bank, H. Yuen, S. Kim and J. S. Harris, Jr., *Electronic Letters*, **38 (6)**, pp 277-278, March 2002.
- [4] G. Ungaro, G. Le Roux, R. Teissier and J.C. Harmand, " GaAsSbN: A new low bandgap material for GaAs Substrates", *Electronic. Lett*, 35, 1246 (1999)
- [5] J-C Harmand, A Caliman, E V K Rao, L Largeau, J Ramos, R Teissier, L Travers, G Ungaro, B Theys and I F L Dias, *Semicond. Sci. Technol.* **17**, 778(2002)
- [6] C. Ungaro, G. Leroux, R. Teissier and J.C. Harmand, *Electronics Letters* **35**, July (1999).
- [7] J. C. Harmond, G. Ungaro, L. Largeau, and G. Le Roux, *Applied Physics Letters*, 77, 2482 (2000).

**A. Comprehensive Study of GaAsSbN/GaAs SQW Structures  
grown by MBE**

**MBE Growth Study of GaAsSbN/GaAs Single Quantum Wells**

Liangjin Wu, Shanthi Iyer

Kalyan Nunna, Jia Li, Sudhakar Bharatan, and Ward Collis,

Department of Electrical & Computer Engineering

North Carolina A&T State University Greensboro, NC 27411

Kevin Matney, Bede Scientific Inc, Englewood, CO

**Abstract**

In this work, the growth and properties of GaAsSbN single quantum wells are investigated. The heterostructures were grown on GaAs substrates in an elemental solid source molecular beam epitaxy system with a RF plasma nitrogen source. A systematic study has been carried out to determine the influence of growth temperature on the optical properties of the layers. For reference low temperature photoluminescence (PL) characteristics of the GaAsSb/GaAs QWs as a function of Sb is also presented. A significant increase in PL intensity with a corresponding blue shift in emission energy and a decrease in full width at half maximum (FWHM) has been observed on annealing the GaAsSbN/GaAs sample in a nitrogen ambient at 700°C. PL emission wavelength as long as 1.52  $\mu\text{m}$  at room temperature has been obtained on annealed samples.

**I. Introduction**

The dilute nitride alloy systems lattice matched to GaAs are currently being extensively investigated as an alternative to InP and GaSb based systems for optoelectronic device applications in the laser based radar and optical communication systems. Amongst the different nitrides, namely InGaAsN, GaInAsSbN and GaAsSbN, the InGaAsN material system is the most extensively studied system. In spite of the rapid progress, there are still some serious problems associated with this alloy system, particularly the difficulty in achieving emission at wavelengths larger than 1.5  $\mu\text{m}$  [1, 2]. Recently the InGaAsSbN system has been demonstrated to overcome this problem using the surfactant properties of Sb and lasers with emission wavelengths as long as 1.49  $\mu\text{m}$  have been fabricated [3]. However, the presence of five components makes the growth complex. Hence, the GaAsSbN alloy still remains attractive due to one lower constituent element and further, longer wavelength can be achieved in comparison to the InGaAsN system due to the smaller strain in the corresponding ternary GaAsSb material for similar N composition.

The work on this system has been carried out primarily by Ungaro et al. [4] and Harmand et al. [5, 6]. N incorporation (as much as 2.5%) has been successfully accomplished in this material system by Harmand et al. [5] using molecular beam epitaxial (MBE) technique. Jones et al. [7] have also published similar results on the growth of this alloy system.

This work is our preliminary report on the effect of the growth temperature on the low temperature PL and limited x-ray characteristics on both as grown and annealed samples of GaAsSbN/GaAs quantum wells grown by MBE technique.

## II. Experimental Details

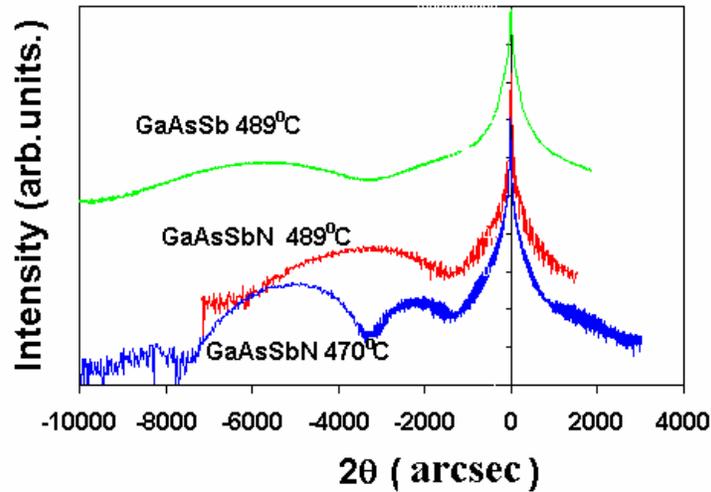
The GaAsSbN QW samples were grown on semi-insulating GaAs (100) substrate in an EPI 930 solid source MBE system using elemental Ga with thermally cracked As and Sb sources. The active N species were generated by flowing ultrahigh pure N<sub>2</sub> through a radio frequency (RF) assisted plasma source. Single quantum wells consisted of GaAsSbN epilayers embedded in GaAs barriers grown at different growth temperatures ranging from 400°C to 490°C. The growth sequence consisted of growth of GaAs buffer layer at 580°C, growth temperature lowered to the desired growth temperature under As overpressure, followed by Sb flushing prior to opening of the Ga and N shutters for GaAsSbN QW growth and terminating with GaAs buffer layer at 580°C. The growth rate of the well layer was estimated to be 1.0 μm/hr as determined from the RHEED intensity oscillations. GaAsSb quantum well layers were also grown at 490°C for different Sb concentrations for reference. For the determination of N composition thick layers of GaAsN were grown at two growth temperatures of 430°C and 490°C for the same elemental flux. The background nitrogen pressure in the growth chamber was about  $2 \times 10^{-7}$  torr for all the nitride samples. The GaAsSbN QW layers were furnace annealed ex-situ at 700°C in the nitrogen ambient for 10 minutes.

In-situ characterization during the growth was carried out utilizing a 15 KV reflection high-energy electron diffraction (RHEED) gun for image acquisition and analyzing the surface reconstruction. Ex-situ characterization consisted of high-resolution x-ray diffraction using a Bede D1 system and low temperature photoluminescence (PL) measurements using a He-Ne laser for a light source, 0.3m grating monochromator and a Ge photodetector. The x-ray experimental data were analyzed using the RADS Mercury software, which employs the dynamical theory to simulate the x-ray intensity diffracted from a layered structure.

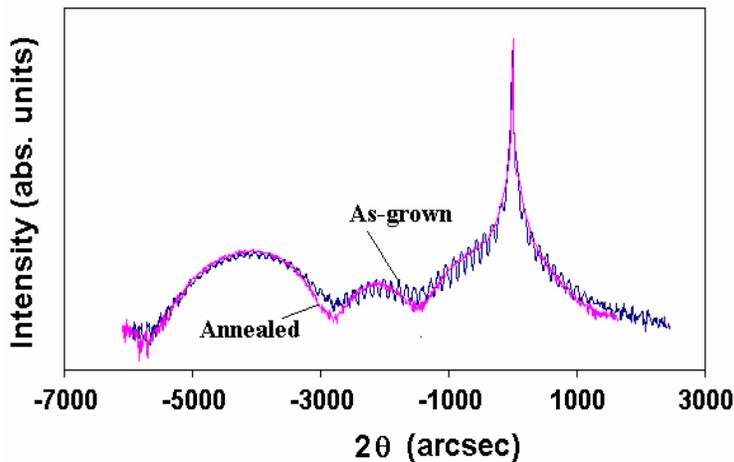
## III. Results and Discussion

For all the GaAsSb and GaAsSbN QW layers the RHEED pattern changed from 2x4 to 1x3 immediately after opening the Sb shutter and the pattern remained streaky throughout the growth for the entire growth temperature range investigated. However, for GaAsN layers grown at 490°C growth, the RHEED pattern became spotty, immediately after opening the N shutter suggesting the 3D growth mode. The spots became dimmer with the increasing thickness, suggesting continued nitridation of the layer makes the layer rough.

Figure 1 shows the HRXRD spectra from (004) of the GaAsSb SQW grown at 490°C and GaAsSbN SQW grown at 490°C and 470°C. The addition of nitrogen to GaAsSb reduces the compressive strain, as expected, in the QW grown at the same growth temperature. A systematic shift of the nitride peak towards longer lattice constant is observed with lower growth temperature. The Sb composition and the thickness of the GaAsSb QW were determined to be  $33.2 \pm 0.005\%$  and  $61.5 \text{ \AA} \pm 0.2 \text{ \AA}$ , respectively, from the best fitting simulation of the experimental x-ray data. The N composition in the GaAsSbN QWs was estimated to be about 1.3-1.4%, from the simulation of the corresponding x-ray data of the GaAsN thick layers grown at the two growth temperatures of 490°C and 430°C. This result is consistent with those of Pan et al. [8] in that the N concentration is almost independent of the growth temperature in the above range in GaAsN.



**Figure 1.** HRXRD spectra of GaAsSb(N) SQW at various substrate temperatures.



**Figure 2.** HRXRD spectra of as-grown and annealed SQW grown at 470°C.

Figure 2 illustrates the HRXRD  $\theta$ - $2\theta$  scan curves for the GaAsSbN/GaAs quantum well of the as-grown and the annealed samples. The presence of Pendellosung fringes and short period fringing from the cap layer for the QW layers (clearly visible in this sample due to a thinner cap layer), are indicative of strained QW layer with sharp and dislocation free interface. On annealing, the spectral peak positions remain invariant with vanishing of the high frequency fringes. The transmission electron microscope micrograph of the corresponding sample (not shown here) indicates an increased interface roughness due to composition modulation at the interface.

Figure 3 shows the systematic shift of 4.5K PL spectra of GaAsSb single QWs from 1.10  $\mu\text{m}$  to 1.21  $\mu\text{m}$  with increasing Sb concentration from 17.1% to 32.8%, respectively. The QW thickness was estimated to be 36Å for the Sb compositions of 17% and 24% and 61Å for Sb composition of 33%, which were obtained from the best fit to the x-ray simulated data. The FWHM of the layers were in the range of 17-21 meV. For double quantum well (DQW) heterostructures with QW thickness of 76Å and barrier layer thickness of 725Å, the PL spectra further shifted to 1.26  $\mu\text{m}$ , which is attributed to

the broadening and shifting of the bound levels to lower energy levels due to the possible interaction between the wells and slightly larger QW thickness.

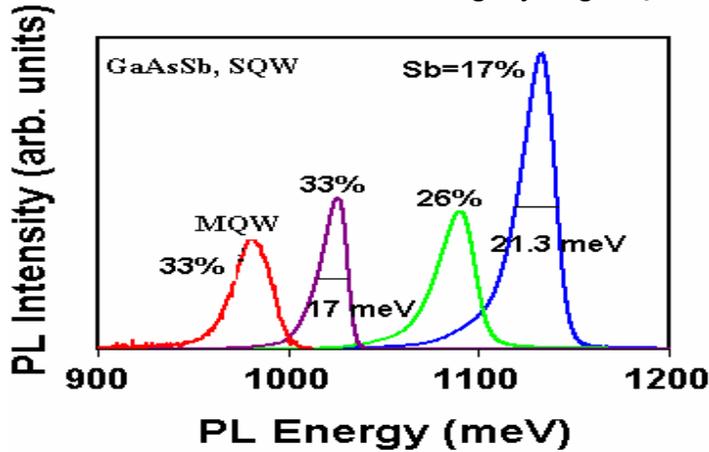


Figure 3. 4.5K PL spectra of GaAsSb SQW & MQW for different Sb compositions.

The addition of N to the SQW of the lowest energy resulted in further lowering of the band gap. Figures 4(a) -4(c) exhibit the variation in the 10K PL energy, intensity and FWHM of as-grown and annealed GaAsSbN /GaAs QW with growth temperature. Also included is the data on the reference GaAsSb QW grown at 490°C and at 430°C. The QW thickness of all these ternary and quaternary layers is estimated to be  $85 \text{ \AA} \pm 5 \text{ \AA}$ . A growth temperature of 450-470°C appears to be the optimum range for the annealed quaternary QWs, exhibiting low PL peak energy with increased intensity and low FWHM. The shift in the PL peak energy in the as grown sample is about 250 meV, which roughly corresponds to N concentration of 1.4% using Ungaro’s [4] value for the shift.

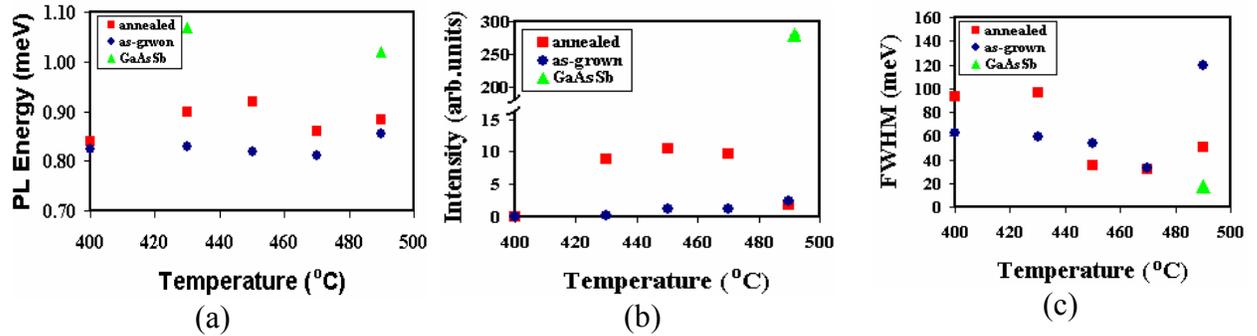


Figure 4. Low temperature PL characterization of energy, intensity and FWHM of GaAsSbN QW at different substrate temperatures.

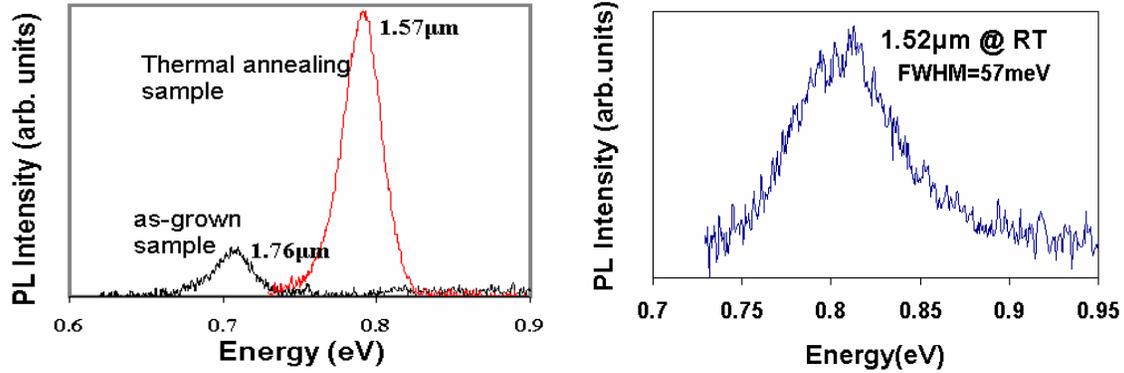


Figure 5. (a) 10 K PL measurement for as-grown and annealed GaAsSbN SQW.  
 (b) RT PL measurement for annealed GaAsSbN SQW with GaAlAs barriers.

Figure 5a shows the typical 10K PL characteristics of the SQW grown at 470°C and nitrogen annealed samples. The longest wavelength that has been achieved on the as grown samples is 1.76 μm, which on annealing shifted to 1.57 μm as measured at 10K. Room temperature PL is observed (Fig. 5b) when GaAlAs is used as the barrier layer, due to the better carrier confinement.

#### IV. Conclusion

Growth temperatures in the range of 450-470°C have been found to be the optimum for the growth of GaAsSbN/GaAs SQWs. The composition of N and Sb is estimated to be around 33% and 1.4%, respectively. Presence of the Pendellosung fringes and high frequency fringes due to the cap layer attest to the good quality of the layers grown. Room temperature PL as long as 1.52 μm has been obtained on these samples.

#### Acknowledgements

This work is supported by Army Research Office (Grant No. DAAD19-02-004), NASA (Grant No. NAG3-2782), and DOD (Contract No. DAAD19-03-2-0016).

#### References:

- [1] H. Saito, T. Makimoto and N. Kobayashi, *J.Cryst.Growth* **195**, 416(1998).
- [2] D.J. Friedman, J.F.Geisz, S.R. Kurtz and J.M. Olson, *J.Cryst.Growth* **195**, 438(1998).
- [3] W. Ha, V. Gambin, M. Wistey, S. Bank, H. Yuen, S. Kim and J. S. Harris, Jr., *Electronic Letters* **38**, 277, (2002).
- [4] G. Ungaro, G.Le Roux, R.Teissier and J.C.Harmand, *Electronics Letters* **35**, 1246 (1999).

- [5] J. C. Harmand, A. Caliman, E. V. K. Rao, L. Largeau, J.Ramos, R. Tessier, L. Travers, G. Ungaro, B. Theys, and I.F.L. Dias, *Semicond. Sci. Technol.* **17**, 778-784 (2002).
- [6] J.C.Harmand, G.Ungaro, J.Ramos, E.V.K.Rao, G.Saint-Girons, R.Teissier, G.Le Roux, L.Largeau, and G.Patriarche, *J.Cryst. Growth* **227-228**, 553 (2001).
- [7] E.D.Jones, N.A.Modine, K.E.Waldrip, F.Jalali, J.F.Klem, and G.M.Peake, *MRS Proc.* **744**, 637 (2002).
- [8] Z.Pan, L.H.Li, W.Zhang, Y.W.Lin, and R.H.Wu, *Appl. Phys. Lett.*, **77(2)**, 214(2000)

**MBE Growth and Properties of GaAsSbN/GaAs Single Quantum Wells**

**L. Wu, S. Iyer, K. Nunna, J. Li, S. Bharatan, W. Collis**

**Department of Electrical and Computer Engineering,  
North Carolina A&T State University, Greensboro, NC 27411**

**K. Matney**

**Bede Scientific Inc, Englewood, CO 80112**

**Abstract**

GaAsSbN single quantum wells (SQWs) growth and properties are investigated in this work. The heterostructures were grown on GaAs substrates in an elemental solid source molecular beam epitaxy (MBE) system assisted with a RF plasma nitrogen source. A systematic study has been carried out to determine the influence of various growth conditions, such as the growth temperature, source shutter opening sequence, and subsequent annealing of these layers ex-situ and in-situ under As overpressure, on the optical properties of the layers. N and Sb incorporations were found to depend strongly on substrate temperature and source shutter opening sequence. The substrate temperature in the range of 450-470 °C was found to be optimum. Simultaneous opening of the source shutters was found to yield sharper QW interfaces. A significant increase in PL intensity with a narrowing of PL line shape and blue shift in emission energy were observed on annealing the GaAsSbN/GaAs SQW in nitrogen ambient. In-situ annealing under As overpressure reveal better results.

PACS: 71.55 Eq, 73.21 Fg, 78.55.Cr, 78.67.De

## I. Introduction

The dilute nitride alloy systems lattice matched to GaAs are currently being extensively investigated as an alternative to InP and GaSb based systems for optoelectronic device applications in the laser based radar and optical communication systems. The addition of small amounts of N offers a unique feature of reducing simultaneously both the energy band gap and the lattice parameters of a given III-V compound or an alloy. The GaInNAs QWs has reduced the bandgap to 1.5  $\mu\text{m}$  and has been developed promised application. Laser diodes based on this material system operating in 1.3  $\mu\text{m}$  to 1.5  $\mu\text{m}$  range have been fabricated and their performance characteristics have been studied [1-3]. In addition, applications of III-V dilute nitrides in solar cells [4] and heterojunction bipolar transistors have been investigated [5].

As an alternative, GaAsSbN has been found to have some advantage. It has been shown [6] that for a given value of Sb composition the value of the band gap in GaAsSb/GaAs quantum well (QW) structures are smaller than in InGaAs/GaAs QW structures for the same value of In composition as long as the widths of the QW structures are smaller than the critical thickness. This opens the possibility of obtaining lower values of the band gaps in GaAsSb/GaAs quantum wells than in InGaAs/GaAs QWs for the same values of the nitrogen composition. Ungaro et al [7] demonstrated that the incorporation of small amount of N (1%) in GaAsSb ternary alloy also reduces its band gap by almost 180 meV as in InGaAs. Much less is known about the structural, electronic and optical values of GaAsSbN epilayer and GaAsSbN/GaAs QWs than in InGaAsN/GaAs QWs. More N mole fraction incorporated will reduce the bandgap further but degrade the layer properties. It has been speculated [6] that GaAsSbN QWs can successfully achieve 1.55  $\mu\text{m}$  with about 36% Sb if remaining N composition about 1%. 17%-25% Sb has been incorporated in most of the MBE grown GaAsSbN structures [6-8]. The Sb sticking coefficient decreases when the growth temperature increase in InAsSb alloys [9]. Kaspi et al [10] found that the shutter opening sequence and substrate temperature play significant roles in Sb-surface segregation and the control of composition at GaAsSb/GaAs interface. Growth parameters such as temperature, growth rate, and thickness, cause variation in nitridation layer which can be sensitive to photoluminescence (PL), full width at half maximum (FWHM) and high resolution x-ray diffraction (HRXRD). In this paper, we have carried out a systematic study on the effect of the substrate temperature, shutter opening sequence, and thermal annealing on the reflection high-energy electron diffraction (RHEED), photoluminescence (PL), and high resolution x-ray diffraction (HRXRD) characteristics of GaAsSbN/GaAs quantum well structures.

## II. Experimental Details

GaAsSbN/GaAs single quantum well structures were grown on undoped (100) GaAs substrates by the molecular beam epitaxial [MBE] technique in an EPI 930 system equipped with elemental Ga, As, and Sb cracker sources and with an EPI plasma RF Unibulb source for N. The SQW structures consisted of GaAsSb(N) grown in the temperature range of 490-400  $^{\circ}\text{C}$ , sandwiched between a GaAs buffer layer of 450 nm thickness and cap layer of 300 nm grown at 580  $^{\circ}\text{C}$ . Two different source shutter opening sequences were used for the growth of the QW. The QWs grown by flushing with Sb for 30-60 seconds prior to the growth will be referred to as a', while QWs grown with a

simultaneous opening of all source shutters will be referred to as b'. Most of the QWs were grown by using procedure a', unless stated otherwise. The thickness of the QWs as estimated from HRXRD was close to 8-9 nm. These were subjected to post growth furnace annealing with a GaAs substrate cover on top, in N ambient at 700 °C-800 °C for 10 minutes. In-situ annealing was also carried in As overpressure at 650 °C for 10 minutes in the growth chamber for a few GaAsSbN/GaAs SQW. For comparison, a GaAsSb/GaAs SQW and GaNAs bulk was also grown at 490 °C and 430 °C.

On some of the samples, Sb and N compositions were determined from a combination of HRXRD and secondary ion mass spectroscopy (SIMS) analysis. The error bars in the N and Sb composition were  $\pm 0.1\%$  and  $\pm 1.5\%$ , respectively.

In situ RHEED was used to monitor the surface reconstruction for layers. HRXRD measurements were performed on a Bede Scientific Metrix-F automated diffractometer, equipped with a Microsource X-ray generator. The motorized detector slit was set to 0.5 mm wide, giving a  $2\theta$  angular resolution of 150 arcsconds.

PL measurements were carried out using He-Ne laser as the light source for excitation, a 0.32 m double grating monochromator for wavelength dispersion with a thermoelectrically cooled Ge detector for the signal detection using a conventional lock-in technique. A closed cycle three stage APD cryogenic system was used to perform the temperature dependence study of the QWs.

### III. Results and Discussion

#### (1) RHEED Study

It is found that for the entire the substrate temperature range of 400 °C - 490 °C, the RHEED pattern exhibited streaks (not shown here) during the GaAsSbN and GaAsSb QW growths, indicating the two dimensional growth mode. However, the RHEED pattern was found to be spotty for GaAsN QW growth under the same growth conditions, indicating a three-dimensional growth mode. In contrast, the onset temperature of three-dimensional islands and formation of clusters in MBE grown GaInNAs and InGaAs layers was found to be 460 °C [11]. This suggests that Sb plays a role of the surfactant as well as the constituent when the Sb flux is about  $2e-7$  torr in our samples. This is in agreement with studies of Sb incorporation in GaNAs [12-14] and GaInNAs [15].

Figures 1 (a) and (b) exhibit the variation in the RHEED intensity and FWHM during the QW growth for different substrate temperatures and shutter sequences. For sample grown by procedure a' at 490 °C, the intensity almost remains invariant with an increase in FWHM when flushing with Sb for 60 seconds prior to QW layer growth, indicating the roughening of the surface. The intensity and FWHM further increase on opening the Ga and N source shutters. The behavior of RHEED intensity for sample grown at 470 °C is similar to the samples grown at 490 °C during the QW growth, while FWHM increases on opening the Sb shutter with a gradual decrease on the opening the G and N shutters. This suggests the formation of discrete island that occurs initially gradually coalesces leading to surface smoothening. For samples grown at 400 °C, the intensity decreases with increase in FWHM, which is indicative of a very rough surface.

For samples grown by procedure b' at 490 °C, the RHEED pattern exhibits somewhat similar trend as in samples grown at 470 °C by procedure a'. The sample grown at 430 °C is as rough as evidenced from the constant intensity and increased FWHM.

The layers grown by both of the different shutter-opening sequences exhibit smooth surface morphology as observed in Normaski microscope for all growth temperatures greater than 430 °C.

## (2) Growth Temperature

Figure 2 shows the (004) HRXRD spectra of the GaAsSbN SQW grown in the temperature range of 400-489 °C. For reference (004) HRXRD spectra of GaAsSb SQWs grown at 489 °C and 430 °C are also included. The addition of nitrogen to GaAsSb reduces the compressive strain in the QW grown at the same growth temperature. The presence of Pendellosung fringes superimposed with high frequency fringes for nitride layers grown at 430 °C and above is indicative of strained QW with sharp and dislocation free interfaces. The absence of these fringes and a broad nitride peak observed on QWs grown at 400 °C is indicative of the poor quality of layers and thus sets the lower limit for the growth temperature of these nitrides.

It is obvious that the compressive strain is reduced with growth temperature increase from 430 °C to 490 °C for both GaAsSb and GaAsSbN systems. In the case of GaAsSb SQW, this has been found to be caused by the reduction in Sb concentration from 33.2% to 29.6% with the increased growth temperature in this region, while in the case of nitride QWs both the reduction in Sb and increased N incorporations (see Figure 3) cause this effect

## (3) Source Shutter Opening Sequences

Figure 4 illustrates the comparison of HRXRD spectra from (004) GaAsSb and GaAsSbN SQWs grown at 490 °C and 430 °C with the two shutter sequences identified earlier as a' and as b'. The layers grown by either of the above procedures exhibit Pendellosung fringes superimposed with high frequency fringes, though the fringes are much more deep for QWs grown by procedure b'. Further, the substrate peak is much sharper for both the ternary and quaternary QWs grown by procedure b'. For layers grown at 490 °C (see Fig.4a), Sb incorporation is larger when grown by procedure a' (29.7% as opposed to 23.7%). On the introduction of N the nitride peak positions for these two SQWs are about 600 arc-seconds apart as shown in Fig.4 (b). SIMS analysis on the sample grown by procedure a' indicated less Sb incorporation of 21.3% with N composition of 1.25%. Assuming the Sb composition to be the same for the nitride layer grown using the procedure b', the N composition in the layer is estimated to be higher around 2.23%. For layers grown at 430 °C, the Sb composition in the ternary SQWs (Fig.4c) is high (approx. 33%), though it remains invariant with the growth procedure used. However, a significant peak shift from 6200 arc-sec to 5000 arc-sec is observable (Fig. 4d) for nitride peaks grown corresponding to N incorporation of 2% grown by procedure b' as compared to 0.5% grown by procedure a'. The latter was confirmed again by SIMS analysis.

The 10 K PL spectra for these GaAsSbN SQW structures are shown in Fig.5. GaAsSbN peaks are much sharper with FWHM ranging from 20-30 meV for layers grown by procedure b'. The peak energy is red shifted by almost 60-80 meV with respect to the layers grown by procedure a', attesting to the higher N concentration in these layers, consistent with the x-ray simulation. Further, the PL intensity is 2-4 times higher for these samples and the spectral shape is also symmetric for these layers. This is

also consistent with the small FWHM values observed for RHEED lines during the growth of these layers.

The above data clearly indicate that the Sb and N incorporations in the layer are not only a strong function of the growth temperature but also the shutter sequences adopted. It is speculated that the Sb flushing prior to growth in procedure a' allows strong bonding to be formed between As and Sb which aids in enhanced incorporation of Sb in the layer, and contributes to significant change in the N incorporation with growth temperature due to the strong temperature dependence of the sticking coefficient of Sb. Further Sb flushing roughens the growth surface and results in relatively broad PL and x-ray peaks. On opening all the shutters simultaneously N is able to knock off Sb due to the small bonding energy of GaSb and modulation in N composition with growth temperature almost vanishes.

In order to obtain the interdependence of N and Sb in the GaAsSbN SQW, thick GaAsN layers were grown at 430 °C and 490 °C using the same flux as in the ternary and quaternary. HRXRD simulation of the experimental data yielded N concentration to be 1.3% to 1.4% in this growth temperature range. This result is consistent with those of Pan et al. [11] in that the N concentration is almost independent of the growth temperature in GaAsN. However, for the same N flux used, N incorporation is less in GaAsN than GaAsSbN. This result is consistent with the reports in literature [6, 15] where N incorporation is found to increase in the presence of Sb.

#### **(4) Annealing Effects**

Figures 6(a)-(c) exhibit the variations of the PL peak energy, intensity and FWHM measured at 10 K for as-grown and ex-situ annealed GaAsSbN/GaAs QWs with growth temperature. The data on the GaAsSb QWs grown at 490 °C and at 430 °C are also included for reference. On annealing, the PL peak energy exhibits a red shift by 50 to 90 meV with increase in intensity by roughly 10 times and has a weak dependence on the growth temperature. However, there is a considerable degradation in the quality of the layers grown at temperatures at and below 430 °C as evidenced by the increased FWHM of the PL spectra and decreased PL intensity on the annealed samples. This may be attributed largely to the increased roughening of the growth surface at lower growth temperatures due to the increased sticking coefficient of Sb, in conjunction with Sb flushing prior to the QW growth. Thus growth temperature of 450-470°C appears to be the optimum range for the QW growth, where the low PL peak energy with increased intensity and low FWHM were achieved on annealed samples.

The effect of annealing temperature on the PL characteristics of SQW samples with both GaAs and GaAlAs barrier are shown in Figs.8 (a)-(c). PL characteristics of in-situ annealed samples at 650 °C and 700 °C is also included. The higher annealing temperature leads to higher blue shift in PL energy with increased intensity and marginal changes in FWHM. In-situ annealing provides the best result in terms of increased intensity and lower FWHM. Samples with GaAlAs barriers show the better emission efficiency compared to the GaAs ones, due to the better carrier confinement. The optimum annealing temperature for ex-situ annealing is around 750 °C.

Figure 9 illustrates the HRXRD  $\theta$ -2 $\theta$  scan curves for the as-grown and the ex-situ annealed SQW structure grown at 470 °C for 13 nm thick with a cap layer of 100 nm. On annealing, the spectral peak positions remain invariant with vanishing of the high

frequency fringes. This, rules out any significant outdiffusion of N and Sb that would alter the composition of the QW. In addition, TEM micrographs taken on this sample [16] do not indicate any extended defects or clusters; however it exhibits some roughness in the order of 3-4 monolayers on either side of the QW. Hence, the changes observed on annealing can be attributed to the reduction of the N-induced nonradiative recombination centers and increased interface roughness with increasing annealing temperature. The latter would affect in the reduction of the effective thickness of the QW. However, both the above effects would result in increased blue shift as observed, and will have an opposing effect on intensity and FWHM. This explains the insensitivity of FWHM to annealing temperature.

However, the PL intensity and peak energy of the in-situ annealed samples are always found to be somewhat higher than the ex-situ annealed samples. The samples were also annealed ex-situ in a roughing vacuum at 700 °C and increased intensity with somewhat higher blue shifts was observed on these samples. Further, temperature dependence of PL energy indicates increased localization in ex-situ annealed samples [16]. This observation suggests that N induced recombination centers at the band edges were more efficiently eliminated in vacuum annealing under As overpressure.

The PL peak energy is 0.815 eV for 8 nm layer and 0.790 eV for 13 nm layer, indicating the transition energy decrease when increasing the quantum well width as expected.

#### **IV. Conclusion**

The incorporations of Sb and N were found to depend strongly on the growth temperature and source shutter opening sequence. Growth temperatures in the range of 450-470°C have been found to be the optimum for achieving good PL and structural characteristics. HRXRD spectra of all the layers grown exhibited the presence of the Pendullosung fringes and high frequency fringes due to the cap layer, attesting to the good quality of the layers grown. PL characteristics were found to improve with annealing. The QW layers grown with simultaneous opening of source shutters indicated relatively narrow nitride peak in the HRXRD spectra. Low PL FWHM in the range of 20-30 meV at 4K PL peak energy of 0.82-0.86eV further attest to the excellent quality of the layers grown. The optimal temperature for ex-situ annealing was found to be 750 °C. PL intensity was found to improve further with in-situ annealing at 650 °C.

#### **Acknowledgements**

This work is supported by Army Research Office (Grant No. DAAD19-02-004, Contract No. W911NF-041-0025), NASA (Grant No. NAG3-2782), and DOD (Contract No. DAAD19-03-2-0016). SIMS measurements were carried out by Evans East.

## V. References

1. M. Reinhardt, M. Fischer, M. Kamp and A. Forchel, *Electron. Lett.* **36**, 1025 (2000).
2. D. A. Livshits, A. Y. Egorov and H. Riechert, *Electron. Lett.* **36**, 1381 (2000).
3. J. S. Harris Jr., *Semicond. Sci. Technol.* **17**, 880 (2002).
4. S. R. Kurtz, J. F. Clem, A. A. Allerman, R. M. Sieg, C. H. Seager and E. D. Jones, *Appl. Phys. Lett.* **80**, 1379 (2002).
5. P. M. Asbeck, R. J. Welty, C. W. Tu, H. P. Xin and R. E. Welser, *Semicond. Sci. Technol.* **17**, 898 (2002).
6. J. C. Harmand, A. Caliman, E. V. K. Rao, L. Largeau, J. Ramos, R. Teissier, L. Traverse, G. Ungaro, B. Theys and I. F. L. Dias, *Semicond. Sci. Technol.* **17**, 778 (2002).
7. G. Ungaro, G. Le Roux, R. Teissier and J. C. Harmand, *Electron. Lett.* **35**, 1246 (1999).
8. S. A. Lourenco, I. F. L. Dias, L. C. Pocas, J. L. Duarte, J. B. B.de Oliveira, and J. C. Hamand, *Journal of Applied Physics*, **93**, 4475 (2003).
9. H. Miyoshi, R. Suzuki, H. Amano, and Y. Horikoshi, *Journal of Crystal Growth* **237-239**, 1519(2002).
10. R. Kaspi, K. R. Evans, *Journal of Crystal Growth* **175/176**, 838(1997).
11. Z. Pan, L. H. Li, W. Zhang, Y. W. Lin and R. H. Wu, *Applied Physics Letters* **77**, 214(2000).
12. W. K. Cheah, W. J. Fan, S. Wicaksono, S. F. Yoon, and K. H. Tan, *Journal of Crystal Growth* **254**, 305(2003).
13. X. Yang, M. J. Jurkovic, J. B. Heroux, and W. I. Wang, *Applied Physics Letters* **75**, 178(1999).
14. X. Yang, J. B. Heroux, L. F. Mei, and W. I. Wang, *Applied Physics Letters* **78**, 4068, (2001).
15. K. Volz, V. Gambin, W. Ha, M. A. Wistey, H. Yuen, S. Bank, and J. S. Harris, *Journal of Crystal Growth* **251**, 360(2003).
16. J. Li, S.Iyer, S.Bharatan, L.Wu, K.Nunna, W.Collis, K.K.Bajaj, K.Matney, and G.Duscher, *Journal of Applied Physics*, submitted.

## VI. Figure Captions

Fig.1. RHEED analysis of (a) intensity and (b) FWHM variation with time for different substrate temperatures and shutter sequences.

Fig.2. HRXRD  $\theta$ - $2\theta$  scans spectra from (004) of GaAsSb and GaAsSbN SQWs grown at various substrate temperatures.

Fig.3. N and Sb composition in GaAsSbN SQWs grown at various substrate temperatures.

Fig.4. HRXRD  $\theta$ - $2\theta$  scans spectra from (004) of GaAsSb and GaAsSbN SQWs grown at 490 °C (a-b) and 430 °C (c-d) by procedure **a'**—flushing Sb prior to growth, **b'**—open Ga, Sb, N simultaneously.

Fig.5. 10 K PL spectra of GaAsSbN SQWs grown at (a) 490 °C and (b) 430 °C by procedure **a'**—flushing Sb prior to growth, **b'**—open Ga, Sb, N simultaneously.

Fig.6. 10 K PL characterization of (a) peak energy, (b) intensity, and (c) FWHM for GaAsSbN SQWs at various substrate temperatures. Symbols open circle represents the as-grown sample, solid circle for annealed samples, and open triangle for GaAsSb as-grown sample.

Fig.7. 10 K PL characterization of (a) peak energy, (b) intensity, and (c) FWHM for GaAsSbN SQWs grown at 470 °C with different N ambient annealing temperatures (solid star). Included are the data of samples annealed in As ambient (open star) and sample with GaAlAs barrier (solid circle).

Fig.8. HRXRD  $\theta$ - $2\theta$  scans spectra from (004) of a GaAsSbN QW grown at 470 °C; 13 nm thick with a cap layer of 100 nm.

Figure 1. Liangjin Wu et al

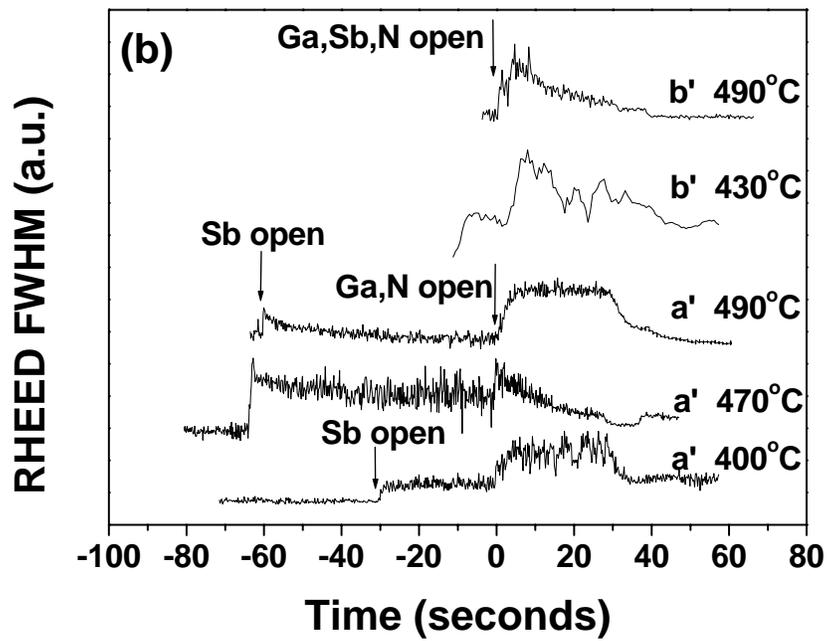
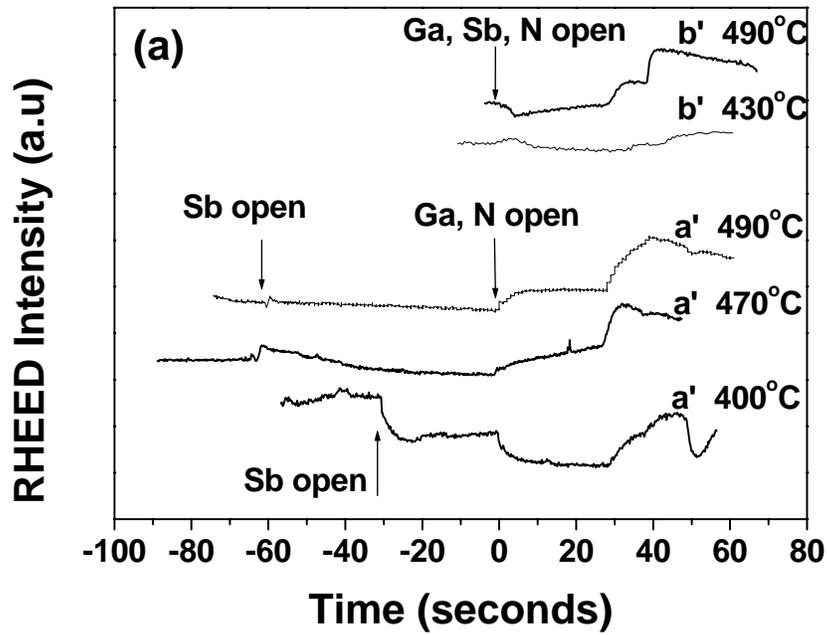


Figure 2. Liangjin Wu et al

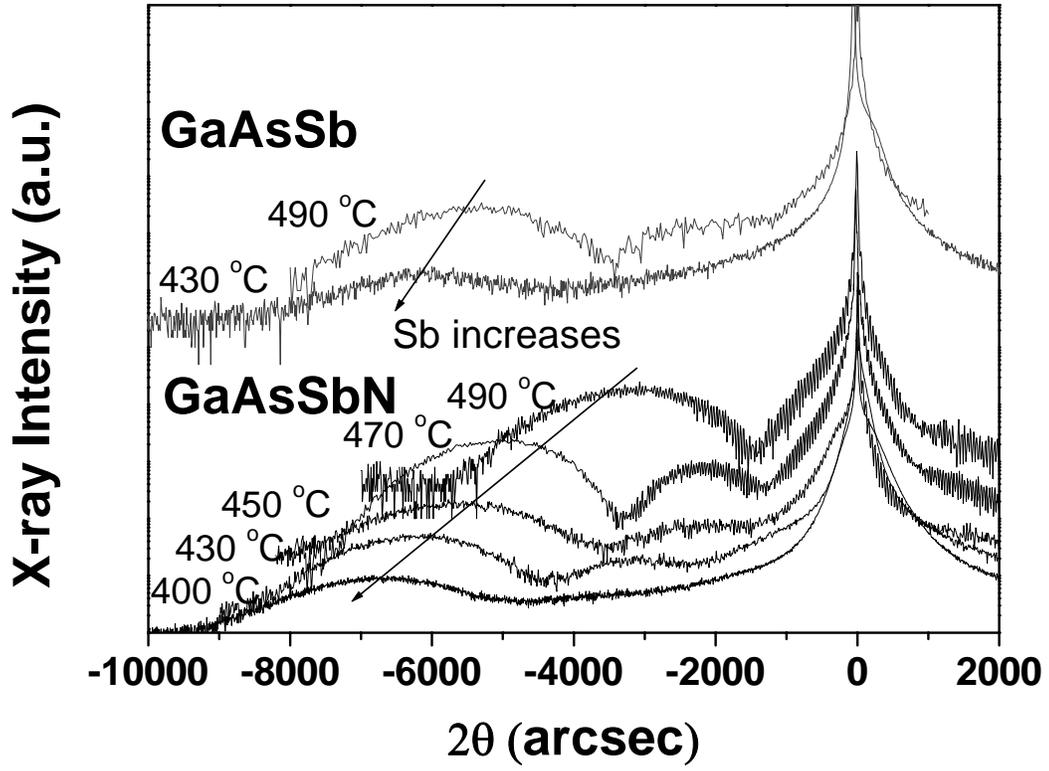


Figure 3. Liangjin Wu et al

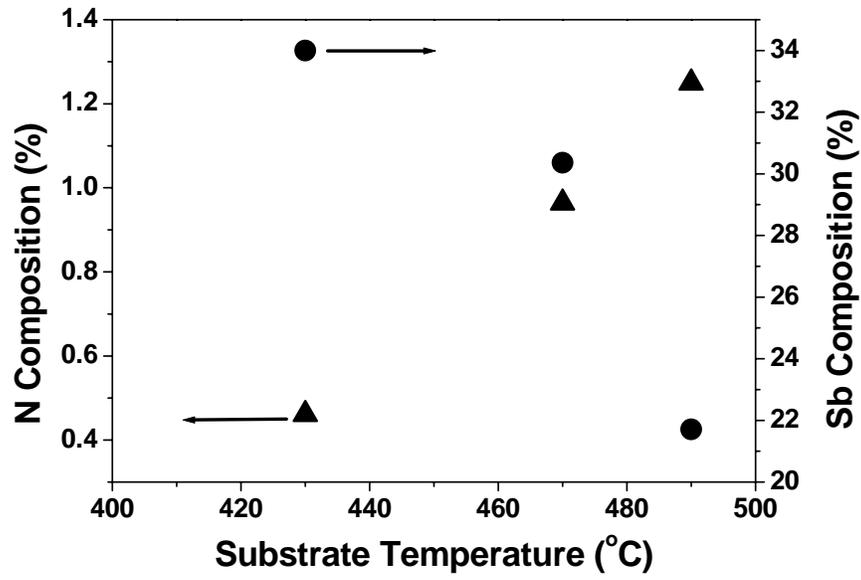


Figure 4. Liangjin Wu et al

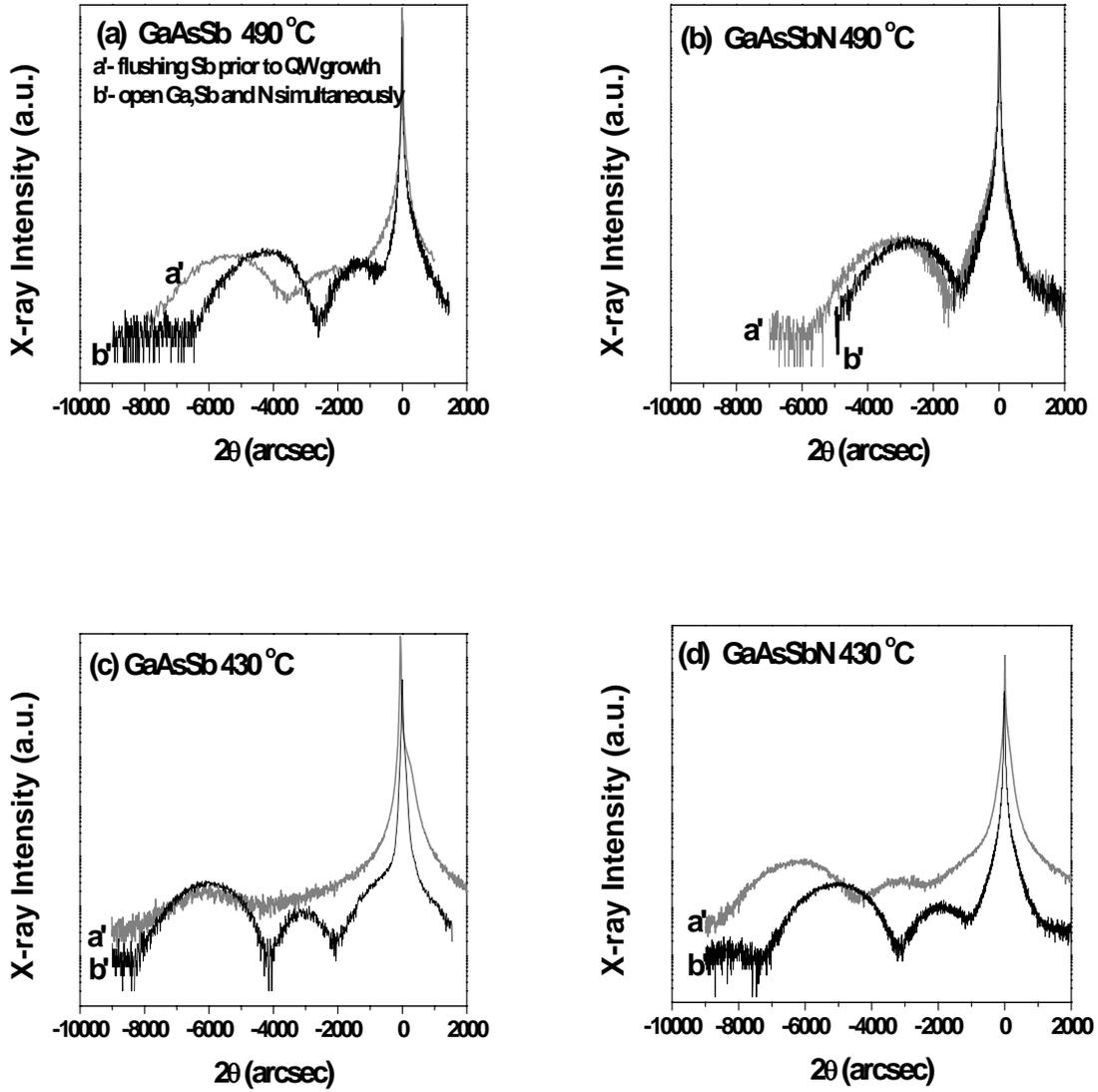


Figure 5. Wu et al

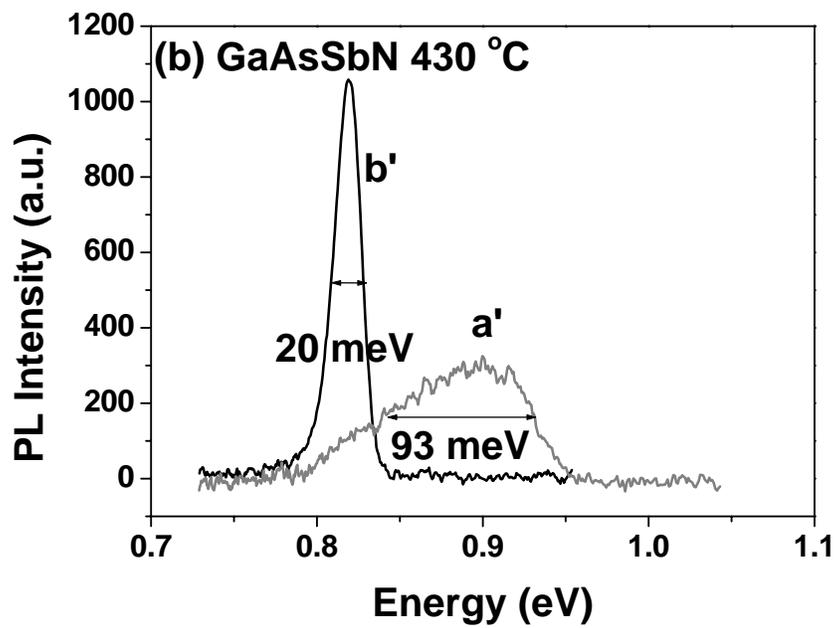
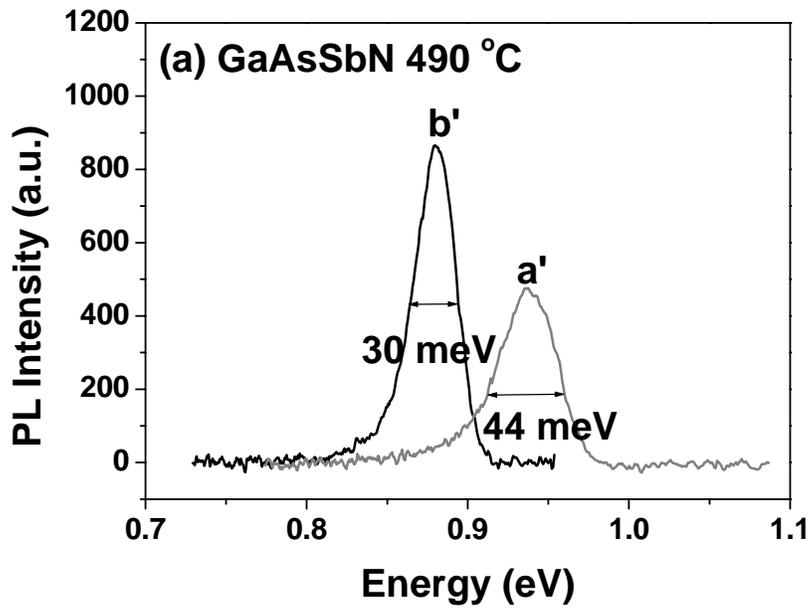


Figure 6. Liangjin Wu et al

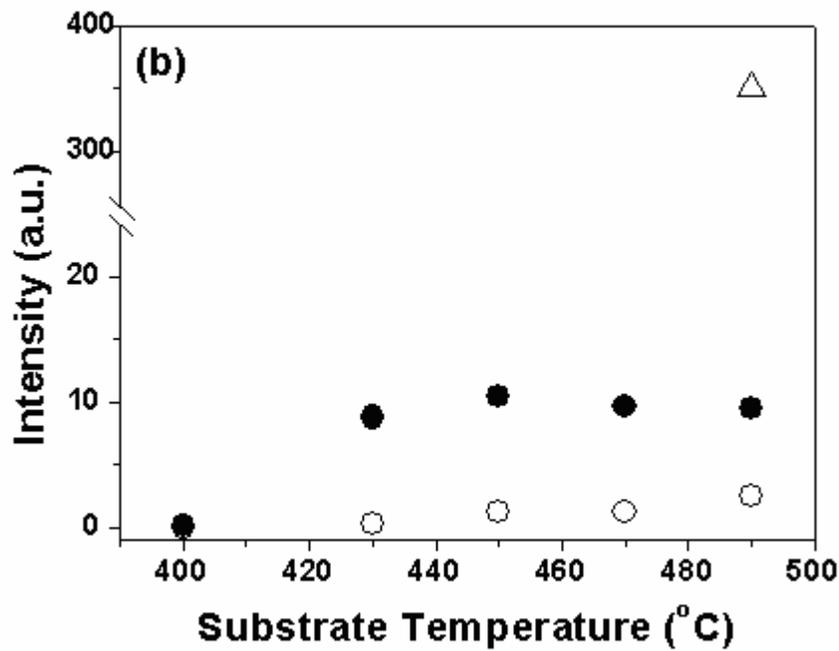
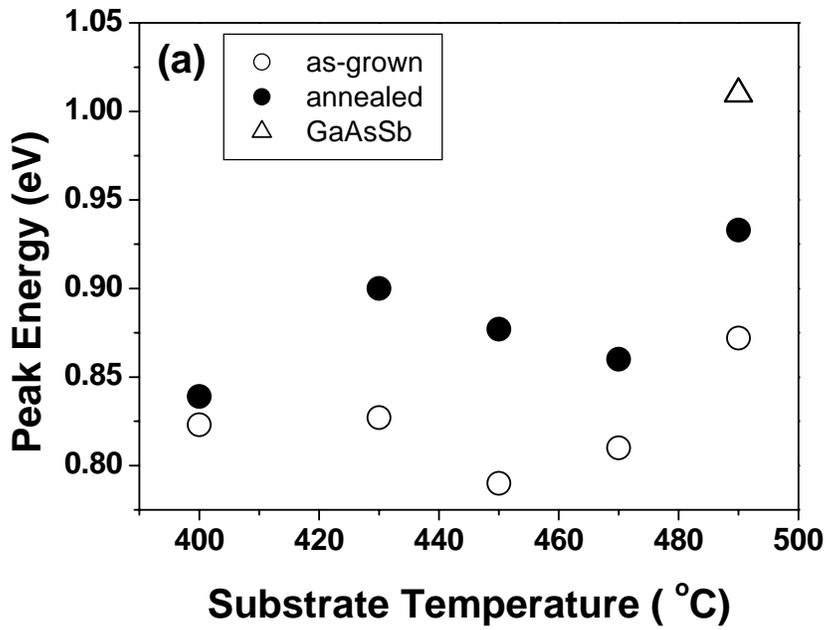


Figure 6. Liangjin Wu et al

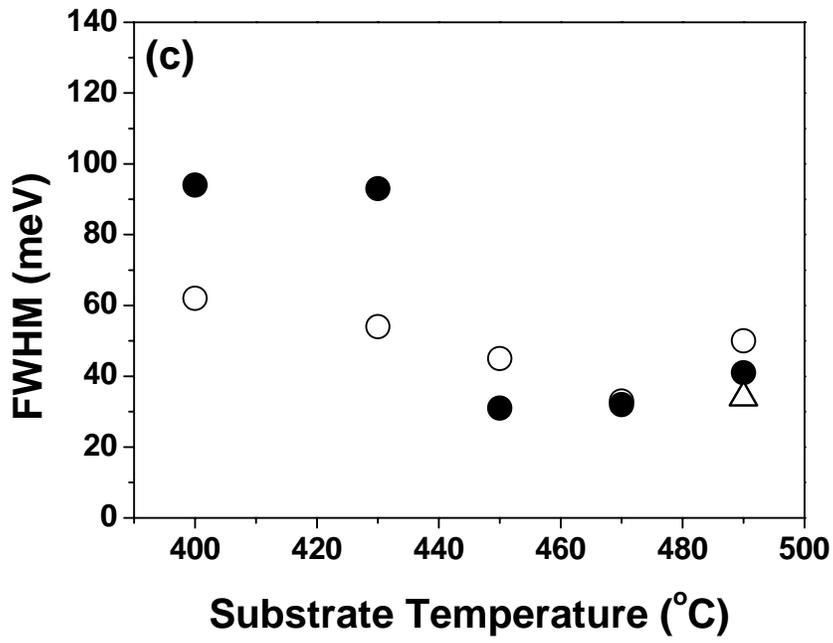


Figure 7. Liangjin Wu et al

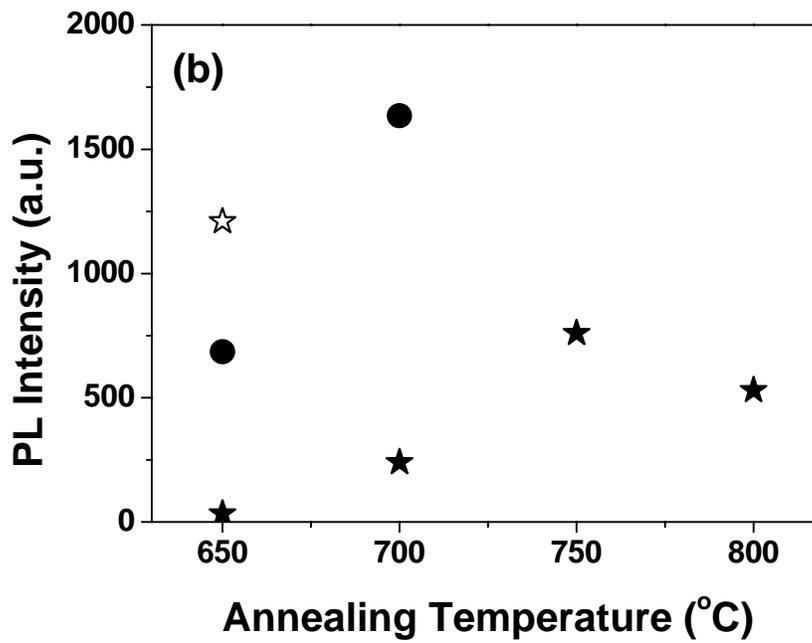
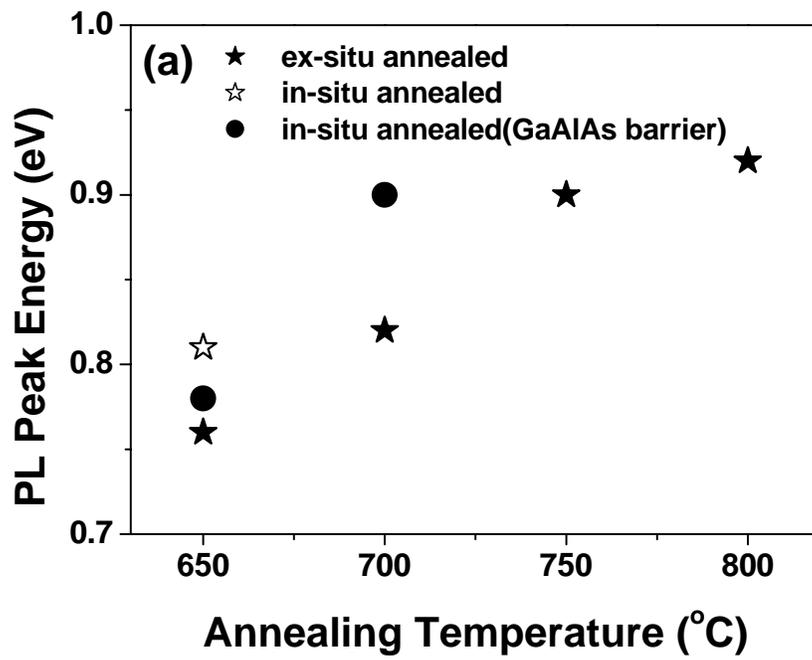


Figure 7. Liangjin Wu et al

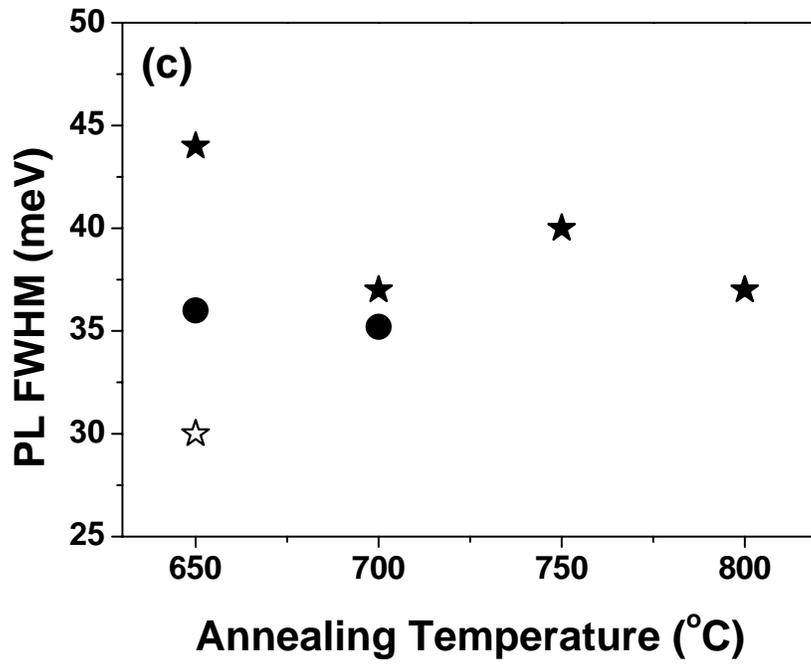
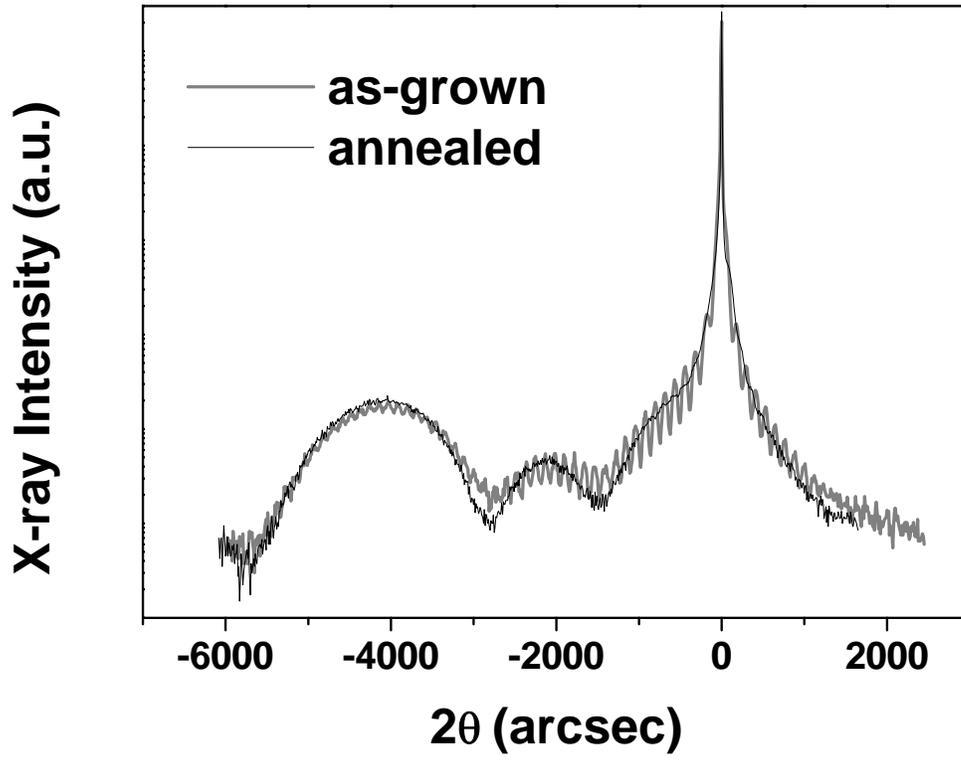


Figure 8. Liangjin Wu et al



**B. Annealing Effects on the Temperature Dependence of  
the Photoluminescence of GaAsSbN SQWs**

*Manuscript submitted to Journal of Applied Physics*

## **Annealing Effects on the Temperature Dependence of the Photoluminescence of GaAsSbN Single Quantum Wells**

**J. Li, S. Iyer, S. Bharatan, L. Wu, K. Nunna, W. Collis**  
Department of Electrical and Computer Engineering,  
North Carolina A&T State University, Greensboro, NC 27411

**K. K. Bajaj**  
Department of Physics, Emory University, Atlanta, GA 30322

**K. Matney**  
Bede Scientific Inc, Englewood, CO 80112

**G. Duscher**  
North Carolina State University, Raleigh, NC 27695

### **Abstract**

In this work the effects of ex-situ annealing in a N ambient and in-situ in an As ambient on the temperature dependence of the photoluminescence (PL) spectral characteristics of GaAsSbN/GaAs single quantum well (SQW) heterostructures have been investigated. The focus of this work is on three representative nitride samples grown by molecular beam epitaxy (MBE) in the temperature range of 450-490 °C. Excitonic transitions in the QWs exhibit the well known “inverted S-curve” behavior in the low temperature regime, the curve being more pronounced in samples annealed in a N ambient. The samples annealed in-situ under an As overpressure exhibit less carrier localization with a more symmetric PL line shape, which is attributed to the reduction in the concentration of nonradiative recombination centers. A room temperature PL peak energy as low as 0.81 eV with a FWHM of 54 meV has been achieved in some of these samples. The values of the activation energies and the thermal efficiencies of the two recombination channels contributing to the nonradiative recombination in these samples have been determined from the temperature dependence of the PL intensity.

PACS: 71.55 Eq, 73.21 Fg, 78.55.Cr, 78.67.De

## I. Introduction

During the last decade a considerable amount of effort has been devoted to the study of the growth and structural, electronic and optical properties of III-V compounds and their alloys containing a small percentage of N. The early observation of Weyers et al. [1] and Kondow et al. [2] of the dramatic reduction of (about 180meV) in the energy band gap of GaAs for instance, with the introduction of small concentration of N (~1%) has stimulated an enormous interest in the opto-electronic devices grown on commonly available GaAs substrates, operating in the 1.3  $\mu\text{m}$  region. Introduction of nitrogen in GaAs also leads to smaller values of the lattice parameter. Thus the addition of small amounts of N also offers a unique feature of reducing simultaneously both the energy band gap and the lattice parameters of a given III-V compound or an alloy.

To reduce the energy band gap of the GaAs-based materials further, an InGaAsN quaternary was proposed by Kondow et al [3]. This allowed the energy band gap of the alloy system to be reduced to the 1.5  $\mu\text{m}$  range. Laser diodes based on this coherently strained system operating in the 1.3 to 1.5  $\mu\text{m}$  range have been fabricated and their performance characteristics have been studied [4-6]. In addition, applications of III-V dilute nitrides in solar cells [7] and heterojunction bipolar transistors have been investigated [8].

Some time ago Ungaro et al. [9] demonstrated that the incorporation of small amount of N (1%) in a GaAsSb ternary alloy also reduces its energy band gap by almost 180meV, almost the same as in GaAs. It has been shown [10] that for a given value of Sb composition the value of the energy band gap in GaAsSb/GaAs quantum well structures is smaller than that of InGaAs/GaAs quantum well structures for the same value of In composition as long as the widths of the quantum well structures are smaller than the critical thickness. This opens the possibility of obtaining lower values of the band gaps in GaAsSb/GaAs quantum wells than in InGaAs/GaAs quantum wells for the same values of the nitrogen composition.

Much less is known about the structural, electronic and optical properties of GaAsSbN epilayers and GaAsSbN/GaAs quantum wells than in InGaAsN/GaAs quantum wells. The PL properties of these systems are strongly influenced by the presence of alloy disorder and the N-induced defects [11-13], which lead to the localization of excitons [11-17] and the presence of nonradiative centers. These centers are responsible for the low PL intensity at low temperatures as well as its further degradation at higher temperatures. Thermal annealing of these quantum well structures is known to improve significantly both of these characteristics as well as their crystalline quality. The temperature dependence of the PL characteristics of as-grown, as well as annealed, GaAsSbN/GaAs single quantum well (SQW) heterostructures for two different values of N composition (N=1.0% and 2.5%) have recently been reported by Harmand et al. [11], and in annealed samples by Lourenco et al. [12]. The temperature at which the detrapping of excitons occurs was found to depend on the N concentration. Lourenco et al. [12] attributed the thermal quenching of PL at higher temperatures to the presence of two nonradiative recombination channels with activation energies close to 45.0 and 7.5 meV, respectively.

In this paper we report on the effect of annealing on the photoluminescence (PL) characteristics of GaAsSbN/GaAs SQW structures. The focus of our work is on the temperature dependence of the PL characteristics of GaAsSbN/GaAs quantum well

structures annealed in-situ under As overpressure and ex-situ in a N ambient. Improved PL characteristics are obtained in the case of in-situ annealing in an As ambient. We observe a very pronounced “S-curve” behavior of the PL peak energy as a function of temperature, thus suggesting a strong localization of excitons by the potential fluctuations and/or defects in GaAsSbN/GaAs SQW structures.

## II. Experimental details

GaAsSbN/GaAs single quantum well structures were grown on undoped (100) GaAs substrates by the molecular beam epitaxial [MBE] technique in an EPI 930 system equipped with elemental Ga, As, and Sb cracker sources and with an EPI plasma RF Unibulb for N. The SQW structures consisted of a 450 nm thick GaAs buffer layer grown at 580 °C, GaAsSb(N) QWs grown in the temperature range of 490-450 °C and a 400 nm GaAs cap layer at 580 °C. GaAsSbN/GaAs SQWS grown at 490 °C and 450 °C will be referred to as sample B (N=1%, Sb= 22.5%, QW thickness=7.4 nm) and C (N=0.7%, Sb= 33.8%, QW thickness=7.6 nm), respectively. These were subjected to post-growth furnace annealing with a GaAs substrate cover on the top in a N ambient at 700 °C for 10 minutes. GaAsSbN/GaAs SQW grown at 470 °C and in-situ annealed in an As overpressure at 650 °C for 10 minutes will be referred to as sample D (N=1.5%, Sb=31.5 %, QW thickness=9.2 nm with thinner cap layer thickness=160 nm). For comparison, a GaAsSb/GaAs SQW was also grown at 490 °C, which will be referred to as sample A (Sb=32.8%).

The Sb and N compositions were estimated from a combination of x-ray diffraction (XRD) and secondary ion mass spectroscopy (SIMS) analysis carried out on these samples. The error bars in the estimation of N and Sb are  $\pm 0.1\%$  and  $\pm 1.5\%$ , respectively, for the samples B and C. The composition of Sb and N for sample D was estimated from x-ray analysis only, and hence represents only a rough estimate of the composition.

High resolution x-ray measurements were performed with a Bede Scientific Metrix-F automated diffractometer, equipped with a Microsource X-ray generator. The motorized detector slit was set to 0.5 mm wide, giving a  $2\theta$  angular resolution of 150 arc-sec.

PL measurements were carried out using a He-Ne laser as the light source for excitation, a 0.32 m double grating monochromator for wavelength dispersion and a Ge detector for the signal detection using the conventional lock-in technique. A closed cycle three stage APD cryogenic system was used to perform the PL temperature dependence study of the QWs.

It may be noted that temperature dependence of the PL was carried out on numerous samples, however our study is focused on the above three representative nitride samples.

Atomic resolution z-contrast imaging was done in an aberration corrected dedicated scanning transmission electron microscope (STEM) with an acceleration voltage of 300 kV. The spatial resolution is about 0.1 nm, more than enough to resolve the dumb-bell structure in GaAs. The mass thickness contrast of a z-contrast image makes it possible to determine the width, homogeneity and interface roughness directly.

### III. Results and discussion

Figures 1a and 1b exhibit the temperature dependence of the PL spectra in the as-grown and annealed sample C, respectively. The low temperature PL spectra of the annealed sample has a highly asymmetric line shape with a sharp high energy cut off and an exponential low energy tail, consistent with the observation of Lourenco et al. [12]. In the as-grown sample C, the PL spectra consist of two overlapping bands of comparable intensity, which are separated by about 20 meV. In the sample B, grown at a higher temperature, an additional low energy peak shifted by 11 meV (not shown) is also observed. At around 130 K in sample C the recombination from the two bands merges into one band. The dominant high energy peak exhibits a red shift with increasing temperature up to 50 K and thereafter shows a slight blue shift. In the annealed sample C, no satellite peaks are observed and the blue shift at 80 K is quite pronounced (see Fig 1b and Fig. 3). Similar behavior in the temperature dependence of the PL characterization was also observed in samples B and D. For the sample D, which was in-situ annealed in an As ambient, the degree of asymmetry in the PL line shape is significantly reduced as shown in Fig. 2 and at 75 K and beyond the PL shape is quite symmetric. In Fig. 3, the PL peak energy of the samples A, B, C and D is plotted as a function of temperature. The temperature dependence of the PL spectra was fitted using the Varshni's equation

$$E_g(T) = E_g(0) - \frac{\alpha T^2}{\beta + T} \quad (1)$$

where T is the absolute temperature,  $E_g(0)$  is the band gap at 0 K and  $\alpha$  and  $\beta$  are constants. The values of these parameters are listed in Table 1. The values of  $\alpha$  and  $\beta$  of nitride QWs are found to be comparable to that of GaAsSb.

Excellent fit is obtained with the Varshni's model for sample A over the entire temperature region, while for samples B, C and D there is a considerable difference (particularly in samples B and C) between the experimental and simulated values for temperatures below 110 K. This is due to the well known S-shaped behavior of the PL peak energy with temperature, indicative of carrier localization. This is commonly observed in nitride samples and has generally been considered an evidence for the exciton localization at potential fluctuations. The values of the maximum localization energy and of the FWHM delocalization temperature for all the samples are listed in Table I. The evolution of the FWHM with temperature is shown in Fig. 4. It increases monotonically with temperature in sample A as expected. However, for other samples it shows a rapid increase with temperature up to the delocalization temperature of 105 K, and thereafter decreases with further increase in temperature up to 180 K and finally increasing with temperature up to 300 K. This anomalous behavior near the delocalization temperature is considered to be a signature of the carrier localization as has been well demonstrated in numerous alloy system such as GaAsN [18-19] and InGaAsN [16-17]. This effect is more pronounced in the sample C than in sample D, consistent with the sharp "S-curve" behavior in the former sample.

In order to understand the nature of the recombination mechanisms, the temperature dependence of integrated PL intensity was also measured on these samples, as illustrated in Figure 5. The PL intensity decreases rapidly with increase in temperature and becomes undetectable for temperatures above 80 K. The temperature dependence of the PL intensity is usually expressed as [12]

$$I(T) = \frac{I_0}{1 + A * \text{Exp}\left(-\frac{E_a}{k * T}\right) + B * \text{Exp}\left(-\frac{E_b}{k * T}\right)} \quad (2)$$

where the presence of two nonradiative recombination channels is assumed.  $I_0$  is the intensity at  $T=0$  K,  $E_a$  and  $E_b$  are the thermal activation energy of the nonradiative center of first and second nonradiative centers, respectively.  $A$  and  $B$  represent the efficiency of these two recombination mechanisms. The values of  $I_0$ ,  $A$ ,  $B$ ,  $E_a$  and  $E_b$  that resulted in the best fit to the experimental data are also listed in Table I. The values of  $E_a$  and  $E_b$  of all the nitride samples are comparable. However, our values for  $E_a$  are smaller than the 45 meV reported by Lourenco et al.[12].

Figure 6 illustrates the high resolution x-ray diffraction spectra of as-grown and annealed sample C. The nitride peak position remains unaltered although the peak counts appear to have increased marginally.

Finally, the atomic resolution cross sectional TEM z-contrast image of the annealed sample D is shown in Fig. 7. It shows excellent quality of the QW layer. It did not reveal presence of any extended defects or clusters, however some contrast modulation, attributed mainly to the composition modulation, at both the interfaces of the QW is observed. The QW width fluctuations are in the 2-4 ML ranges at each of the interfaces.

The difference in the PL peak energies observed at low temperature and at room temperature for GaAsSb and GaAsSbN QW structures are 60 meV and ~40 meV, respectively. These values are smaller than 76 meV reported by Chiu et al.[20] for GaAsSb QW and 60 meV for nitrides by Lourenco et al. [12]. A similar, small shift in the PL peak energy with temperature has also been observed in InGaAsN by Pinault et al.[16]. The origin of the lower energy peak in as-grown sample C is not understood at this time.

The effects of annealing in N and As ambients on the PL line shape and its temperature dependence appear to be different. The samples annealed in a N ambient exhibit stronger localization in the order of 25-32 meV as exhibited by the pronounced inverted S-curve in the temperature dependence of the PL peak energy, while it is shallow (~16 meV) in samples annealed in-situ under an As overpressure. The PL line shape is highly asymmetric for N-annealed samples. By comparison, the PL shape of sample D is symmetric and exhibits the lowest FWHM of 54 meV at room temperature. Invariance of x-ray diffraction peaks with annealing in sample C rules out any significant outdiffusion of N and Sb that would alter the composition of the QW. Hence it is speculated that the overpressure of As from the covered substrate during the ex-situ annealing in N ambient, may not be sufficient to overcome the As desorption from the cap layer. This in turn can lead to some vacancies at the interface, which then becomes the driving force for the outdiffusion of N and Sb locally at the interface, as suggested by Bhat et al. [21] in the case of InGaAsN, leading to increased composition modulation and/or rearrangement of bonding at the interface. This increased composition modulation can cause the nonuniformity in local strain due to the large difference in the sizes of N and Sb atoms, which can result in both the increased localization energy and nonradiative recombination centers at the band edges.

The growth temperature has only minor effect on the degree of localization. The PL of the sample grown at 490 °C exhibits the largest carrier localization energy as indicated in the Table 1. The large localization energy for this sample results likely from the presence of increased N concentration in this sample, which enhances the induced defect density. This is also consistent with the observation that low temperature PL spectra of sample B, exhibiting an additional N induced low energy band in comparison to sample C.

The activation energy  $E_a$  determined from the intensity dependence of the PL spectra in the higher temperature region is of the same magnitude as the exciton localization energy. This suggests that  $E_a$  represents the activation energy of the nonradiative mechanism of the carrier delocalized from the trapped exciton in the localized state. The PL quenching at lower temperature (less than 100 K) is due to the low activation energy  $E_b$  (6-11 meV) recombination process occurring at the tail states. This value of activation energy is close to the energy 9.0 meV corresponding to the delocalization temperature. It may be noted that the ratio  $A/B$ , which represents the ratio of efficiency of these two recombination mechanisms is higher for sample D, indicating the dominance of recombination mechanism corresponding to  $E_a$ , that is responsible for PL quenching at higher temperature. This explains the large change in the intensity observed in this sample as compared to other nitride samples. Further, the PL efficiency of this QW is comparable to the other samples though the N composition is higher than other samples. This is also consistent with our earlier conclusion that in the As annealed samples, the presence of As significantly reduces the density of nonradiative recombination centers arising from the band edge states. In addition, the higher value of  $\beta$  obtained on this sample suggests a more stable temperature dependence. The localization observed in this sample D is attributed mainly to the compositional modulation at the interface as evidenced from the TEM micrograph.

As mentioned earlier Harmand et al. [11] have studied the variation of the PL peak position with temperature in as-grown as well as annealed SQWs of GaAsSbN/GaAs. They find that the values of the localization energies in as-grown samples are considerably larger than those in the annealed samples. In our sample C, however we find the opposite result. Annealing appears to have deepened the localizing centers. In addition, our values for the localization energies are much larger than those observed by Harmand et al. [11] and Lourenco et al. [12]. The reason for this difference in behavior is not clear at this time.

A detailed study of the effects of growth conditions and different annealing procedures on the optical characteristics will be presented elsewhere.

## V. Conclusions

In summary, the temperature dependent PL characteristics of GaAsSbN/GaAs SQWs exhibit an “inverted S-curve” at low temperatures indicative of carrier localization. The carrier localization is shallow in as-grown samples and becomes pronounced on annealing in a N ambient. This is attributed to the enhanced defect-induced diffusion at the interface, thereby leading to composition modulation and/or rearrangement of bonding at the interface. Annealing in an As ambient therefore improves significantly the PL characteristics with reduced carrier localization. The thermal annealing of PL intensity has been attributed to the presence of two nonradiative

recombination centers with activation energies in the range of 24-32 meV and 7-11 meV, respectively. SQWs with room temperature PL energy as low as 0.81 eV with FWHM of 54 meV have been achieved in these samples.

### **Acknowledgements**

This work is supported by Army Research Office (Grant No. DAAD19-02-004, Contract No. W911NF-041-0025), NASA (Grant No. NAG3-2782), and DOD (Contract No. DAAD19-03-2-0016). SIMS measurements were carried out by Evans East of the Evans Analytical Group.

## References

1. M. Weyers, M. Sato and H. Ando, *Jpn. J. Appl. Phys.* **31**, L853 (1992).
2. M. Kondow, K. Uomi, K. Hosomi and T. Mozume, *Jpn. J. Appl. Phys.* **33**, L1056 (1994).
3. M. Kondow, K. Uomi, A. Niwa, T. Kitatani, S. Watahiki and Y. Yazawa, *Jpn. J. Appl. Phys.* **35**, 1273 (1996).
4. M. Reinhardt, M. Fischer, M. Kamp and A. Forchel, *Electron. Lett.* **36**, 1025 (2000).
5. D. A. Livshits, A. Y. Egorov and H. Riechert, *Electron. Lett.* **36**, 1381 (2000).
6. J. S. Harris Jr., *Semicond. Sci. Technol.* **17**, 880 (2002).
7. S. R. Kurtz, J. F. Clem, A. A. Allerman, R. M. Sieg, C. H. Seager and E. D. Jones, *Appl. Phys. Lett.* **80**, 1379 (2002).
8. P. M. Asbeck, R. J. Welty, C. W. Tu, H. P. Xin and R. E. Welser, *Semicond. Sci. Technol.* **17**, 898 (2002).
9. G. Ungaro, G. Le Roux, R. Teissier and J. C. Harmand, *Electron. Lett.* **35**, 1246 (1999).
10. J. C. Harmand, A. Caliman, E. V. K. Rao, L. Largeau, J. Ramos, R. Teissier, L. Traverse, G. Ungaro, B. Theys and I. F. L. Dias, *Semicond. Sci. Technol.* **17**, 778 (2002).
11. J. C. Harmand, G. Ungaro, J. Ramos, E. V. K. Rao, G. Saint-Girons, R. Teissier, G. Le Roux, L. Largeau, and G. Patriarche, *J. Crystal Growth* **227-228**, 553 (2001).
12. S. A. Lourenco, I. F. L. Dias, L. C. Pocas and J. L. Duarte, *J. Appl. Phys.* **93**, 4475(2003).
13. T. K. Ng, S. F. Yoon, S. Z. Wang, W. K. Loke, and W. J. Fan, *J. Vac. Sci. Technol.* **B 20**, 964(2002).
14. J. Misiewicz, G. Sek, R. Kudrawiec, K. Ryczko, D. Gollub, J. P. Reithmaier and A. Forchel, *Microelectronics Journal* **34**, 351(2003).
15. J. Misiewicz, P. Sitarek, K. Ryczko, R. Kudrawiec, M. Fischer, M. Reinhardt and A. Forchel, *Microelectronics Journal* **34**, 737(2003).
16. M.-A. Pinault and E. Tournie, *Appl. Phys. Lett.* **78**, 1562(2001).
17. S. Shirakata, M. Kondow and T. Kitatani, *Appl. Phys. Lett.* **79**, 54(2001).
18. X. D. Luo, Z. Y. Xu, W. K. Ge, Z. Pan, L. H. Li and Y. W. Lin, *Appl. Phys. Lett.* **79**, 958(2001).
19. I. A. Buyanova, W. M. Chen, G. Pozina, J. P. Bergman, B. Monemar, H. P. Xin and C. W. Tu, *Appl. Phys. Lett.* **75**, 501(1999).
20. Y. S. Chiu, M. H. Ya, W. S. Su, and Y. F. Chen, *J. Appl. Phys.* **92**, 5810 (2002).
21. R. Bhat, C. Caneau, L. Samlamanca-Riba, W. Bi and C. Tu, *J. Crystal Growth* **196**, 427 (1998).

## Figure Captions

FIG.1. Variation in the PL spectra with temperature of sample C: (a) as-grown sample, (b) annealed in N ambient.

FIG.2. Variation in the PL spectra with temperature of in-situ annealed sample D.

FIG.3. PL peak energy variation with temperature for samples A, B, C and D. Superimposed is the fit from Varshni's model .

FIG.4. Temperature dependence of FWHM of samples A, C and D.

FIG.5. Temperature dependence of integrated PL intensity for samples A, B, C and D.

FIG.6. HRXRD spectra of as-grown and annealed sample C.

FIG.7. The atomic resolution cross sectional TEM z-contrast image of sample D.

Table I. Jia Li *et al*

Sample	$E_{10K}$ (eV)	$\alpha$ (meV/K)	$\beta$ (K)	$E_{loc}^{max}$ (eV)	$T_{deloc}$ (K)	$E_a$ (meV)	$E_b$ (meV)	<b>A/B</b>
A	1.039	0.363	245			32	8	83
B	0.997	0.38	256	0.0322	110	24	7	22
C	0.915	0.40	283	0.0256	105	26	11	24
D	0.849	0.37	350	0.0168	105	27	9	42

Figure 1. Jia Li *et al*

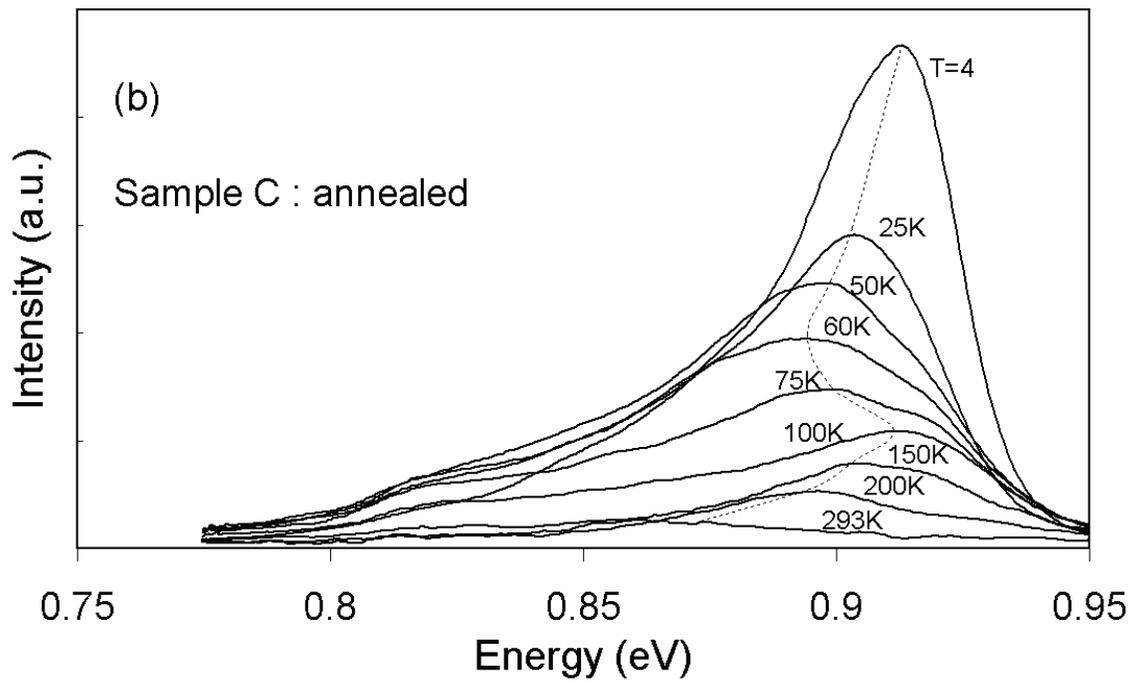
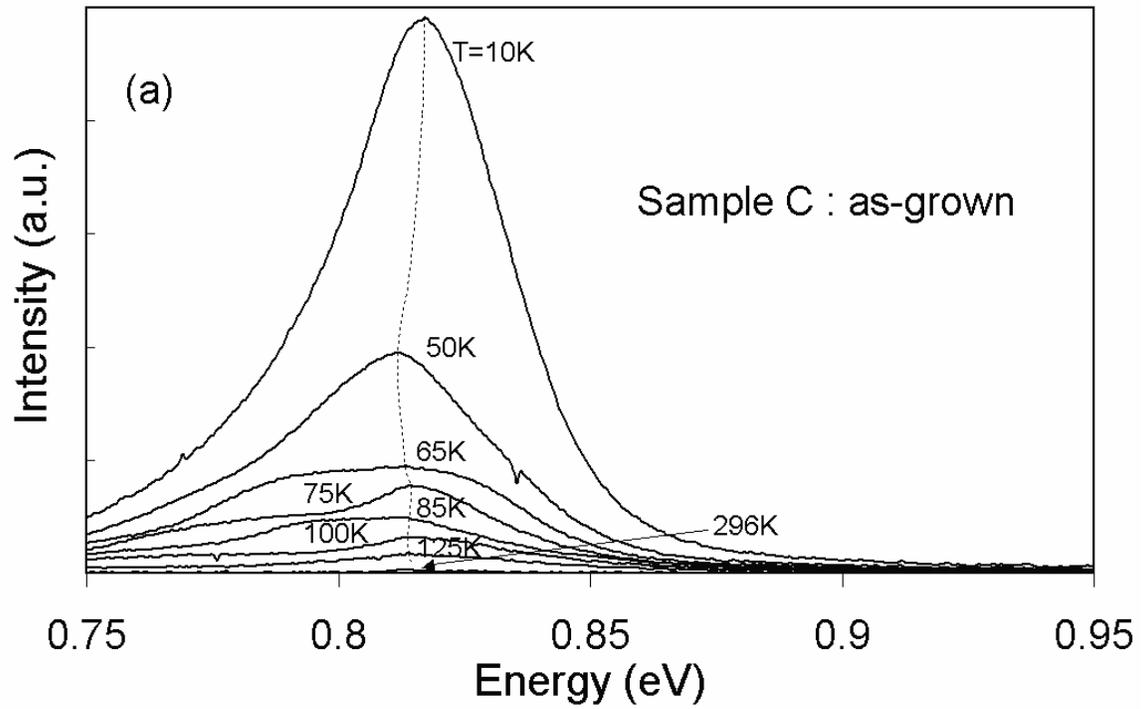


Figure 2. Jia Li *et al*

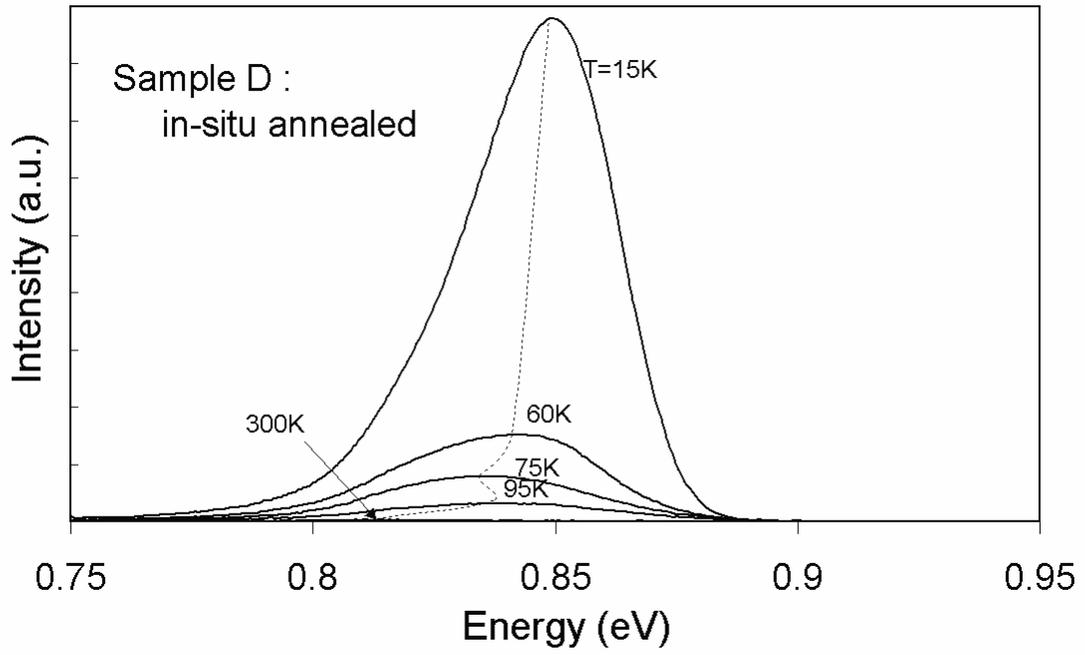


Figure 3. Jia Li *et al*

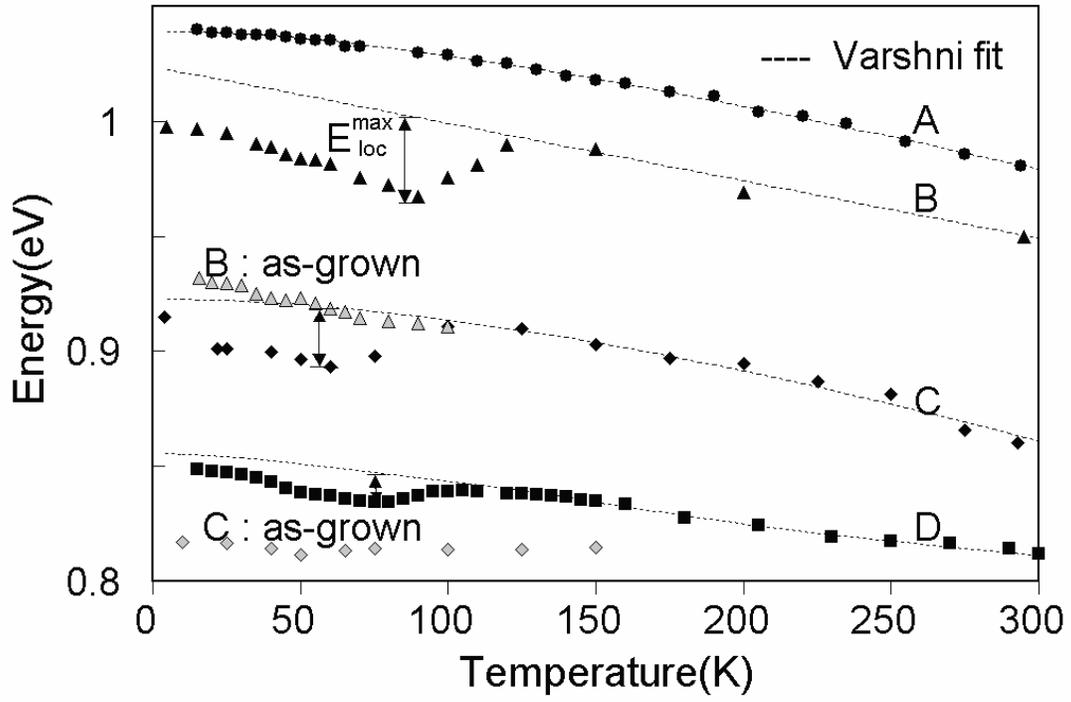


Figure 4. Jia Li *et al*

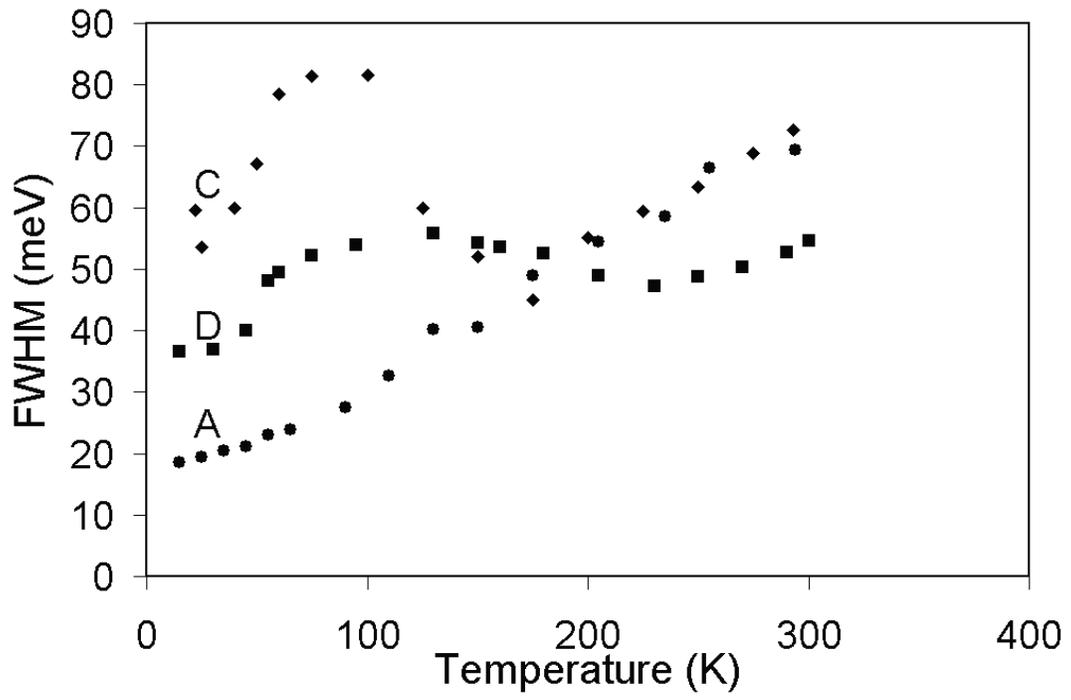


Figure 5. Jia Li *et al*

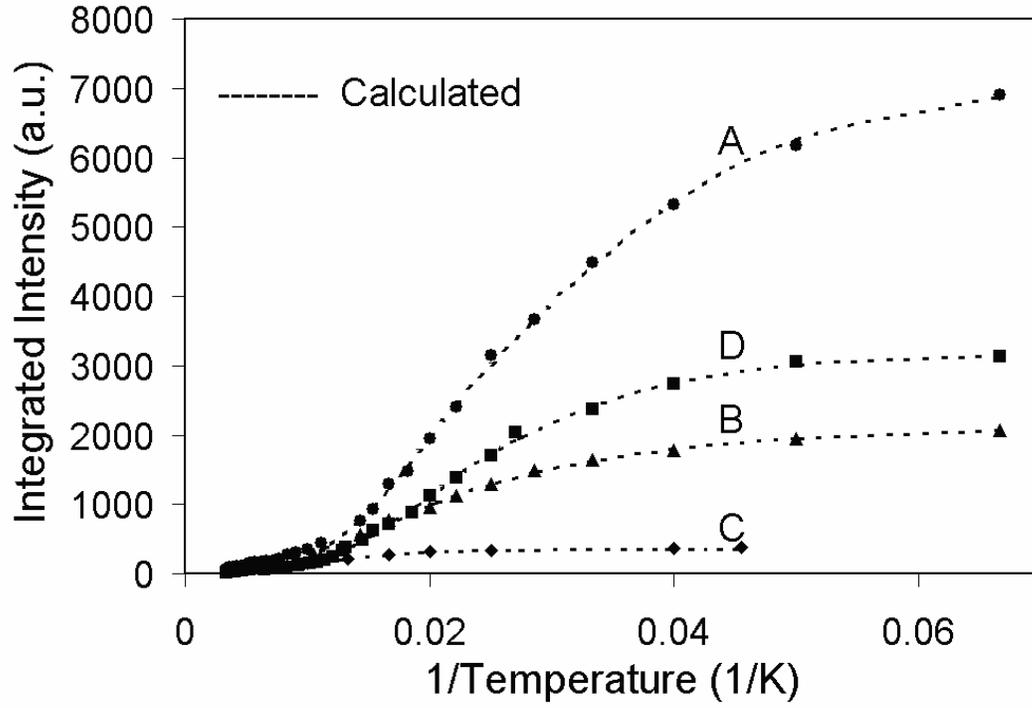


Figure 6. Jia Li *et al*

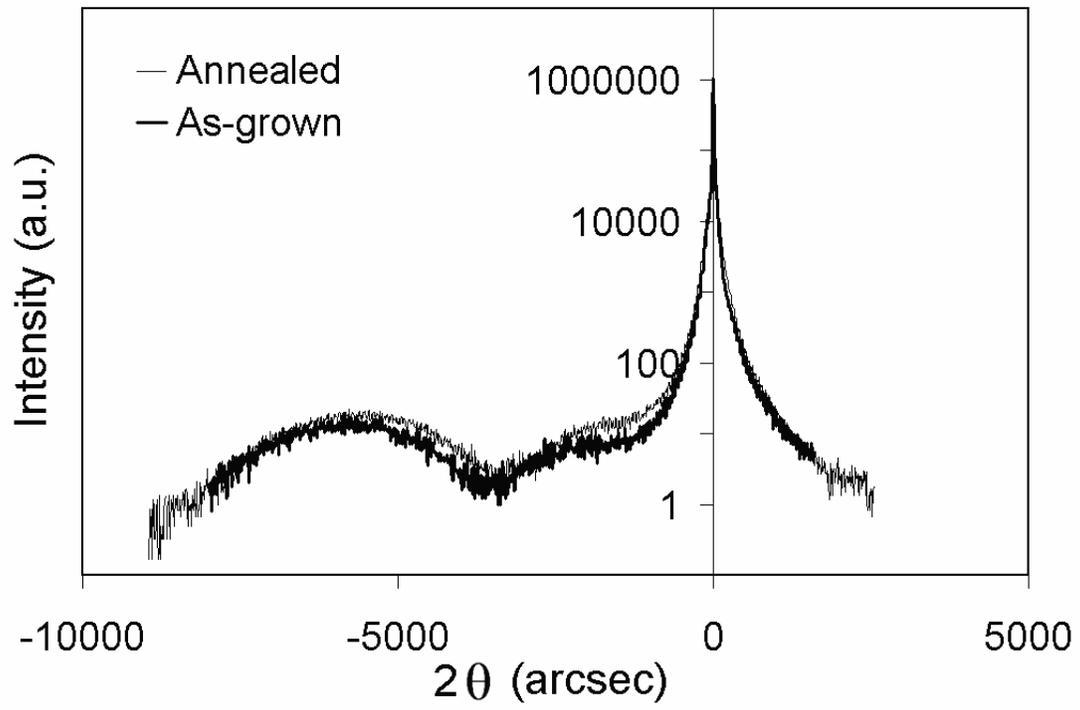
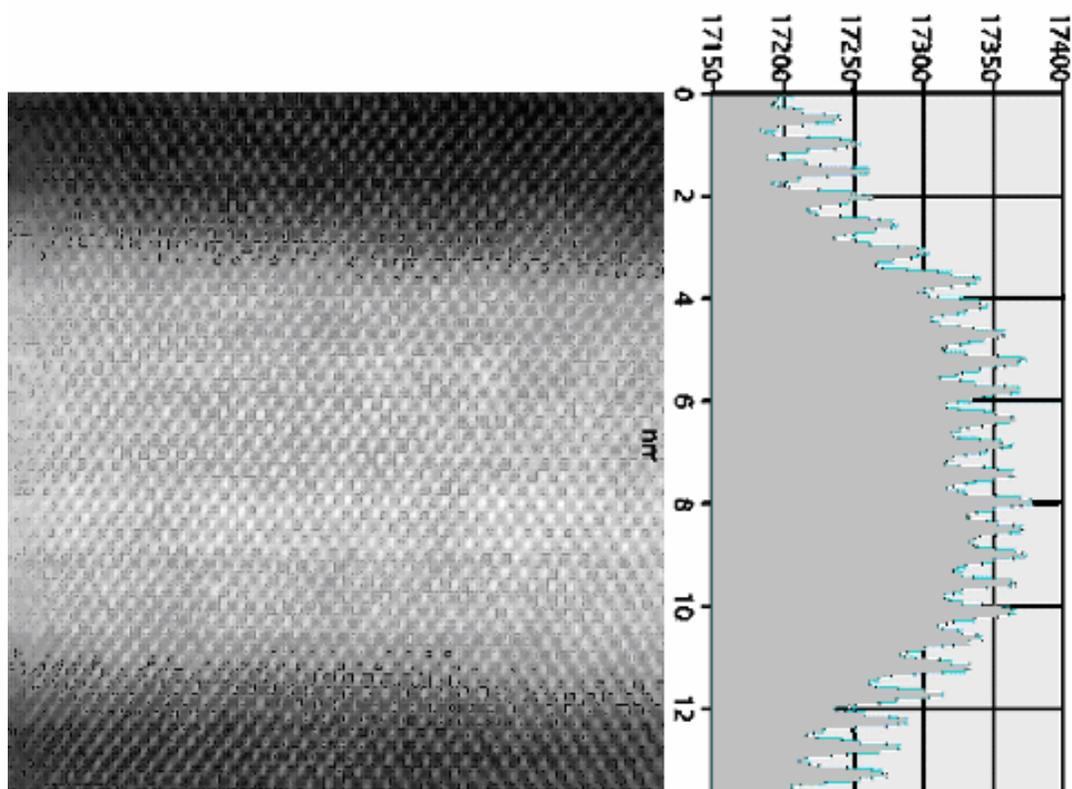


Figure 7. Jia Li *et al*



### **C. Effect of Nitrogen on GaAsSbN/GaAs SQWs**

## **Effect of Nitrogen on GaAsSbN/GaAs Single Quantum Wells**

### **Abstract**

In this work we report on the effect of nitrogen (N) on the structural and optical properties of GaAsSbN single quantum wells (SQWs) grown by solid source molecular beam epitaxy (MBE). GaAlAs layers were used as the barrier. The samples are grown at 470°C and annealed in-situ in As ambient at 650°C for 10 min. The Sb composition in the layers was 28-30%. A 4K PL peak energy of 0.78 eV with low FWHM of 28 meV was achieved on SQWs for N concentration of 1.4%. The presence of Pendullosong and high frequency fringes due to the cap layer for the entire composition range of N varying up to 2.3% in high-resolution x-ray diffraction (HRXRD) spectra of the SQWs, attest to the excellent structural quality of the grown layers and interfaces. With increase in N concentration up to 0.7% the energy gap decreases at a rate of 270meV/% change in N, while with further increase in N concentration the change in PL peak energy reduces to 30meV/% change in N and for 2.3% N concentration the PL peak energy increases.

PACS: 71.55 Eq, 73.21 Fg, 78.55.Cr, 78.67.De

## I. Experiment

GaAsSbN/GaAs single quantum well structures were grown on undoped (100) GaAs substrates by the solid source molecular beam epitaxial [MBE] technique in an EPI 930 system equipped with elemental Ga, As, and Sb cracker sources and with an EPI RF plasma Unibulb for N. The SQW structures are sandwiched between a 35 nm thick GaAs layer followed by a 150 nm thick GaAlAs grown at 580°C. The substrate temperature for the QW layer grown is 470°C and the thickness is about 8-9 nm. All the grown layers were subjected to in-situ annealing in an As overpressure at 650°C for 10 minutes.

High-resolution x-ray measurements were performed with a Bede Scientific Metrix-F automated diffractometer, equipped with a Microsource X-ray generator. The motorized detector slit was set to 0.5 mm wide, giving a  $2\theta$  angular resolution of 150 arc-seconds.

PL measurements were carried out using a He-Ne laser as the light source for excitation, a 0.32 m double grating monochromator for wavelength dispersion and a Ge detector for the signal detection using a conventional lock-in technique. A closed cycle three-stage APD cryogenic system was used to perform the PL temperature dependence study of the QWs.

SIMS was carried out on selected QWs with N composition of 0.53%, 1.41% and 2.32% corresponding to the N flux of 1, 4 and  $9 \times 10^{-7}$  torr, respectively. The composition of Sb in these samples were then determined using Bede RADS Mercury software. The error bars in the estimation of N and Sb are  $\pm 0.1\%$  and  $\pm 1.5\%$ , respectively in these samples. The compositions of Sb and N for other samples were estimated from N flux used and x-ray analysis, and hence represent only a rough estimate of the composition.

## II. Results and Discussion

Figure 1 shows the HRXRD spectra of a GaAsSbN SQW with GaAlAs barriers grown for different N fluxes. The presence of Pendullossung fringes and superimposed high frequency fringes indicate sharp interfaces and excellent quality of the layers grown. The Sb composition was found to be 28.4% for the reference GaAsSb SQW structure. With the introduction of small amount of N (N=0.53%), the compressive strain is enhanced due to the increased incorporation of Sb. With further increase in the N flux up to  $9 \times 10^{-7}$  torr, the compressive strain reduces monotonically as expected. The N composition in the layer increases with N flux as shown in Fig. 2, however, Sb incorporation shows an initial increase to 35% with N composition varying up to 0.7% and thereafter it comes back to the composition as in the reference GaAsSb QW.

Figure 3 exhibits the variation of PL peak energy and FWHM as a function of N flux. Introduction of N reduces the peak energy as expected. However, it does not vary linearly in the entire range. The reduction is faster initially corresponding to a decrease of 270meV/% change in N, thereafter the decrease is small 30meV/% change if N. The QW with 2.3% N exhibits an increase in PL peak energy with increased FWHM. For the above sample the N composition is estimated to be 0.8-1% from the position of PL peak energy, which is much less than that determined from SIMS measurement. These suggest that N is probably incorporated in the interstitial position and sets the upper limit.

Figures 4(a) - 4(c) exhibits the variation in the low temperature PL spectra for different values of the laser excitation power, for SQWs grown at different N flux. An asymmetric PL line is observed at low-excitation power. With increasing excitation

power, the PL intensity of the low energy tail is raised due to the gradual filling of the empty energy states in the band tails of the GaAsSbN SQW, yielding a more symmetric line shape. The PL peak energy blue shift observed in these samples is in the range of 1-2.4 meV/decade for excitation power increase from 10 mW ( $0.3 \text{ W/cm}^2$ ) to 326 mW ( $10 \text{ W/cm}^2$ ). This value is much less than those observed GaAsN SQWs (12-18 meV/decade) [1] or epilayers (16 meV/decade) [2]. The energy shift being so low can be assigned to the recombination of excitons trapped by the lesser potential fluctuations at the band edge.

In Figure 5, a plot of PL intensity versus excitation power is shown for all the N flux. The behavior of the PL intensity with excitation power is found to be similar for all the samples with an initial increase and saturating beyond excitation power of 4-5  $\text{W/cm}^2$ ,

### III. References

- [1] H.Dumont, L. Auvray, Y. Monteil, F.Saidi, F. Hassen, H. Maaref, Optic Materials, 24 (2003), 303-308.
- [2] F.Saidi, F. Hassen, H. Maaref, L. Auvray, H.Dumont, Y. Monteil, Materials Science and Engineering C, 21 (2002), 245-249.

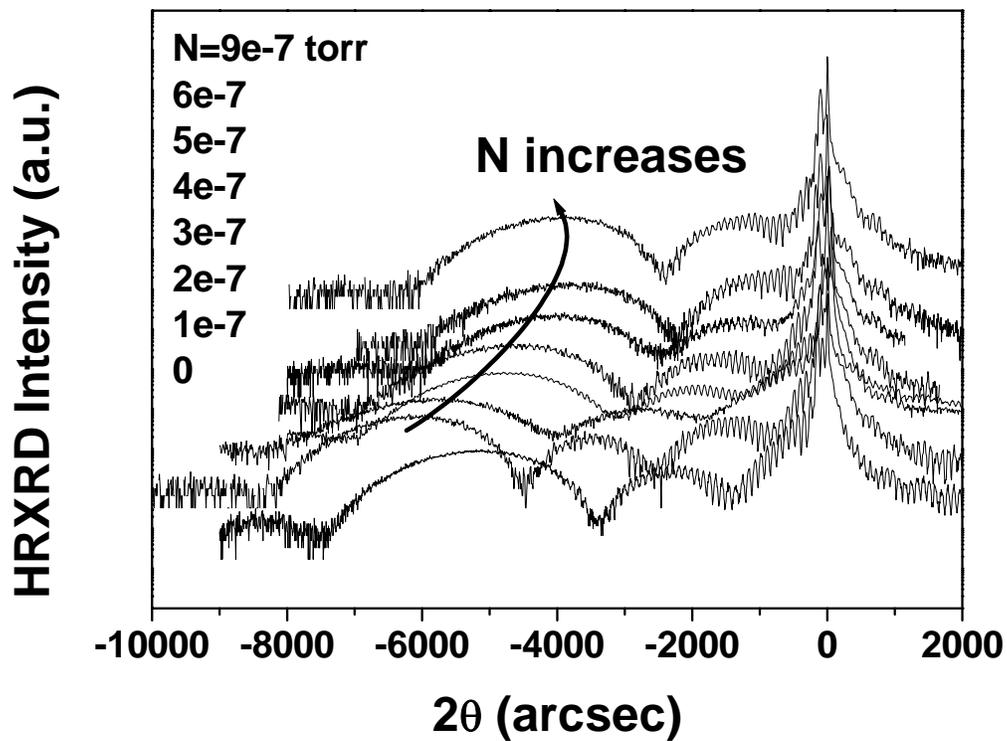


Figure 1. High resolution x-ray spectra of GaAsSbN/GaAs and GaAsSb/GaAs SQWs with varying N flux from  $1-9 \times 10^{-7}$  torr showing a shift in the nitride peak.

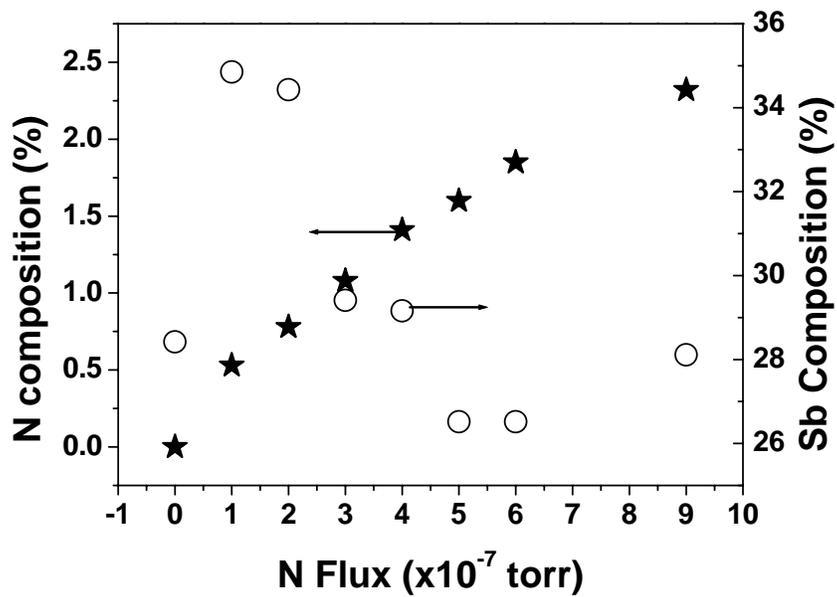
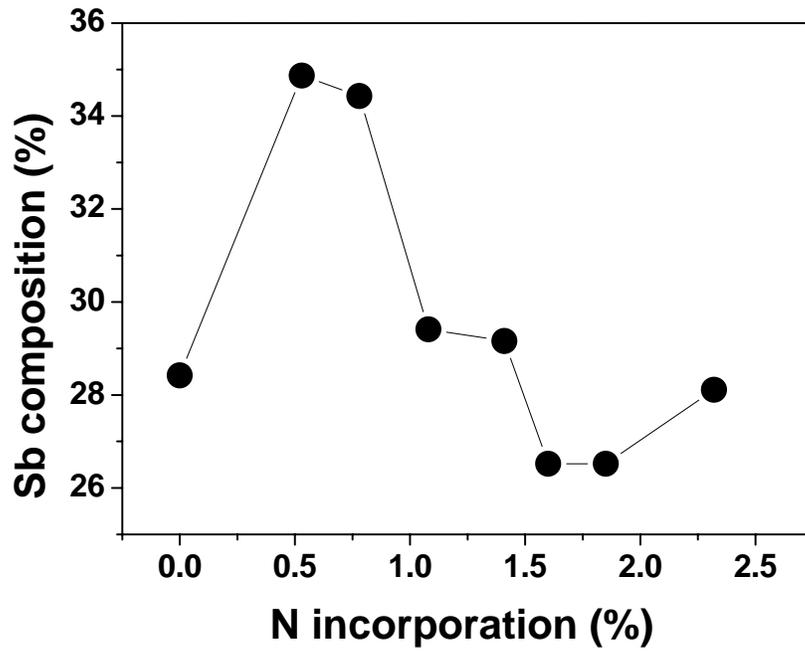


Figure 2. The compositional variations in Sb and N with (a) N incorporation and (b) N flux

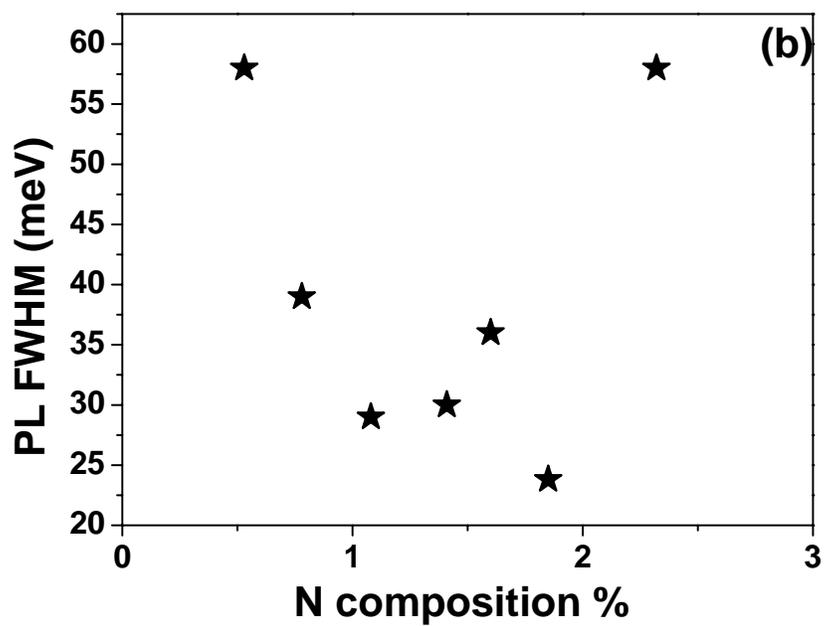
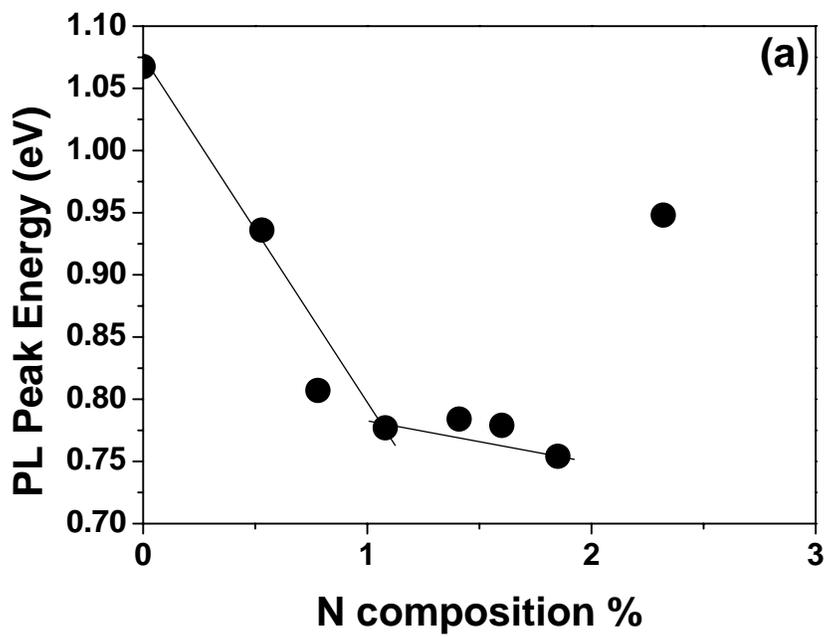
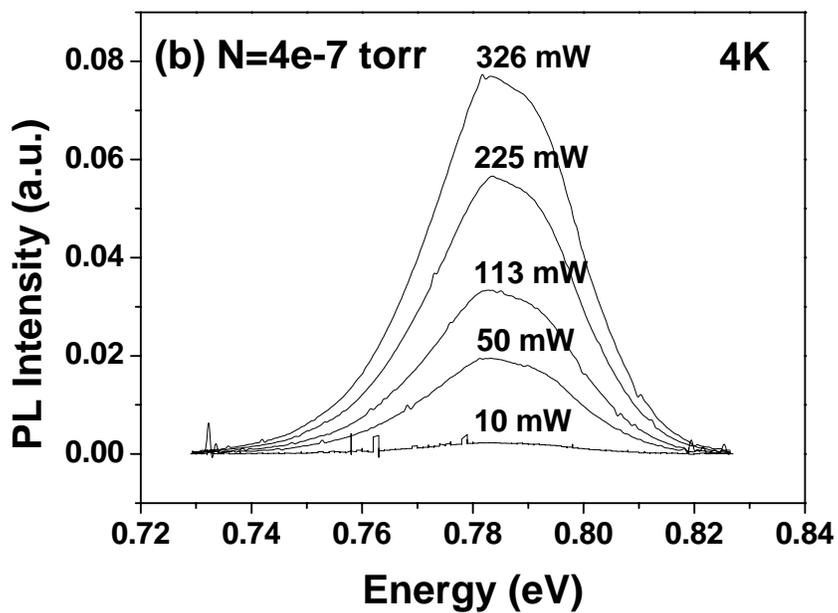
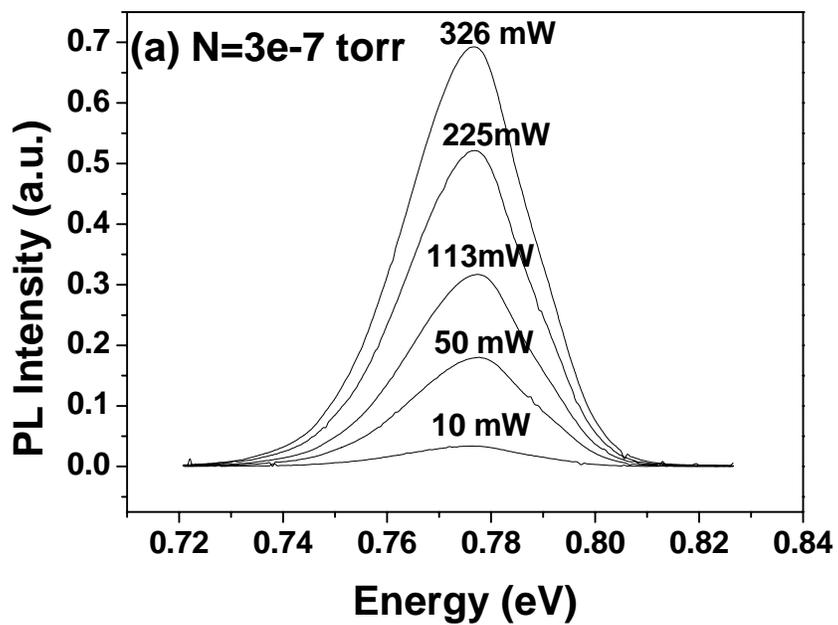


Figure 3. The variations in (a) PL peak energy and (b) PL FWHM with N composition



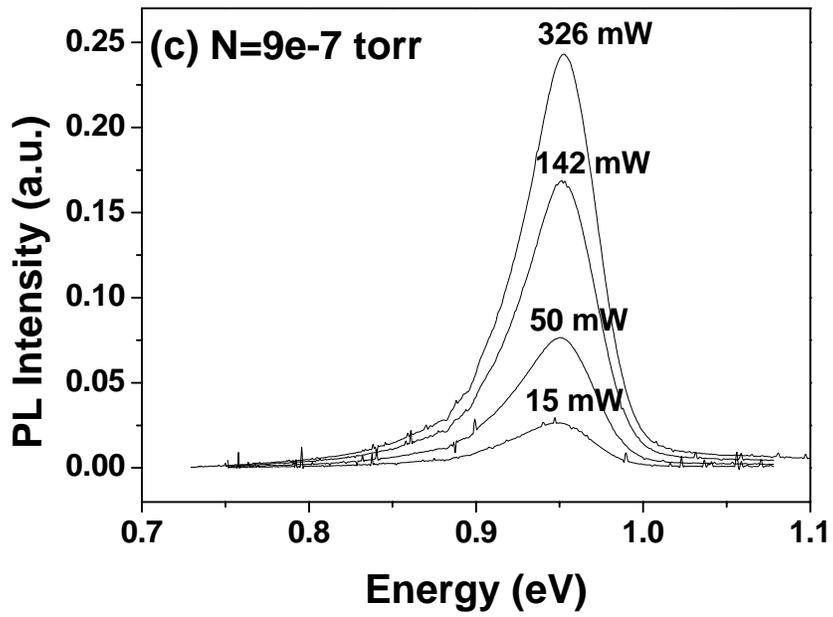


Figure 4. Low temperature PL peak intensity and peak energy shift dependence on the excitation energy for samples of N flux (a)  $3 \times 10^{-7}$  torr, (b)  $4 \times 10^{-7}$  torr and (c)  $9 \times 10^{-7}$  torr.

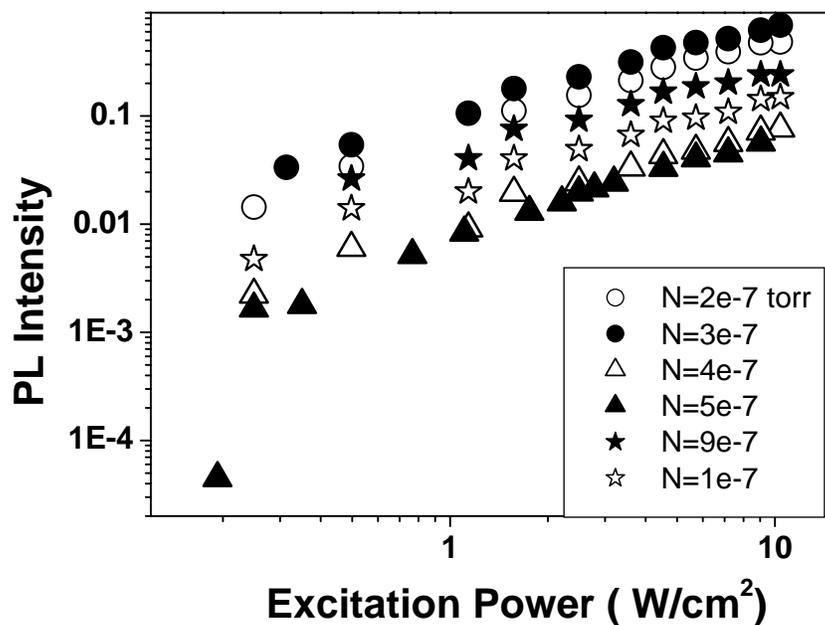


Figure 5. Low temperature PL peak intensity dependence on laser excitation power for samples of various N flux varying from  $1-9 \times 10^{-7}$  torr.

## IV. PUBLICATIONS & THESIS ARISING FROM ARO

### Refereed Publications

Liangjin Wu, **Shanthi Iyer**, Kalyan Nunna, Jia Li, Sudhakar Bharatan, Ward Collis and Kevin Matney “MBE Growth Study of GaAsSbN/GaAs Single Quantum Wells” Mat. Res. Soc. Symp. Proc. 799, Z1.9.1 (2003).

### Non-refereed Publications and Presentations

Liangjin Wu, Kalyan Nunna, Jia Li, Sreenivasa Kothamasu, Ward Collis, **Shanthi Iyer**, Kevin Matney, K. Bajaj, “MBE Growth Study of GaAsSbN/GaAs Single Quantum Wells” Presented at MRS Fall Meeting 2003.

Jia Li, Sudhakar Bharatan, Kalyan Nunna, Liangjin Wu, **Shanthi Iyer**, K. Bajaj, “Carrier Localization in GaAsSbN/GaAs Quantum Wells” Presented at MRS Fall Meeting 2003.

*David L. Jones*, **Shanthi Iyer**, Liangjin Wu, Jia Li, Ward Collis, S. M. Hegde, Kevin Matney, K. K. Bajaj and R.T. Senger “MBE Growth and PL Studies of GaAsSb/GaAs And GaAsSbN/GaAs Quantum Well Heterostructures” APS March Meeting, Austin, TX March 3, 2003.

*David Jones*, Liangjin Wu, Jia Li, Sreenivasa Kothamasu, Ward Collis, and **Shanthi Iyer** “MBE Growth and Properties of GaAsSb/GaAs Quantum Wells” , Ron McNair Symposium on Science and Technology Frontiers Jan. 27, 2003.

*Sreenivasa Kothamasu*, Jial Li , Liangjin Wu, David Jones, Kalyan Nunna and **Shanthi Iyer**, “Characterization of GaAs<sub>1-x</sub>Sb<sub>x</sub>N/GaAs Quantum Well Heterostructures Grown by Molecular Beam Epitaxy using High Resolution X-Ray Diffraction” Ron McNair Symposium on Science and Technology Frontiers Jan. 28, 2003.

*David Jones*, Liangjin Wu, Jia Li, Sreenivasa Kothamasu, Ward Collis, and **Shanthi Iyer** “MBE Growth and Properties of GaAsSb/GaAs Quantum Wells” presented at MRS NC Section Meeting, MCNC, NC, Nov. 15, 2002.

### Manuscript Submitted

Jia Li, **Shanthi Iyer**, Sudhakar Bharatan, Liangjin Wu, Kalyan Nunna, Ward Collis, K. Bajaj, K. Matney and Gerd Duscher “Annealing Effects on the Temperature Dependence of the Photoluminescence of GaAsSbN Single Quantum Wells”- submitted to J. Appl. Phys.

L. Wu, **S. Iyer**, K. Nunna, J. Li, S. Bharatan, W. Collis and K. Matney “MBE Growth and Properties of GaAsSbN/GaAs Single Quantum Wells”- submitted to J. Appl. Phys.

#### Manuscript in Preparation

“Effect of Nitrogen on GaAsSbN/GaAs SQWs”

#### Graduate Degrees Awarded

“X-ray Diffraction Study of GaAsSb/GaAs and GaAsSbN/GaAs Quantum Well Heterostructures”, Sreenivasa Kothamasu, 11/03

“Characterization of NiFe/Cu/Co Pseudo-Spin Vales by SQUID”, Jamil Woods, 07/03.

“A Photoluminescence Study of GaAsSbN/GaAs SQW Heterostructures”, Sudhakar Bharatan, 07/03.

“Molecular Beam Epitaxial Growth of GaAsSbN/GaAs Quantum Wells for 1.3-1.5  $\mu\text{m}$  Emission”, David Jones, 04/03.

“A Study of GaAsSb/GaAs Quantum Well Structures Grown by Molecular Beam Epitaxy”, Kalyan Nunna, 04/03

#### Undergraduate Project

David Le Clair, ( Winner of NC Space Grant Consortium Scholarship) “MBE Growth and X-Ray Characterization of GaAsSb/GaAs Quantum Well Heterostructures” Funded for \$4000, Summer 2002

#### High School Project

Stephanie Santiago, NASA SHARP apprentice, “X-ray Diffraction of Semiconductors”, Summer 2003.

## **V. PARTICIPATING SCIENTIFIC PERSONNEL AND REPORTS SUBMITTED**

### **Faculty**

*Shanthi Iyer, Professor*

### **Graduate Students**

David Jones, MSEE  
Sreenivasa Kothamasu, MSEE  
Sudhakar Bharatan, MSEE  
Kalyan Nunna, MSEE  
Liangjin Wu, Ph.D. candidate  
Sudhakar Bharatan, Ph.D. candidate  
Kalyan Nunna, Ph.D. candidate

### **Undergraduate Students**

David Leclaire

### **Collaborators**

Dr. Ralph Dawson, University of New Mexico  
Dr. Ward Collis, North Carolina A&T State University  
Dr. Kevin Matney, Bede Scientific Inc.  
Dr. K. K. Bajaj, Emory University  
Dr. Gerd Duscher, N.C. State University

### **Progress Reports**

Interim Progress Report: 11/01/01 -7/31/02  
Interim Progress Report, 8/1/02-04/30/03