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13. ABSTRACT (Maximum 200 words)

The wide bandgap III-nitride system $Al_yGa_xIn_{1-y-x}N$ has a myriad of applications in areas such as light emitting devices, and high frequency/high temperature electronics. Salient results derived from this research include such topics as the first report of the growth and characterization of the quarternary system $Al_yGa_xIn_{1-y-x}N$, the first description of the effect of H_2 gas on indium and impurity incorporation in $In_xGa_{1-x}N$, the experimental verification of ordering and phase separation in $In_xGa_{1-x}N$, the discovery of an optical memory effect in GaN, and the growth and characterization of novel multicolor emitting GaN/InGaN based double heterostructures.

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Final Progress Report

I. This final progress report for the research project "Development of InGaN and AlGaInN compounds for Optoelectronic Devices" summarizes results that have been obtained in the study of the $Al_yGa_xIn_{1-y-x}N$ system. This wide bandgap III-nitride system has a myriad of applications in areas such as light emitting devices, and high frequency/high temperature electronics. Results from our research include both material development and characterization and the growth of novel device structures.

II. Given below is a summary of some of the more important results derived from this research. Abstracts of these salient results are given below and include such topics as the first report of $Al_yGa_xIn_{1-y-x}N$ growth and characterization, the first description of the effect of H_2 gas on indium and impurity incorporation in $In_xGa_{1-x}N$, and the experimental verification of ordering and phase separation in $In_xGa_{1-x}N$. The abstract title, personnel who were involved in the work, and general time frame the material was peer reviewed in the literature are given.

1. Growth and characterization of AlInGaN quaternary alloys

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(Received 1 September 1995; accepted 20 October 1995)

We report on the deposition of $Al_yIn_xGa_{1-x-y}N$ in the $(0 < y < 0.15)$ and $(0 < x < 0.14)$ composition range by metalorganic chemical vapor deposition. AlInGaN quaternary alloys offer a lattice-matched platform for InGaN-based light emitting heterostructure devices. Epitaxial growth of AlInGaN on (0001) sapphire substrates has been achieved at 750 °C. Alloy composition, lattice constants, and band gaps were obtained by energy dispersive spectroscopy, x-ray diffraction, and room temperature PL. Band edge emissions dominate the PL spectra of these quaternary films. Preliminary data suggest that the lattice constant of AlInGaN can be deduced from chemical composition using Vegard's law, indicating solid solution in the grown quaternary films. ©1996 American Institute of Physics.

2. Effect of hydrogen on the indium incorporation in InGaN epitaxial films

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(Received 11 October 1996; accepted 22 November 1996)

The InN percent in metalorganic chemical vapor deposition (MOCVD) and atomic layer epitaxy (ALE) grown InGaN was found to be significantly influenced by the amount of hydrogen flowing into the reactor. The temperature ranges for this study are 710–780 °C for MOCVD, and 650–700 °C for ALE. For a given set of growth conditions, an increase of up to 25% InN in InGaN, as determined by x-ray diffraction, can be achieved by reducing the hydrogen flow from 100 to 0 sccm. Additionally, the hydrogen produced from the decomposition of ammonia does not seem to change the InN percent in the films, indicating that the ammonia decomposition rate is less than 0.1%. The phenomenon of having hydrogen control the indium incorporation was not reported in the growth of any other III–V compound previously studied. ©1997 American Institute of Physics.

3. Optical memory effect in GaN epitaxial films

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We report on memory effects in the optical properties of GaN and AlN epitaxial-films grown by atmospheric pressure metal organic chemical vapor deposition. After exposing selected areas of particular samples with He–Cd laser light (3.8 eV), we observed a persistent and marked decrease in the near band edge photoluminescence (PL) intensity emitted from these areas. This effect has been observed in epitaxial films that typically have a pyramid-like hillock surface. This ability to modulate PL emission intensity at individual points in these materials can be exploited as a method for optical data storage. A means of erasing information stored using this effect has also been investigated using lower energy (~ 2 eV). ©1997 American Institute of Physics.

4. Impurity dependence on hydrogen and ammonia flow rates in InGaN bulk films

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H, C, and O impurity concentrations in metalorganic chemical vapor deposition grown InGaN were found to be dependent on the hydrogen and NH3 flow rates. By increasing the hydrogen flow rate from 0 to 100 sccm, a decrease of greater than two orders of magnitude in the C and O impurity levels and one order of magnitude in the H impurity level was observed. Increasing the NH3 flow rate from 1 to 5 slm results in a decrease in the C concentration and an increase in the H and O concentrations indicating that high purity NH3 (99.999%) can be a significant source of O contamination. Additional studies show that when the InN percent in the InGaN films increases, the impurity concentrations increase regardless of changes in the growth conditions. The InGaN films were grown from 710 to 780 °C and the impurity concentrations were characterized by secondary ion mass spectrometry.

5. Phase separation in InGaN grown by metalorganic chemical vapor deposition

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We report on phase separation in thick InGaN films with up to 50% InN grown by metalorganic chemical vapor deposition from 690 to 780 °C. InGaN films with thicknesses of 0.5 μm were analyzed by Θ–2Θ x-ray diffraction, transmission electron microscopy (TEM), and selected area diffraction (SAD). Single phase InGaN was obtained for the as-grown films with < 28% InN. However, for films with higher than 28% InN, the samples showed a spinodally decomposed microstructure as confirmed by TEM and extra spots in SAD patterns that corresponded to multiphase InGaN. ©1998 American Institute of Physics.

6. Phase separation and ordering coexisting in $In_xGa_{1-x}N$ grown by metal organic chemical vapor deposition

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We have recently reported the occurrence of phase separation in $In_xGa_{1-x}N$ samples with x>0.25. Theoretical studies have suggested that InxGa1–xN can phase-separate asymmetrically into a low InN phase and an ordered high InN phase. In this letter, we report on the existence of simultaneous phase separation and ordering of InxGa1–xN samples with x>0.25. In these samples, phase separation was detected by both transmission electron microscopy selected area diffraction (TEM-SAD) and x-ray diffraction. Ordering was detected by both imaging and TEM-SAD. ©1999 American Institute of Physics.

III. Below is a list of publications and technical reports derived from this research project:

1. "Phase separation in InGaN grown by metalorganic chemical vapor deposition", N. A. El-Masry, E. L. Piner, S. X. Liu, and S. M. Bedair, Appl. Phys. Lett. **72**, 40 (1998).
2. "Phase separation and ordering coexisting in $In_xGa_{1-x}N$ grown by metal organic chemical vapor deposition", M. K. Behbehani, E. L. Piner, S. X. Liu, N. A. El-Masry, and S. M. Bedair, Appl. Phys. Lett. **75**, 2202 (1999).
3. "Effect of Hydrogen on the In incorporation in InGaN Epitaxial Films", E. L. Piner, M. K. Behbehani, N. A. El-Masry, F. G. McIntosh, J. C. Roberts, and S. M. Bedair, Appl. Phys. Lett. **70**, 461 (1997).
4. "Impurity Dependence on Hydrogen and Ammonia Flow Rates in InGaN Bulk Films Grown by Metalorganic Chemical Vapor Deposition", E. L. Piner, M. K. Behbehani, N. A. El-Masry, J. C. Roberts, F. G. McIntosh, and S. M. Bedair, Appl. Phys. Lett. **71**, 2023 (1997).

5. "Growth and Properties of InGaN and AlInGaN Thin Films on (0001) Sapphire", E. L. Piner, F. G. McIntosh, J. C. Roberts, M. E. Aumer, V. A. Joshkin, S. M. Bedair, and N. A. El-Masry, MRS Internet J. Nitride Semicond. Res. **1**, 43 (1996).
6. "New Buffer Layers for GaN on Sapphire by Atomic Layer Epitaxy and Molecular Stream Epitaxy", E. L. Piner, Y. W. He, K. S. Boutros, F. G. McIntosh, J. C. Roberts, S. M. Bedair, and N. A. El-Masry, Mater. Res. Soc. Symp. Proc. **395**, 307 (1996).
7. "Growth and Characterization of In-based Nitride Compounds", S. M. Bedair, F. G. McIntosh, J. C. Roberts, E. L. Piner, K. S. Boutros, and N. A. El-Masry, J. Crystal Growth **178**, 32 (1997).
8. "Optical Memory Effect in GaN Epitaxial Films", V. A. Joshkin, J. C. Roberts, F. G. McIntosh, E. L. Piner, and M. K. Behbehani, Appl. Phys. Lett. **71**, 234 (1997).
9. "Epitaxial Deposition of GaInN and InN Using the Rotating Susceptor ALE System", F. G. McIntosh, E. L. Piner, J. C. Roberts, M. K. Behbehani, M. E. Aumer, N. A. El-Masry, and S. M. Bedair, Appl. Surf. Sci. **112**, 98 (1997).
10. "Growth and Characterization of AlInGaN/InGaN Heterostructures", J. C. Roberts, F. G. McIntosh, M. Aumer, V. Joshkin, K. S. Boutros, E. L. Piner, Y. W. He, N. A. El-Masry, and S. M. Bedair, Mater. Res. Soc. Symp. Proc. **423**, 341 (1996).
11. "Optical Transitions in InGaN/AlGaN Single Quantum Wells:, K. C. Zeng, M. Smith, J. Y. Lin, H. X. Jiang, J. C. Roberts, E. L. Piner, F. G. McIntosh, S. M. Bedair, J. Zavada, J. Vac. Sci. Technol. B **15**, 1139 (1997).
12. "Stacked InGaN/AlGaN Double Heterostructures", J. C. Roberts, F. G. McIntosh, M. E. Aumer, E. L. Piner, V. A. Joshkin, S. Liu, N. A. El-Masry, and S. M. Bediar, Mater. Res. Soc. Symp. Proc. **449**, 1161 (1997).
13. "Growth and Characterization of AlInGaN Quaternary Alloys", F. G. McIntosh, K. S. Boutros, J. C. Roberts, S. M. Bedair, E. L. Piner, and N. A. El-Masry, Appl. Phys. Lett. **68**, 40 (1996).
14. "Low Temperature Growth of High Quality $In_xGa_{1-x}N$ by Atomic Layer Epitaxy", K. S. Boutros, J. C. Roberts, F. G. McIntosh, E. L. Piner, N. A. El-Masry, and S. M. Bedair, Mater. Res. Soc. Symp. Proc. **395**, 213 (1996).
15. "Growth and Characterization of In-Based Nitride Compounds and their Double Heterostructures", V. A. Joshkin, J. C. Roberts, E. L. Piner, M. K. Behbehani, F. G. McIntosh, L. Wang, S. Lin, I. Shmagin, S. Krishnankutty, R. M. Kolbas, N. A. El-Masry, and S. M. Bedair, Mater. Res. Soc. Symp. Proc. **468**, 13 (1997).
16. "Growth of High Quality InGaN Films by Metalorganic Chemical Vapor Deposition", J. C. Roberts, F. G. McIntosh, K. S. Boutros, S. M. Bedair, M. Moussa, E. L. Piner, Y. He, and N. A. El-Masry, Mater. Res. Soc. Symp. Proc. **395**, 273 (1996).

17. "AlGaInN Quaternary Alloy by MOCVD", F. G. McIntosh, E. L. Piner, K. S. Boutros, J. C. Roberts, Y. He, M. Moussa, N. A. El-Masry, and S. M. Bedair, Mater. Res. Soc. Symp. Proc. **395**, 219 (1996).

IV. Below is a list of all participating scientific personnel showing any advanced degrees earned by them while employed on the project:

1. K. S. Boutros - degree earned: Ph.D.
2. E. L. Piner - degree earned: Ph.D.

V. Below is a report of inventions that arose out of the funded research:

1. ***Stacked Quantum Well Aluminum Indium Gallium Nitride Light Emitting Diodes***, U. S. Patent 5,684,309, issued November 4, 1997.
2. ***Methods of Forming Indium Gallium Nitride or Aluminum Indium Gallium Nitride Using Controlled Hydrogen Gas Flows***, U. S. Patent 5,851,905, issued December 22, 1998