Appl. Phys. A 86, 521–524 (2007)
DOI: 10.1007/s00339-006-3804-9

Applied Physics A
Materials Science & Processing

S.J. CHUNG[1,✉]
B. KARUNAGARAN[1]
S. VELUMANI[2]
C.-H. HONG[1]
H.J. LEE[1]
E.-K. SUH[1]

Photoluminescence and persistent photoconductivity of $Al_xGa_{1-x}N/GaN$ heterostructures

[1] Semiconductor Physics Research Center and Department of Semiconductor Science and Technology, Chonbuk National University, Chonju 561-756, Korea
[2] Departamento de Física, TEC de Monterrey – Campus Monterrey, E. Garza-Sada 2501, Monterrey, N.L., C.P. 64849, México

Received: 2 November 2006/Accepted: 21 November 2006
Published online: 23 December 2006 •

ABSTRACT We have investigated the optical properties of $Al_xGa_{1-x}N/GaN$ heterostructures ($x = 0.08$, 0.15, 0.33) grown by metal organic chemical vapor deposition on sapphire using photoluminescence (PL) and persistent photoconductivity (PPC) measurements. For the $Al_xGa_{1-x}N/GaN$ heterostructures (HS) containing high Al composition, we observed an anomalous temperature-dependent photoluminescence and persistent photoconductivity effects. These results show a strong dependence of the physical properties of $Al_xGa_{1-x}N/GaN$ HS on the Al content and layer thickness. The anomalous temperature-dependent PL is usually attributed to the presence of carrier localization states. These phenomena are explained based on the alloy compositional fluctuations in the $Al_xGa_{1-x}N/GaN$ HS. From the PPC measurements, the photocurrent (PC) quenching was observed for $Al_xGa_{1-x}N/GaN$ HS and it is explained by the metastable states formed in the underlying GaN layer. Also, the mechanisms behind the PC quenching and PPC phenomena are explained in detail.

PACS 72.20.Jv; 72.40.+w; 78.55.Cr

1 Introduction

$Al_xGa_{1-x}N$ epilayers have recently attracted much attention as promising materials for the fabrication of light-emitting diodes (LEDs) with wavelengths ranging from blue to ultraviolet (UV) and for high-power electronic devices. However, present understanding of optical properties of $Al_xGa_{1-x}N$ epilayers is still insufficient to realize efficient devices in short-wavelength optoelectronics. Previous reports on $Al_xGa_{1-x}N$ epilayers mostly dealt with photoluminescence (PL), photoluminescence excitation (PLE), photocurrent (PC), optical absorption (OA), and PL decay time explaining the effect of alloy potential fluctuations (APFs) [1–3]. It is expected that alloy fluctuations will play an important role in determining the optical and transport properties of $Al_xGa_{1-x}N$ alloys due to the large energy band gap difference between AlN and GaN. According to thermodynamical calculations, $Al_xGa_{1-x}N$ alloys are expected not to have unstable mixing regions, in contrast to $In_xGa_{1-x}N$ alloys. Accordingly, no phase separation is predicted in $Al_xGa_{1-x}N$ alloys [4]. However, in practice, $Al_xGa_{1-x}N$ alloy samples with high Al composition also show an S-shaped PL shift as observed in $In_xGa_{1-x}N$ alloys or InGaN/GaN multi-quantum wells (MQWs), and the S-shape behavior and Stokes shift have been explained in terms of the effects of localized states induced by alloy fluctuations in $Al_xGa_{1-x}N$ alloys [3, 5].

In this paper, we report optical properties of $Al_xGa_{1-x}N/GaN$ heterostructures (HS) investigated by using PL and persistent photoconductivity (PPC) measurements. The temperature-dependent PL peak energy shift exhibits S-shape behavior and the decay time of PPC spectra strongly depends on Al content and layer thickness. These S-shape peak shifts are usually attributed to the presence of carrier localization states and are explained based on the alloy compositional fluctuations in the $Al_xGa_{1-x}N$ alloys. In PPC measurements, PC quenching phenomena depending on layer thickness are explained by the metastable states in the GaN interface layer.

2 Experiment

In order to study the optical properties of these alloys, $Al_xGa_{1-x}N/GaN$ heterostructures (HS) having Al composition ranging from 0 to 0.33 were grown by metal organic chemical vapor deposition (MOCVD) on sapphire. A 2.0-μm GaN epilayer was first deposited on the 25-nm low-temperature-grown GaN nucleation layer grown on a c-plane sapphire substrate, followed by an $Al_xGa_{1-x}N$ epilayer whose thickness was varied from 0.2 to 1 μm. The growth temperature and pressure for the $Al_xGa_{1-x}N$ epilayer were 1080 °C and 300 Torr, respectively. The Al content was determined by X-ray diffraction (XRD) using Cu $K_{\alpha 1}$ radiation. The PL experiments were performed using a 325-nm He–Cd laser and a 244-nm double cw Ar^+ laser. For the persistent photoconductivity (PPC) measurements, samples were cut into $3 \times 3\ mm^2$ and two coplanar electric contacts with 1-mm spacing were formed with indium solder. The ohmic nature of the contacts was confirmed by I–V characteristics. A detailed description of our PPC measurement system is reported elsewhere [6].

3 Results and discussion

The PL peak energy positions of the $Al_xGa_{1-x}N$ epilayers for compositions $x = 0.08$, 0.15, and 0.33 are plot-

✉ Fax: +82-63-270-3585, E-mail: chungsj40@chonbuk.ac.kr

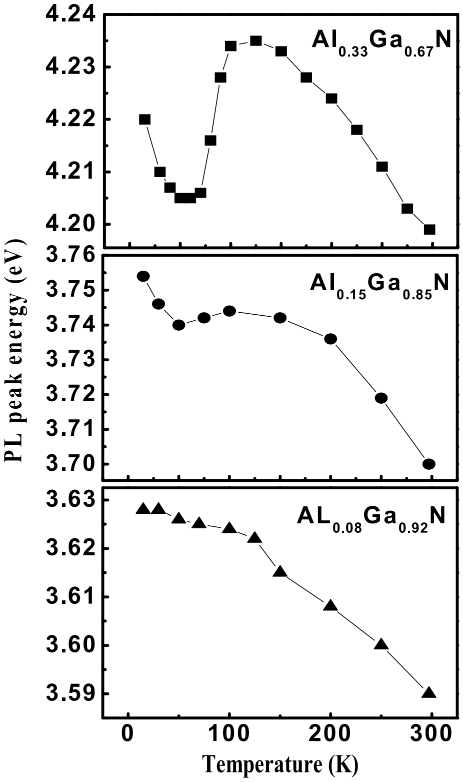

FIGURE 1 Temperature-dependent PL spectra of $Al_xGa_{1-x}N$ epilayers with various Al compositions

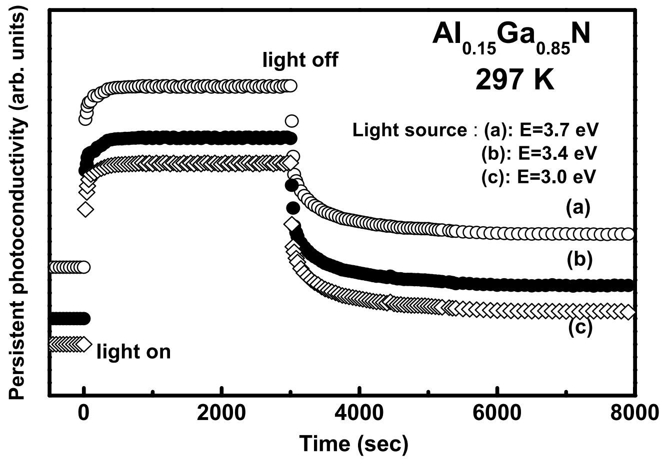

FIGURE 2 RT-PPC spectra of $Al_{0.15}Ga_{0.85}N$/GaN HS with thick AlGaN layers (> 0.5 μm) as a function of incident-photon energy

ted as a function of temperature in Fig. 1. These PL data were taken from our previous work [3]. The PL peak energy for the $Al_{0.08}Ga_{0.92}N$ epilayer follows the typical temperature dependence of the energy band gap shrinkage described by Varshni's equation. But, the PL emission spectra for the $Al_{0.33}Ga_{0.67}N$ epilayer show the 'S-shaped' emission peak shift, i.e. decrease–increase–decrease, with increasing measurement temperature. The increase of Al content clearly shows the aggravation of the S-shaped behavior of PL peak energy. Similar behavior has been reported previously for the temperature-dependent PL emission energy shift in $In_xGa_{1-x}N$, InGaN/GaN MQWs [7, 8], $Al_xGa_{1-x}N$, and pseudomorphic AlGaN/GaN HS [2, 3, 9, 10]. According to Lin et al. [11], the recombination of an electron–hole (e–h) pair is considered to occur around the localized states. The localized e–h pair recombination increases with Al content in the $Al_xGa_{1-x}N$ epilayers due to strong alloy potential fluctuations. At low temperatures, carriers can recombine at energy states of local potential minima. As the temperature slightly increases, weakly localized carriers are thermally excited and would either recombine nonradiatively or be redistributed to other strongly localized states. Thus, the PL peak energy decreases with increasing temperature. After the effect of redistribution is saturated, the thermal energy can excite carriers to higher localized states, and hence the PL peak energy again increases. As the temperature increases further, a red shift occurs due to ordinary band gap shrinkage. Cho et al. [1] explained that the anomalous temperature-induced emission peak shift is deeply related to the thermal population in localized energy tail states due to alloy potential inhomogeneities in the $Al_xGa_{1-x}N$ epilayers. A similar pattern of temperature dependence for PL peak energy has been reported in ordered $GaInP_2$ and in disordered superlattices consisting of $(AlAs)_m(GaAs)_n$ with m and n randomly chosen, in contrast to the case of random homogeneous direct gap III–V alloys or ordered AlAs/GaAs superlattices [1, 12, 13].

Additional evidence for the existence of localized states due to alloy potential fluctuations is observed in the measurement of PPC. Figure 2 shows PPC spectra of $Al_{0.15}Ga_{0.85}N$/GaN HS with thick AlGaN layers (> 0.5 μm) as a function of excitation photon energy at room temperature (RT). The origin of the PPC can be explained in that the photoexcited carriers are trapped and spatially separated by local potential fluctuations, which suppresses the recombination of carriers. With decreasing Al content, the PPC decay becomes faster. The carrier transport process can be used for interpreting the slow decay in samples with high Al compositions. This slow decay is essentially due to the carrier capture process from weakly localized states into strongly localized states of deeper potential by alloy fluctuations. Moreover, the stretched exponential relaxation is commonly observed in disordered samples. According to the previous report [14], the density of states is exponentially distributed in terms of energy, such as the band tail states in disordered semiconductors. Therefore, the stretched exponential decay reveals similarities of the present system with disordered samples.

In order to obtain the degree of alloy potential fluctuations in $Al_{0.15}Ga_{0.85}N$/GaN HS, we performed the temperature-dependent PPC experiments and reported our observations elsewhere [3]. Figure 3 displays the Stokes shift and depth of alloy potential fluctuations as a function of Al composition that is obtained by temperature-dependent PPC; the experimental details of the PPC measurement have been reported in our previous work [3]. The results clearly indicate the existence of localized states in Al–Ga–N alloy systems, especially with high Al contents.

Figure 4 presents room-temperature PPC spectra of $Al_{0.15}Ga_{0.85}N$/GaN HS with AlGaN layers thinner than 0.5 μm, as a function of incident-photon energy. As shown in Fig. 4, however, the PPC phenomenon observed in our sam-

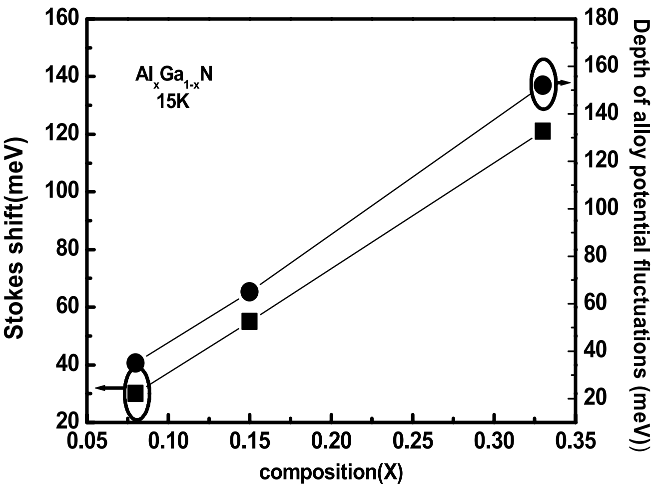

FIGURE 3 The Stokes shift and the average depth of APFs as a function of Al composition

ple indicates that PC quenching occurs after a certain amount of time, despite the continuous light irradiation. When the light is switched off, a negative PC signal arises, followed by slow recovery to the dark-current level. These observations are quite different from the PPC of n-GaN, p-GaN, and $Al_xGa_{1-x}N$ epilayers previously reported [15–17].

When light corresponding to the energy band gap of an $Al_{0.15}Ga_{0.85}N$ epilayer is irradiated, PC quenching occurs, but only down to the dark-current level without negative PC phenomena, as shown in Fig. 4a. However, as shown in Fig. 4b and c, the PC quenches below dark current so as to allow negative current, even under continuous light irradiation, if the photon energy is lower than the band-gap energy. It is also observed that the PC quenches more severely with lower incident-photon energy. This may be explained as follows: when the energy of the incident light decreases below the band-gap energy of the AlGaN layer, i.e. < 3.7 eV, defects such as dislocations in the GaN layer influence the PPC spectra since the penetration depth increases with decreasing incident-photon energy and probes the GaN epilayer. A similar phenomenon has been reported in n-type GaN unintentionally doped with impurities [18], InSb/GaAs epilayers [19], and Sn modulation doped $Al_xGa_{1-x}As$/GaAs ($x > 35\%$) HS [20]. No photocurrent quenching or negative dark current phenomenon has been found in AlGaN layers of thickness larger than 0.5 μm as shown in Fig. 2, which agrees well with the previous reports [6, 18]. On the other hand, similar kinds of results have been obtained for thin AlGaN layers (< 0.5 μm) grown on more-than-2-μm-thick GaN layers, as well as for more-than-0.5-μm-thick $Al_xGa_{1-x}N$ layers grown on thin GaN layers of less than 2 μm. This observation is very similar to previous results on undoped n-GaN epilayers [6, 18] and implies that the PPC of AlGaN/GaN HS contains a certain degree of influence from both GaN and AlGaN layer thicknesses.

In order to confirm the above observation, PPC of an $Al_xGa_{1-x}N$ epilayer grown on an AlN buffer layer was measured and the outcome was exactly a typical PPC without an anomalous behavior, as shown in Fig. 2. Therefore, it can be concluded that the anomalous PPC observed in an $Al_xGa_{1-x}N$ layer of less than 0.2 μm is caused purely due to the effect of the metastable state formed in the GaN underlayer [18].

To understand the metastable state in the GaN layer, the PPC spectrum was taken with applied thermal energy at the moment of PC quenching and is given in Fig. 5. It is obvious that the PC quenching could be recovered immediately with direct heating of the sample. In general, ordinary white light is believed to be insufficient to transit back the bound electrons in the metastable state into the conduction band, due to the wide band gap nature of GaN. As a consequence, even though the electrons captured by the metastable state can be thermally recovered when the potential barrier is not too high, they cannot induce any optical transition, due to their low energy. Therefore, the electrons captured at the metastable state which cause PC quenching are recovered back to the conduction band due to applied thermal energy by direct heating. The increased amount of recovered electrons in the conduction band gradually restores the PC. In addition, thermally excited electrons can also contribute to enhance the PC. This phenomenon is quite similar to the behavior of the EL2 metastable state in GaAs, where the PC is completely quenched under the applied

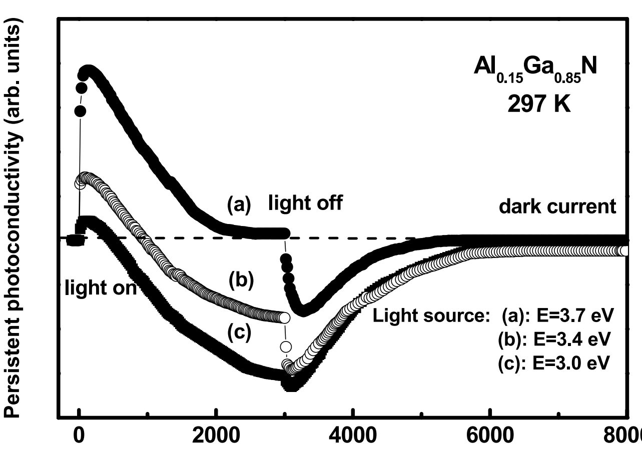

FIGURE 4 RT-PPC spectra of $Al_{0.15}Ga_{0.85}N$/GaN HS with thin AlGaN layers (< 0.5 μm) as a function of incident-photon energy

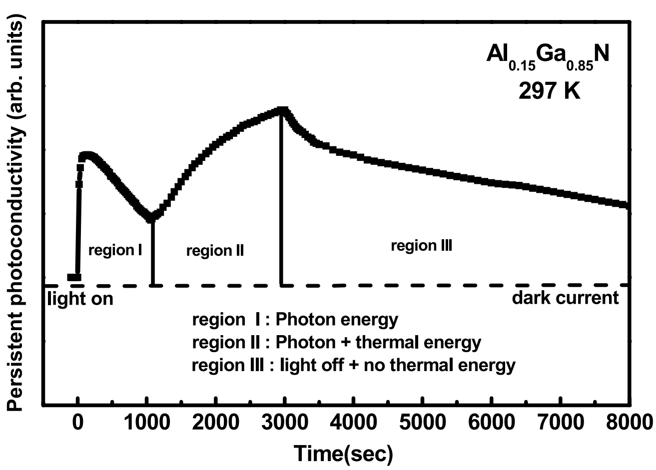

FIGURE 5 RT-PPC spectrum of $Al_{0.15}Ga_{0.85}N$ HS measured with additional thermal energy at the moment of PC quenching

light energy of 1.1 eV at a temperature below 100 K, but it is, however, recovered by thermal energy at a temperature above 100 K [19–22].

The mechanism of PC quenching and the anomalous PPC phenomenon associated with negative PC can be explained as follows. The photogenerated conduction electrons contributing to PC are gradually transferred to metastable states, which results in PC quenching. When the light is off, the remaining electrons in the conduction band recombine with holes in the valence band and a negative PC flows due to excess holes in the valence band. The recovery of negative PC to the dark-current state can be explained in that the electrons in a metastable state gradually return to the conduction band over the potential barrier and recombine with the excess holes in the valence band.

4 Summary

$Al_xGa_{1-x}N/GaN$ heterostructures (HS) were grown by MOCVD and the optical properties of the $Al_xGa_{1-x}N/GaN$ HS were studied using PL and PPC measurements. In PL measurements, the increase of Al content clearly shows the aggravation of the S-shaped behavior of PL peak energy. These behaviors are usually attributed to the presence of carrier localization states and are explained based on the alloy compositional fluctuations in the $Al_xGa_{1-x}N$ alloys. In PPC measurements, there was PC quenching despite continuous light irradiation and an anomalous PPC phenomenon accompanied by negative PC was observed. The PC drastically decreases after light interruption and is gradually recovered back to the dark-current value. It also shows a strong dependence on the energy of the incident photon. Such a phenomenon has been shown to depend on the thickness of either the AlGaN or the GaN layer, suggesting that the metastable states formed in either the GaN or the AlGaN interface layer play a major role.

ACKNOWLEDGEMENTS This work was supported by a Korea Research Foundation grant funded by the Korean Government (MOEHRD) (KRF-2005-005-J07501).

REFERENCES

1 Y.H. Cho, G.H. Gainer, J.B. Lam, J.J. Song, W. Yang, W. Jhe, Phys. Rev. B **61**, 7203 (2000)
2 H.S. Kim, R.A. Mair, J. Li, J.Y. Lin, H.X. Jiang, Appl. Phys. Lett. **76**, 1252 (2000)
3 S.J. Chung, M.S. Kumar, H.J. Lee, E.-K. Suh, J. Appl. Phys. **95**, 3565 (2004)
4 T. Matsuoka, MRS Internet J. Nitride Semicond. Res. **1**, 11 (1996)
5 M. Leroux, S. Dalmasso, F. Natali, S. Helin, C. Touzi, S. Laugt, M. Passerel, F. Ommes, F. Semond, J. Massies, P. Gibert, Phys. Stat. Solidi B **234**, 887 (2002)
6 S.J. Chung, M.S. Jeong, O.H. Cha, C.-H. Hong, E.-K. Suh, H.J. Lee, Y.S. Kim, B.H. Kim, Appl. Phys. Lett. **76**, 1021 (2000)
7 P. Perlin, V. Iota, B.A. Weinstein, P. Wisniewski, T. Suki, P.G. Eliseev, M. Osinski, Appl. Phys. Lett. **70**, 2993 (1997)
8 Y. Narukawa, Y. Kawakami, M. Funato, Sz. Fujita, Sg. Fujita, S. Nakamura, Appl. Phys. Lett. **70**, 981 (1997)
9 M. Leroux, S. Dalmasso, F. Natali, S. Helin, C. Touzi, S. Laugt, M. Passerel, F. Ommes, F. Semond, J. Massies, P. Gibert, Phys. Stat. Solidi B **234**, 887 (2002)
10 G. Steude, B.K. Meyer, A. Goldner, A. Hoffman, F. Bertran, J. Christen, H. Amano, I. Akasaki, Appl. Phys. Lett. **74**, 2456 (1999)
11 T.Y. Lin, J.C. Fan, Y.F. Chen, Semicond. Sci. Technol. **14**, 406 (1999)
12 A. Zunger, S. Mahajan, in *Handbook of Semiconductors*, vol. 3, ed. by S. Mahajan (Elsevier, Amsterdam, 1994)
13 B. Monemar, Phys. Rev. B **61**, 676 (1974)
14 S.W. Feng, Y.-C. Cheng, Y.-Y. Chung, C.C. Yang, Y.-S. Lin, C. Hsu, K.-J. Ma, J.-I. Chyi, J. Appl. Phys. **92**, 4441 (2002)
15 C. Johnson, J.Y. Lin, H.X. Jiang, M.A. Khan, C.J. Sun, Appl. Phys. Lett. **68**, 1808 (1996)
16 K.J. Kim, S.J. Chung, Appl. Phys. Lett. **80**, 1767 (2002)
17 C.V. Reddy, K. Balakrishnan, H. Okumura, J.M. Redwing, Appl. Phys. Lett. **73**, 244 (1998)
18 S.J. Chung, O.H. Cha, Y.S. Kim, M.S. Jeong, C.-H. Hong, H.J. Lee, M.S. Jeong, J.O. White, E.-K. Suh, J. Appl. Phys. **89**, 5454 (2001)
19 Y. Beaulieu, J.B. Webb, J.L. Brebner, Solid State Commun. **76**, 233 (1990)
20 Z. Peng, T. Saku, Y. Horikoshi, J. Appl. Phys. **79**, 3592 (1996)
21 B.H. Kim, Ph.D. thesis, Chonbuk National University, Korea (1989)
22 J.W. Kim, M.S. thesis, Kunsan National University, Korea (1991)