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Deep Defects Annihilation in $GaAs_{1-x}N_x$ Layers by Si-doping

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Abstract: The photoluminescence (PL) properties of Si-doped $GaAs_{0.985}N_{0.015}$ with different silicon content were investigated. The study was carried out on a set of three samples grown by Molecular Beam Epitaxy (MBE) on GaAs (001) oriented substrate using a radio frequency nitrogen beam source. For all samples, the PL measurements show the presence of a wide band situated at 0.83 eV which intensity decreases by increasing silicon content. This wide band was attributed to the presence of deep localized states induced by a three-dimensional growth of the GaAsN layer. In addition, these deep localized states are annihilated by the free carriers from silicon atoms. PL measurements in the range of 10 to 300 K were also performed to identify the band gap energy of $GaAs_{1-x}N_x$ structure. The decrease of the activation energies with increasing silicon content was observed.

Key words: $GaAs_{1-x}N_x$, molecular beam epitaxy, photoluminescence spectroscopy, Si-doping, deep defects

INTRODUCTION

Direct-band gap III-V compound semiconductors are widely used to produce optoelectronic devices, such as laser diodes, light emitting diodes and photodiodes[1,2]. Recently the nitrogen incorporation in III-V materials is promising the flexibility in choice of semiconductor band gap[2] due to many interesting physical properties such as a strong redshift of the band gap energy[3-7]. Silicon is the donor of choice for GaN and GaAs materials because of its low activation energy: at room temperature, nearly all of the donors are ionized[8-13]. The behavior of Si as n-type dopant in GaAs has been the subject of investigation for four decades[9,13,14]. Actually, doping of the III-V-N alloys with donors is an important yet very poorly understood issue[6,15].

In this work, we present the effect of the silicon n-doping on three samples using photoluminescence measurements (PL). Samples (a), (b) and (c) consist respectively of a $GaAs_{0.985}N_{0.015}$ active region with different silicon content: undoped, 0.28×10^{18} cm^{-3} and 2.10×10^{18} cm^{-3}. The study is based on PL measurements by varying temperature and laser power excitation.

MATERIALS AND METHODS

The studied samples were grown by Molecular Beam Epitaxy (MBE) system on (001) GaAs oriented substrate using a Riber system equiped with solid source for Ga and As elements and with an Addon radio-frequency (rf) plasma source for N. The growth temperature of GaAs substrate is around 600 °C, it's used to optimize the crystal quality. However, the temperature was decreased to 400 – 450 °C for growing $GaAs_{1-x}N_x$ layers to avoid phase separation. The samples (Table 1) are composed by a semi-insulator GaAs substrate covered by a GaAs buffer (0.1μm) and an active region (0.5μm) with different silicon content. The PL measurements were performed by using a variable temperature (10 – 300 K) close-cycle cryostat under 514.5 nm line of an Argon ion Ar+ laser as excitation source. The signal was detected through a 250 mm Jobin-Yvon monochromator and by a GaInAs photodiode associated with a standard lock-in technique.

RESULTS AND DISCUSSION

Figure 1 shows the normalized photoluminescence spectra of the three samples at low temperature under a laser power excitation of 10 Wcm^{-2}. For the undoped sample (a), the PL spectrum is essentially formed by a wide band situated at 0.83 eV and two fine structures at the high energy side. For the Si-doped samples (b) and (c) respectively with 0.28×10^{18} cm^{-3} and 2.10×10^{18} cm^{-3}, we note also the presence of the wide band and only one fine structure at high energy.

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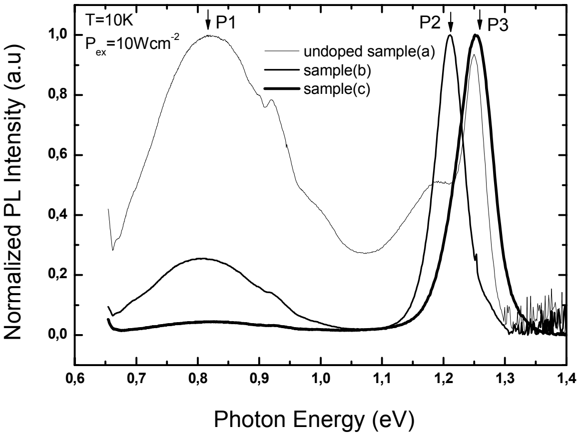

Fig. 1: 10 K normalized photoluminescence spectra of the samples (a), (b) and (c) describing the influence of silicon doping under a laser power excitation of 10Wcm^{-2}

Table 1: Si- doping content in the different $GaAs_{1-x}N_x$ (x = 1.5%) samples

Samples	Active region	Si-doping (cm^{-3})
(a)	$GaAs_{1-x}N_x$	undoped
(b)	$GaAs_{1-x}N_x$:Si	0.28×10^{18}
(c)	$GaAs_{1-x}N_x$:Si	2.10×10^{18}

Table 2: Fitting parameters using the Bose-Einstein expression (Eq. 1)

$GaAs_{1-x}N_x$ band gap	E_B (eV)	a (meV)	θ (K)
undoped sample (a)	1.27	20.5	155
sample (b), 0.28×10^{18} cm^{-3}	1.285	35	286
sample (c), 2.10×10^{18} cm^{-3}	1.3	29.5	216

Table 3: Fitting parameters using the Arrhenius model (Eq. 2) of the wide band situated at 0.83 eV

0,83 eV Wide band	E_a (meV)	a_1	a_2
undoped sample (a)	29	110^{-4}	2110^{-2}
sample (b), 0.28×10^{18} cm^{-3}	13.5	3.510^{-4}	510^{-2}
sample (c), 2.10×10^{18} cm^{-3}	10.3	1110^{-4}	0.9810^{-2}

The decrease of the wide band PL intensity is noted by increasing the silicon content, this effect is firstly attributed to the deep nitrogen localized states, but recently, we found that the same wide band was also observed by Viturro *et al.*[16] in GaAs at low growth temperature. We remind that the growth temperature of GaAsN is decreased to avoid the nitrogen evaporation, that causes a three-dimensional growth and then the formation of deep defects. The decreasing of the wide band PL intensity by increasing the silicon content as seen in the highly doped sample (c), attribute to this band a trap character for the free carriers.

In order to determine the nature of the structures at high energy, PL measurements in a range of 10-300 K were performed (Fig. 2a-2c). The analysis of the samples (a) and (b) PL spectra show that the emission peak P_2 situated at 1.21 eV disappears by increasing temperature. This behavior is related to nitrogen localized state. On the other hand, the three samples PL

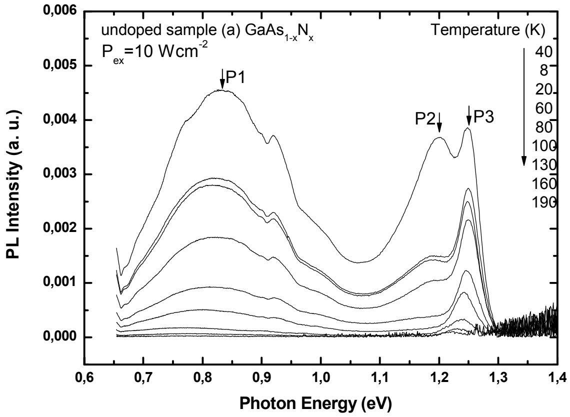

Fig. 2a: Temperature dependence of photoluminescence spectra of the $GaAs_{1-x}N_x$ (x = 1.5%) undoped sample (a) under 10Wcm^{-2}

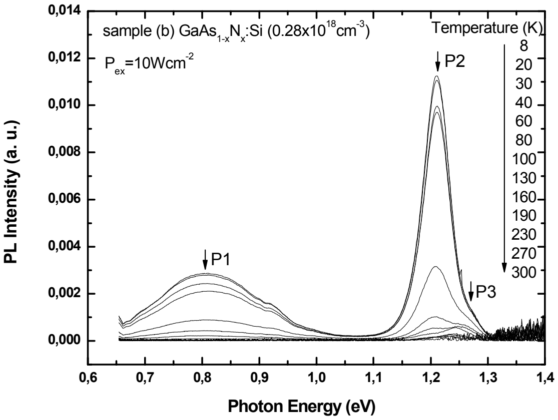

Fig. 2b: Temperature dependence of photoluminescence spectra of the Si-doped $GaAs_{1-x}N_x$ (x = 1.5%) sample (b) with 0.28×10^{18} cm^{-3} under 10Wcm^{-2}

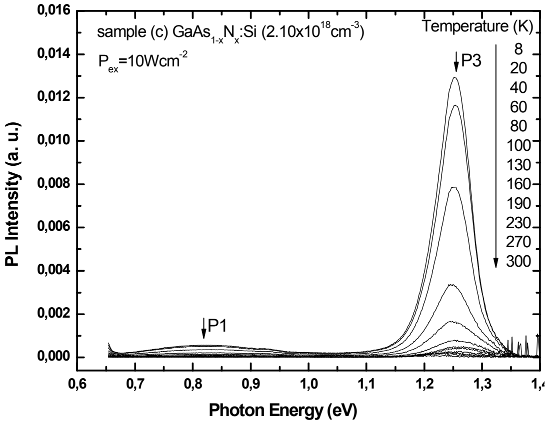

Fig. 2c: Temperature dependence of photoluminescence spectra of the Si-doped $GaAs_{1-x}N_x$ (x = 1.5%) sample (c) with 2.10×10^{18} cm^{-3} under 10Wcm^{-2}

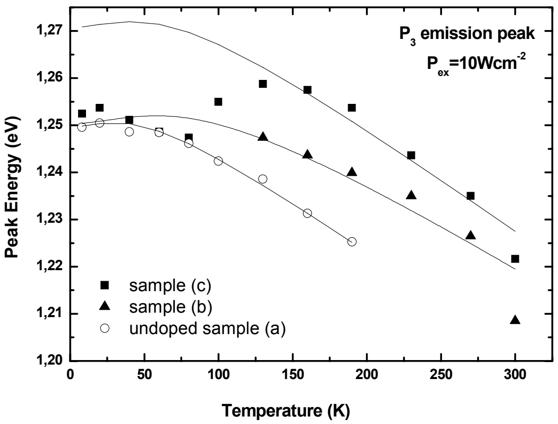

Fig. 3: Temperature dependence of the energy emission peak P_3 for the samples (a), (b) and (c)

spectra (Fig. 2a-2c) are composed by an emission peak P_3 that appears at 1.25 eV and becomes dominant at high temperature. The P_3 peak shifts to lower energies with increasing temperature; this indicates that it is attributed to the $GaAs_{1-x}N_x$ band edge emission. The calculated band gap energy of $GaAs_{1-x}N_x$ alloys with x =1.5% is found equal to 1.25 eV at 10 K using the band-anticrossing model[17] with C_{MN}=2.7 eV[7] and E_N = 1.67 eV.

The temperature dependence of the PL peak energy P_3 for the three samples is shown in Fig. 3. We note that the PL peak energies decrease with temperature but the shift is smaller than observed in GaAs material[7]. An "S-Shape" phenomenon appears for the highly Si-doped sample(c) in the temperature range of 10-130 K; it is attributed to the recombination of photogenerated carriers trapped by localized states in GaAsN, as previously observed by Dumont *et al.*[18]. The PL peak energies at high temperature were fitted using the Bose-Einstein statistical expression[19]:

$$E_g(T) = E_B - a.\left[\frac{2}{\exp(\frac{\theta}{T})-1}+1\right]+\xi.k_BT \qquad (1)$$

where a(eV) represents the electron-phonon interaction strength, θ(K) is related to the average phonon energy, k_B is the Boltzmann constant and ξ is the parameter reflecting the shape of the joint density of states, which is assumed to be 1/2 here. The best fit (Fig. 3) was obtained with the parameters listed in Table 2, which confirm that the emission P_3 is attributed to the $GaAs_{1-x}N_x$ band gap energy. Both the electron-phonon interaction strength and the average phonon energy are found to increase for the samples (b) and (c) comparing to the undoped sample (a). The increase of the 'a' parameter can be attributed to the decrease of the defects density leading to an increase of the PL efficiency. While the increase of the 'θ' parameter can be related to the silicon vibration mode.

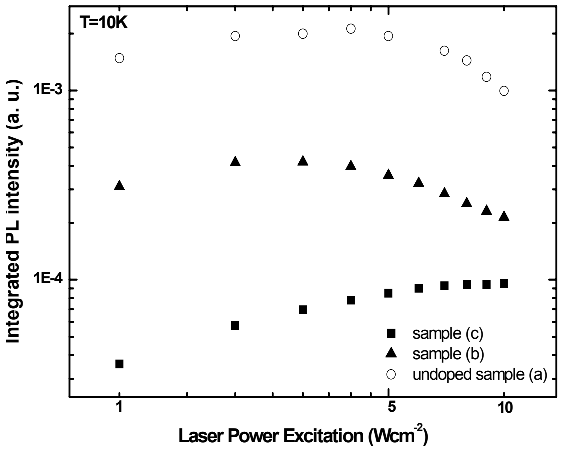

Fig. 4: 10K integrated PL intensity as function of the laser power excitation of the wide band situated at 0.83 eV for the samples (a), (b) and (c)

We can also access to the carriers thermal activation E_a of the wide band by using the Arrhenius model[20,21]:

$$I_{PL}(T)=\frac{I_0}{1+a_1T^{\frac{3}{2}}+a_2T^{\frac{3}{2}}\exp(-\frac{E_a}{k_BT})} \qquad (2)$$

where $I_{PL}(T)$ is the integrated PL intensity, I_0 is a proportionality constant, E_a is the thermal activation energy, k_B is the Boltzmann constant and a_1, a_2 are fitting parameters.

We reported in Table 3 the fitting parameters using the Arrhenius model for the 0.83 eV wide band. We note a decrease of the activation energy by increasing the silicon contents; this result can be explained by the fact that the silicon decreases the potential barrier between the wide band and the non radiative states.

The study of the integrated PL intensity (Fig. 4.) as function of the laser power excitation in the 1-10 Wcm^{-2} range of the wide band situated at 0.83 eV, shows an uncommon phenomenon for the undoped (a) and the lower Si-doped (b) samples. In fact, we note that the integrated PL intensity increases from 1 to 3 Wcm^{-2} and decreases beyond. For the highly Si-doped sample (c), the evolution of the integrated PL intensity as function of the laser power excitation shows a linear behavior with a slope lower than the unit (slope = 0.41). Such a slope value[20,22] allows us to attribute the wide band to an emission peak relative to excitons bound to defects. This is another confirmation that it can be attributed to deep localized states. For high power excitation values, the observed decrease in the integrated PL intensity is much pronounced for the undoped sample (a) and disappears for the highly doped sample (c). This character can be attributed to a non radiative recombination process.

CONCLUSION

We have presented photoluminescence measurements of Si-doped $GaAs_{0.985}N_{0.015}$ samples grown on GaAs substrates by Molecular Beam Epitaxy. We have noted that the PL spectrum is essentially formed by a wide band situated at 0.83 eV and one or two fine structures at 1.21 eV and 1.25 eV attributed respectively to nitrogen localized state and to the $GaAs_{1-x}N_x$ band gap. The introduction of the silicon decreases considerably the wide band PL intensity. We assume that this wide band is related to the presence of deep localized states caused by a three-dimensional GaAsN layer growth, which is annihilated by Si-doping. At high laser power excitation, the strong decrease of the PL intensity observed for the undoped sample (a) is reduced by the Si-doping; this effect can be interpreted by the reduction of the non radiative recombination process.

REFERENCES

1. Ferreria, M.D. and E.A. Imhoff, Power-dependent photoluminescence spectral shift in InGaAsP semiconductors, 1992. AMP J. Technol., 2: 70.
2. Mascharenhas, A., Y. Zhang, J. Verley and M.J. Seong, Overcoming limitations in semiconductor alloy design , 2001. Superlattices and Microstructures 29: 395.
3. Walukiewicz, W., Narrow band gap group III-nitride alloys, 2004. Physica E, 20: 300.
4. Weyers, M., M. Sato and H. Ando, Red Shift of Photoluminescence and Absorption in Dilute GaAsN Alloy Layers, 1992. Jpn. J. Appl. Phys., 31: L853.
5. Yaguchi, H., S. Kikuchi, Y. Hijikata, S. Yoshida, D. Aoki and K. Onabe, Photoluminescence study on temperature dependence of band gap energy of GaAsN alloys, 2001. Phys. Stat. Sol. (b), 228: 273.
6. Ager, J.W.III and W. Walukiewicz, Current status of research and developpement of III-N-V semiconductor alloys, 2002. Semiconductor Sci. Technol., 17: 741.
7. Chtourou, R., F. Bousbih, S. Ben Bouzid and F.F. Charfi, Effect of nitrogen and temperature on the electronic band structure of $GaAs_{1-x}N_x$ alloys, 2002. Appl. Phys. Lett., 80: 2075.
8. Lama, T.E., A.A. Quivy, S. Martini, M.J. Da Silva and J.R. Leite, Smooth p-type GaAs (001) films grown by molecular beam epitaxy using silicon as the dopant , 2005. Thin Solid Films, 474: 25.
9. Li, Z.Q., H. Chen, H.F. Liu, L. Wan, Q. Huang and J.M. Zhou, Photoluminescence study of Si doping cubic GaN grown on (001) GaAs substrates by molecular beam epitaxy, 2001. J. Crystal Growth, 227-228: 420.
10. Miyagawa, A., T. Yamamoto, Y. Ohnishi, J.T. Nelson and T. Ohachi, Silicon doping into MBE-grown GaAs at high arsenic vapor pressures, 2002. J. Crystal Growth, 237-239: 1434.
11. Goepfert, I.D., E.F. Schubert, A. Osinsky, P.E. Norris and N.N. Faleev, Experimental and theoretical study of acceptor activation and transport properties in p-type $As_xGa_{1-x}N$/GaN superlattices, 2000. J. Appl. Phys., 88: 2030.
12. Grieshaber, W., E.F. Schubert, I.D. Goepfert, R.F. Karlicek, M.J. Schurman and C. Tran, Competition between band gap and yellow luminescence in GaN and its relevance for optoelectronic devices, 1996. J. Appl. Phys., 80: 4615.
13. Yu, P.Y. and M. Cardona, 2001. Fundamentals of Semiconductors: Physics and Materials Properties. Springer, Third Edn.
14. Jakiela, R. and A. Barcz, Diffusion and activation of Si implanted into GaAs, 2003. Vacuum, 70: 97.
15. Wu, J., W. Walukiewicz and E.E. Haller, Calculation of the ground state of shallow donors in $GaAs_{1-x}N_x$, 2001. J. Appl. Phys., 89: 789.
16. Viturro, R.E., M.R. Melloch and J.M. Woodall, Optical emission properties of semi-insulating GaAs grown at low temperatures by molecular beam epitaxy, 1992. Appl. Phys. Lett., 60: 24.
17. Shan, W., W. Walukiewicz, J.W. Ager III, E.E. Haller, J.F. Geisz, D.J. Friedman, J.M. Olson and S.R. Kurtz, Band Anticrossing in GaInNAs Alloys , 1999. Phys. Rev. Lett., 82: 1221.
18. Dumont, H., L. Auvray, Y. Monteil, F. Saidi, F. Hassen and H. Maaref, Radiative N-localized recombination and confinement in GaAsN/GaAs epilayers and quantum well structures, 2003. Optical Materials, 24: 303.
19. Viña, L., S. Logothetidis and M. Cardona, Temperature dependence of the dielectric function of germanium, 1984. Phys. Rev. B, 30: 1979.
20. Gasanly, N.M., A. Aydinli and N.S. Yuksek, Temperature- and excitation intensity-dependent photoluminescence in TlInSeS single crystals, 1997. J. Phys.: Condensed Matter, 14: 13685.
21. Krustok, J., H. Collan and K. Hjelt, Does the low-temperature Arrhenius plot of the photoluminescence intensity in CdTe point towards an erroneous activation energy?, 1997. J. Appl. Phys., 81: 1442.
22. Schmidt, T., K. Lischka and W. Zulehner, Excitation-power dependence of the near-band-edge photoluminescence of semiconductors, 1992. Phys. Rev. B, 45: 8989.