Progress report (Oct. 2005 – October 2008)

We have investigated the fundamental material properties of AlN and high Al-content AlGaN alloys and further developed MOCVD growth technologies for obtaining these materials with improved crystalline quality and conductivities. We highlight a few examples of our studies below:

1. Determining the temperature and compositional dependence of the energy bandgap of AlGaN Alloys

AlGaN alloys are very attractive for the fabrication of ultraviolet (UV) and deep UV emitters, detectors, and other optoelectronic devices due to the capability of tuning the direct bandgap in a large energy range (3.4 to 6.1 eV). They have high mechanical hardness, high thermal conductivity, large dielectric constant, and high resistance to harsh environment. For device applications based on AlGaN alloys, improving the material quality and understanding of the fundamental properties are essential. Many properties of $Al_xGa_{1-x}N$ alloys are unknown due to the increased difficulties to grow high quality materials and decreased emission efficiency with increasing x. Among the basic properties and parameters of $Al_xGa_{1-x}N$ alloys, the determination of the bandgap and its variation with temperature and composition are fundamentally important to the design of practical devices based on these materials.

During this funding period, we have employed deep-ultraviolet PL spectroscopy to study the temperature and compositional dependence of the bandgap of $Al_xGa_{1-x}N$ alloys grown on sapphire in the temperature range between 10 to 800 K. Band-edge emission peaks in $Al_xGa_{1-x}N$ alloys were fitted by Varshni equation to obtain Varshni coefficients, which increase nonlinearly with x. Based on the experimental data, we have determined empirically the compositional and temperature dependence of the bangap of $Al_xGa_{1-x}N$ alloy for the entire alloy range.

2. Probing exciton localization in AlGaN alloys

Another important property, which affects the optical and electrical properties of AlGaN alloy, is the carrier and exciton localization. Exciton localization energy and PL emission linewidth yield the information about the compositional and potential fluctuations occurring in semiconductor alloys. During the supporting period, we have studied the exciton localization effect in $Al_xGa_{1-x}N$ alloys for the entire composition range, $0 \le x \le 1$. Our experimental results demonstrated that the localized exitons in AlGaN alloys have the largest localization energies compared to all other semiconductor alloys. We have established three parallel methods for directly measuring the exciton localization energies (E_{Loc}) in AlGaN alloys and confirmed that E_{Loc} can be obtained by measuring either the deviation of the exciton emission peak energy from the Varshni equation at low temperatures or the thermal activation energy of the exciton emission intensity or the exciton emission linewidth. The exciton localization energy in $Al_xGa_{1-x}N$ alloys was observed to increase with x and reach a maximum for x ~ 0.7, implying that the potential fluctuation caused by alloy disorder is also a maximum at that value of x, consistent with the theoretical calculation result assuming completely random alloys. Exciton localization is prominent in wide-gap AlGaN alloys due to their small Bohr radius and a large difference in energy band gaps between GaN and AlN. This large exciton localization may give rise to increased quantum efficiency due to reduction of nonradiative recombination rate under the influence of the carrier localization effect. However, carrier localization will reduce significantly the conductivity of AlGaN alloys, particularly for Al content around 70%. Thus, strong carrier and exciton localization can have a significant effect on the optical and electrical properties of deep UV optoelectronic devices.

3. Identification of cation vacancies and n-type conductivity control in AlGaN alloys

In the past, AlGaN alloys with high Al contents and pure AlN were known as excellent insulators due to their large energy bandgap up to 6.1 eV. It has been very difficult to convert them to semiconductors due to the large activation energies of dopants and the simultaneous generation of free electron traps during crystal growth. We have utilized DUV picosecond time-resolved PL spectroscopy as an effective approach to identify the presence of compensating centers for n-type doping for materials grown under different conditions. We have identified three types of Al vacancies and their complexes that are free electron traps in AlGaN alloys with high Al contents and pure AlN. By monitoring/minimizing the Al vacancy related emission intensity, optimal growth conditions and layer structures for obtaining highly conductive n-type AlGaN alloys were obtained. We have achieved record high room temperature n-type conductivity for $Al_xGa_{1-x}N$ alloys with high x. Furthermore, guided by PL and Hall effect measurements, we have established the fact that it is possible to obtained n-type conduction in pure AlN with Si doping.

4. Growth and photoluminescence studies of Zn-doped AlN epilayers

While n-type AlN epilayers with reasonable conductivities have been achieved by Si-doping, p-type conductivity is extremely difficult to obtain due to the large activation energy of Mg acceptors (0.51 eV). It would be desirable to identify an alternative acceptor that may have a lower ionization energy. A previous calculation by F. Mireles and S. E. Ulloa [Phys. Rev. B **58**, 3879 (1998)] predicted that Zn occupies Al site in AlN and the activation energy of Zn acceptor in AlN is in the range of 0.22 – 0.44 eV, which is significantly smaller than that of Mg in AlN. Since the free hole concentration increases exponentially with a decrease of the acceptor activation energy, any strategies that have the potential to reduce the activation energies of acceptors in AlN are worth pursuing. During this funding period, we have carried out the growth and photoluminescence studies of Zn-doped AlN epilayers. By comparing the PL spectra of Zn-doped, Mg-doped and undoped AlN epilayers, the energy level of Zn acceptor in AlN was deduced to be about 0.74 eV, which is about 0.23 eV deeper than Mg level in AlN. In contrary to the previous theoretical prediction, our results thus suggest that Zn is not a better candidate than Mg as a p-type dopant in AlN. More theoretical and experimental investigations are required to further understand doping issues in AlN, particularly pertaining p-type doping.

5. High crystalline quality AlN epilayer growth technology development

AlN has the widest direct band gap among the III-nitrides and possesses outstanding properties such as high temperature stability, high thermal conductivity, and DUV transparency. These properties make it a good candidate for high power/temperature electronic and optoelectronic device applications such as UV emitters and detectors active in the spectral wavelength region down to 200 nm. AlN epilayers are commonly grown on foreign substrates, such as sapphire and SiC, since high quality AlN bulk wafers are not readily available. Like GaN, AlN thin films grown on sapphires are plagued by a high threading dislocation (TD) density on the order of ~ 10^9-10^{10} cm^{-2}. It is well documented that the presence of high TD density in III-nitrides is a major obstacle for the realization of high performance devices. Thus, approaches for obtaining low defect AlN epilayers as well as effective methods to probe the TD density in AlN need to be sought. During the reporting period, we have carried out the growth and systematic characterization of the optoelectronic and structural properties of AlN epilayers through the measurements of XRD and PL.

The results revealed that the TD density, in particular the edge TD density, decreases considerably with increasing the epilayer thickness. XRD rocking curves of the (002) and (102) reflections of a 4 μm epilayer show FWHMs as small as 63 and 437 arcsec, respectively. These are among the smallest values reported for AlN epilayers and are even smaller than those of the best GaN epilayers grown on sapphire (GaN (002) reflection peak has a FWHM of about 150 arcsec).

From the tilt (out-of plane rotation) and twist (in-plane rotation) spread caused by the mosaicity of the AlN film, the dislocation density was estimated. The screw dislocation density was ~$5x10^6$ cm^{-2} in the 4 μm thick AlN epilaye and is more than one order of magnitude lower than that in GaN of the same thickness (~10^8 cm^{-2}). This clearly indicates that AlN epilayer is an effective dislocation filter. This reduction in screw dislocation density is particularly important for vertical optoelectronic devices such as light emitting devices and detectors because screw dislocations are one of the major sources of current leakage paths, which increase with increasing current density. Screw dislocations also behave as non-radiative recombination centers that reduce the output intensity from optical devices.

6. Fundamental studies of Mg-doped AlGaN alloys and AlN

For applications using AlN or AlGaN with an extremely high Al content as an active material, highly conductive n-type and p-type AlN materials are desirable. However, it is very difficult to improve the conductivity of AlN due to the large activation energy of dopants and the simultaneous generation of compensating centers and defects during the crystal growth. During this funding period, we have carried out epitaxial growth by MOCVD and studies of the correlation between the electrical and optical properties of Mg-doped AlN epilayers. Besides the large activation energy of the Mg acceptors in AlN, we have experimentally observed that compensation of free holes by “donor-like” intrinsic defects (nitrogen vacancies) is another cause for the highly resistive nature of Mg-doped high Al-content AlGaN alloys and AlN. By monitoring the ratio of the native vacancy related emission to the band edge emission intensity, we have obtained Mg-doped AlN with measureable p-type conductivity at elevated temperatures (T > 600 K). This improvement allows us for the first time to determine the Mg acceptor activation energy by conventional Hall-effect measurements. From the variable temperature Hall-effect measurements performed at elevated temperatures (up to 850 K), the activation energy of Mg in AlN was measured to be about 0.51 eV, which is consistent with the value obtained from our previous optical measurements.

II. Publications, invited talks, etc during the supporting period (acknowledged DOE support)

Publications during the funding period (10/2005 - current)

1. N. Nepal, J. Li, M. L. Nakarmi, J. Y. Lin, and H. X. Jiang, “Temperature and compositional dependence of the energy band gap of AlGaN alloys,” Appl. Phys. Lett. 87, 242104 (2005).
2. N. Nepal, J. Li, M. L. Nakarmi J. Y. Lin, and H. X. Jiang, “Exciton localization in AlGaN alloys,” Appl. Phys. Lett. 88, 062103 (2006).
3. M. L. Nakarmi, N. Nepal, C. Ugolini, T. M. Altahtamouni, J. Y. Lin, and H. X. Jiang, “Correlation between optical and electrical properties of Mg doped AlN epilayers,” Appl. Phys. Lett. 89, 152120 (2006).
4. Z. Y. Fan, J. Y. Lin, and H. X. Jiang, “III-nitride deep ultraviolet micro- and nano-photonics,” Proc. SPIE 6127, 61271C (2006).
5. N. Nepal, M. L. Nakarmi, H. U. Jang, J. Y. Lin, and H. X. Jiang, “Growth and photoluminescence studies of Zn-doped AlN epilayers,” Appl. Phys. Lett. 89, 192111 (2006).
6. X. H. Ji, S. P. Lau, S. F. Yu, H. Y. Yang, T. S. Herng, A. Sedhain, J. Y. Lin, H. X. Jiang, K. S. Teng, and J. S. Chen, "Ultraviolet photoluminescence from ferromagnetic Fe-doped AlN nanorods," Appl. Phys. Lett. 90, 193118 (2007).
7. F. Wang, S. S. Li, J. B. Xia, H. X. Jiang, J. Y. Lin, J. Li, and S. H. Wei, “Effects of the wavefunction localization in $Al_xIn_yGa_{1-x-y}N$ quaternary alloys,” Appl. Phys. Lett. 91, 061125 (2007).
8. Z. Y. Fan, J. Y. Lin, and H. X. Jiang, “Achieving conductive high Al-content AlGaN alloys for

deep UV photonics," Proc. SPIE 6479, 64791I (2007).

9. B. N. Pantha, N. Nepal, T. M. Al Tahtamouni, M. L. Nakarmi, J. Li, J. Y. Lin, and H. X. Jiang, "Correlation between biaxial stress and free exciton transition in AlN epilayers," Appl. Phys. Lett. 91, 121117 (2007).

10. B. N. Pantha, R. Dahal, J. Li, J. Y. Lin, H. X. Jiang, and G. Pomrenke, "Thermoelectric properties of $In_xGa_{1-x}N$ alloys," Appl. Phys. Lett. 92, 042112 (2008).

11. A. Sedhain, J. Y. Lin, and H. X. Jiang, "Valence band structure of AlN probed by photoluminescence," Appl. Phys. Lett. 92, 041114 (2008).

12. A. BenMoussa, J. F. Hochedez, R. Dahal, J. Li, J. Y. Lin, H. X. Jiang, A. Soltani and J.-C. De Jaeger, "Characterization of AlN metal-semiconductor-metal diodes in the spectral range of 44–360 nm: Photoemission assessments," Appl. Phys. Lett. 92, 022108 (2008).

13. T. M. Al tahtamouni, A. Sedhain, J. Y. Lin, and H. X. Jiang, "Si-doped high Al-content AlGaN epilayers with improved quality and conductivity using indium as a surfactant," Appl. Phys. Lett, 92, 092105 (2008).

14. T. M. Al tahtamouni, A. Sedhain, J. Y. Lin, and H. X. Jiang, "Growth and optical properties of a-plane AlN and quantum wells grown on r-plane sapphire substrates," phys. status solidi (c) 5, 1568 (2008).

Invited reviews and book chapters

15. Z. Y. Fan, J. Y. Lin, and H. X. Jiang, "III-nitride micro-emitter arrays: development and applications," Special Issue, J. Phys. D: Appl. Phys. 41 094001 (2008).

16. H. X. Jiang and J. Y. Lin, "III-Nitride Micro-Cavity Light-Emitters," – in *"Wide Bandgap Light-Emitting Materials and Devices,"* edited by G.F. Neumark, I. Kuskovsky, and H. X. Jiang, published by Wiley –VCH Verlag GmbH, 2007.

Meetings organized (Chair/co-chair or program committee member)

1. 2006 Fall Meeting, Material Research Society, Symposium I: Advances in III-V Nitride Semiconductor Materials and Devices.
2. National Organizing Committee – 14th Semiconducting and Insulating Materials Conference, University of Arkansas, May 2007.
3. Editorial Board Member - International Journal of Materials Sciences, (2004 – current).
4. Second International Symposium on Growth of III-Nitrides, Laforet Shuzenji, Izu, Japan, July 6-9, 2008.
5. Summer School 2008 on Wide-bandgap Semiconductor Physics and Devices, July 28 - August 3, 2008 – Dalian, China.

Books edited

- "Advances in III-V Nitride Semiconductor Materials and Devices," MRS Fall 2006 Meeting Proceedings Vol. 955E, Edited by C.R. Abernathy, H. Jiang, J.M. Zavada.
- *"Wide Bandgap Light-Emitting Materials and Devices,"* edited by G.F. Neumark, I. Kuskovsky, and H. X. Jiang, published by Wiley –VCH Verlag GmbH, 2007.

Tutorials/short courses offered

1. "Processes and Devices of GaN Materials on Si," H. X. Jiang, Spring MRS meeting 2008 Tutorial, San Francisco.
2. "III-Nitride deep UV photonics," H. X. Jiang & J. Y. Lin, Short Course, SPIE - Photonic West Meeting, San Jose Jan. 2006.

Invited talks

1. "III-nitride photonic crystals," H. X. Jiang, University of Illinois, Urbana-Champing, April 2008.

2. "Recent Development and Applications AC-LEDs," H. X. Jiang, 2007 International workshop on new materials research and industrialization," Wenzhou, China, December 2007.
3. "Progress and challenges of p-type doping in III-nitrides," H. X. Jiang, III-Nitride Workshop, Richmond, VA, Oct. 2007.
4. "III-nitride deep UV nano-photonics," H. X. Jiang, the 54th Midwest Solid State Conference, University of Nebraska – Lincoln, Oct. 2007.
5. "III-Nitride UV Photonics," H. X. Jiang, 3rd Asian Pacific Workshop on Wide Gap Semiconductors, March 11-14, 2007, Jeonju, Korea.
6. "Achieving highly conductive Al-rich AlGaN alloys for deep UV photonics," H. X. Jiang Photonic, West Meeting, San Jose, Jan. 2007.
7. "Er-dopedGaN synthesized by MOCVD," IBEDM, Japan, Nov. 2006.
8. "Nitride Deep UV Photonics," H. X. Jiang, H. X. Jiang, the 53rd Midwest Solid State Conference, University of Missouri, Kansas City, Oct. 2006.
9. "III-Nitride Wide Bandgap Semiconductors for Optical Communications," H. X. Jiang, Lasers & Electro-Optics Society, LEOS 2006 Annual Meeting, Montreal, Canada, Oct 2006.
10. "Conductive high Al-content AlGaN alloys for deep UV photonics," 2006 Lester Eastman Conference on High Performance Devices, Cornell University, August 2006.
11. "Fundamental doping issues in Al-rich AlGaN alloys for deep UV photonics," H. X. Jiang, OIDA (Optoelectronics Industry Development Association) Roadmap Forum on Nitride LED and Laser Technology, Palo Alto, CA, May 2006.
12. "III-nitride deep ultraviolet micro- and nano-photonics," H. X. Jiang, Delivered in the Symposium on Quantum Sensing: Evolution and Revolution from Past to Future, SPIE - Photonic West Meeting, San Jose, Jan. 2006.
13. Nitride Photonic Crystals," H. X. Jiang, Meijo International Symposium on Nitride Semiconductors, Meijo University, Japan, Dec. 2005.
14. "Si and Mg-doped Al-rich AlGaN alloys for deep UV photonics," H. X. Jiang, 5th Akasaki Research Center Symposium, Dec., Nagoya, Japan, Dec. 2005.
15. "III-Nitride Photonic Crystals and AC LEDs," H. X. Jiang, 2005 International Forum on LED & Solid-State Lighting, Xiamen China.
16. "III-Nitride Photonic Crystals," H. X. Jiang, SPIE Symposium on Integrated Opto electronic Devices 2005 Conference on Light-Emitting Diodes – Research, Manufacturing, and Applications IX, San Jose, CA 2005.
17. "III-nitride deep UV photonics," H. X. Jiang, presented in Institute of Electron Technology, Warsaw, Poland, Aug. 2005.
18. "Recent advances in III-Nitride UV photonics," H. X. Jiang, NASA Goddard Space Flight Center, May 2005.
19. "Recent advances in III-nitride micro- and nano-photonics," H. X. Jiang, Navel Research Lab., May 2005.
20. "III-Nitride UV Photonics," J. Y. Lin, 2007 International workshop on new materials research and industrialization, Wenzhou, China, December 2007.
21. "Nitride Photonic Crystals," J. Y. Lin, Eighth International Symposium on Contemporary Photonics Technology, Tokyo, Japan (January 2005).
22. "III-Nitride Ultraviolet Micro- and Nano-Photonics," J. Y. Lin, 2005 Conference on Lasers and Electro-Optics Quantum Electronics & Laser Science Conference, Baltimore, MD (May, 2005).