phys. stat. sol. (b) **228**, No. 1, 273–277 (2001)

Photoluminescence Study on Temperature Dependence of Band Gap Energy of GaAsN Alloys

H. YAGUCHI[1]) (a), S. KIKUCHI (a), Y. HIJIKATA (a), S. YOSHIDA (a), D. AOKI (b), and K. ONABE (b)

(a) Department of Electrical and Electronic Systems, Saitama University, 255 Shimo-Okubo, Saitama-shi, Saitama 338-8570, Japan

(b) Department of Advanced Materials Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

(Received July 16, 2001; accepted July 19, 2001)

Subject classification: 78.55.Cr; 78.66.Fd; S7.14

We have studied the temperature dependence of photoluminescence (PL) spectra of GaAsN alloys. The PL peak energy shift due to the temperature change decreases with increasing N concentration of GaAsN alloys. The localized state emission partly contributes to the decrease in the PL peak energy shift. In addition, the small PL peak energy shift at high temperatures is due to the reduction in the temperature dependence of the band gap energy. From the analysis using the Bose-Einstein statistical expression, the average phonon energy is much larger than that expected from the linear interpolation between GaAs and GaN, indicating that the interaction between electrons and phonons localized at N atoms plays an important role in the reduction of the temperature dependence of the band gap energy of GaAsN alloys.

1. Introduction

Recently, III–V–N alloys have been receiving much attention because of their unique electronic and optical properties, and thus they are expected to be novel materials for optoelectronic device applications. Particularly, (In)GaAsN alloy is a promising material for long-wavelength semiconductor lasers with superior characteristic. As for optical-fiber communications, the stability against temperature during operation of the semiconductor lasers is desirable, i.e. the output power stability and the wavelength stability. Large conduction band offsets between (In)GaAsN and (Al)GaAs leads to an output power stability against temperature, and indeed a high characteristic temperature for the InGaAsN semiconductor laser is obtained [1]. On the other hand, the temperature dependence of the band gap energy determines the wavelength stability. It is reported that the temperature dependence of the band gap energy of GaAsN alloy is smaller than GaAs from absorption measurements [2] and is explained in terms of band anticrossing model [3]. In addition, photoluminescence measurements revealed that the emission related to localized states results in the reduction of the PL peak energy shift due to the temperature change [4]. In this study we have measured photoluminescence (PL) spectra of GaAsN alloys to examine in detail the temperature dependence of the band gap energy and the influence of localized states.

[1]) Corresponding author; Phone: +81 48 858 3841; Fax: +81 48 858 3841; e-mail: yaguchi@opt.ees.saitama-u.ac.jp

 0370-1972/01/22811-0273 $ 17.50+.50/0

2. Experimental

The samples used in this study were GaAsN alloys the N concentration of which ranged from 0 to 3.1% on GaAs (001) substrates grown by metalorganic vapor phase epitaxy (MOVPE). The sources for Ga, As, and N were trimethylgallium, arsine, and 1,1-dimethylhydrazine, respectively. Owing to the optimization of the MOVPE growth, highly luminescent samples have been successfully obtained even without post annealing [4]. The PL measurements were carried out at 10 to 300 K using an Ar ion laser (488 nm, 10 mW) as the excitation source.

3. Results and Discussion

Figure 1 shows the temperature dependence of the PL spectra of GaAsN alloy with the N concentration of 0.97%. Emission related to localized states is observed at low temperatures and vanishes with increasing temperature. On the other hand, a new PL peak appears at the higher energy side and becomes dominant with increasing temperature. The PL peak which is dominant at high temperatures shifts to lower energies with increasing temperature. This indicates that the PL observed at high temperatures is the band edge emission and that the peak shift is related to the temperature dependence of the band gap energy.

The temperature dependence of the PL peak energy of GaAsN alloys is shown in Fig. 2. At low temperatures, except GaAs, the PL peak energy rapidly decreases with increasing temperature and the peak intensity also rapidly decreases, and at the end the lower energy emission vanishes. As can be seen from this figure, the PL peak energy shift is reduced as the N concentration increases, which partly results from the influence of the localized states in GaAsN alloys. However, the PL peak shift at high temperatures also decreases with increasing N concentration. This indicates that the temperature dependence of the band gap energy of GaAsN alloys decreases with increasing N concentration. Thus, the PL peak energies at high temperatures were analyzed by the Bose-Einstein statistical expression [5],

$$E_{\mathrm{PL}} = E_{\mathrm{B}} - a_{\mathrm{B}} \left[\frac{2}{\exp\left(\Theta_{\mathrm{B}}/T\right) - 1} + 1 \right] + \alpha k_{\mathrm{B}} T \,, \qquad (1)$$

where a_{B} represents the electron–phonon interaction strength, Θ_{B} is

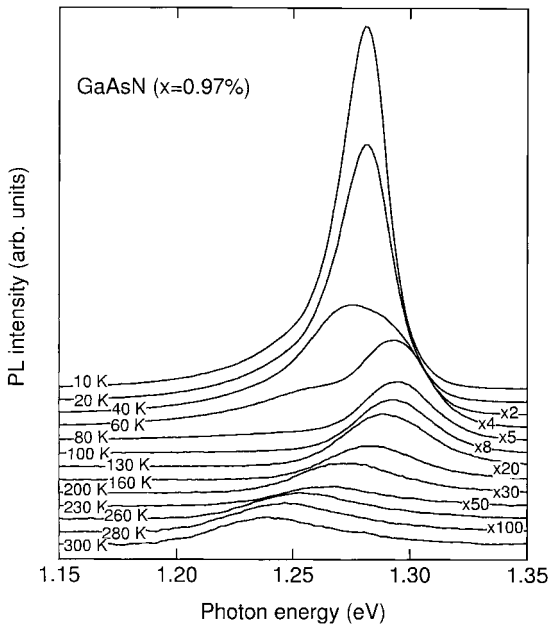

Fig. 1. Temperature dependence of photoluminescence spectra of $GaAs_{1-x}N_x$ ($x = 0.97\%$)

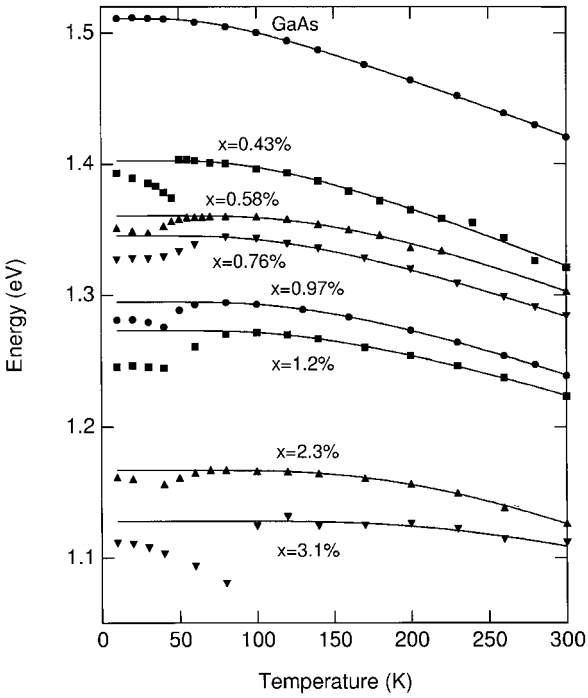

Fig. 2. Temperature dependence of photoluminescence peak energy of GaAsN alloys

the average phonon energy, and α is the parameter reflecting the shape of the joint density of states, which is assumed to be 1/2 here. T is the temperature and k_B is Boltzmann constant. Equation (1) was fitted to the PL peak energies at high temperatures where the localized states and exciton is less influenced. The solid curves shown in Fig. 2 are obtained from this analysis. The band gap energy shift due to temperature change can be also estimated.

Figure 3 shows the band gap energy differences between 0 and 300 K as a function of N concentration. The N concentration dependence of the band gap shift of GaAsN alloys with temperature changed between 25 and 297 K estimated from absorption measurements by Uesugi et al. [2] is also

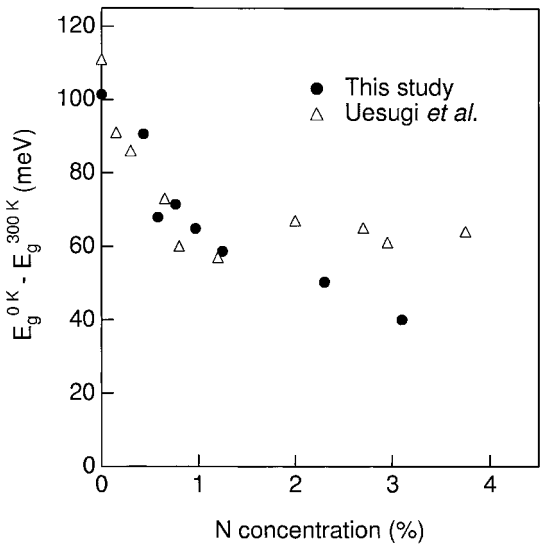

Fig. 3. Band gap energy differences between 0 and 300 K as a function of N concentration

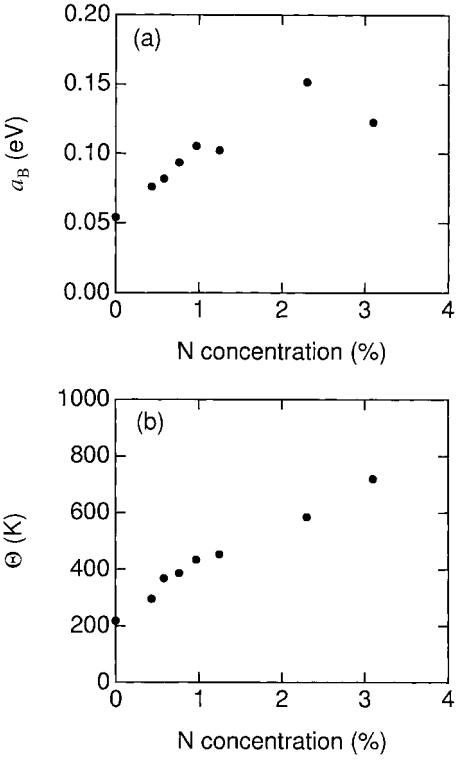

Fig. 4. Electron–phonon interaction strength and average phonon energy obtained from the analysis using the Bose-Einstein statistical expression

shown in this figure. For N concentration of 1%, our results are in good agreement with the results by Uesugi et al. However, the band gap energy shift still decreases with increasing N concentration for $x > 1\%$ in our results.

Figure 4 shows the electron–phonon interaction strength and the average phonon energy obtained from the analysis using the Bose-Einstein expression. Both the electron–phonon interaction strength and the average phonon energy are found to increase with increasing N concentration. However, the average phonon energy obtained from the analysis is much larger than that estimated from the linear interpolation between the longitudinal optical (LO) phonon energies of GaAs ($293\,\mathrm{cm}^{-1}$) and cubic GaN ($741\,\mathrm{cm}^{-1}$) [6]. For example, the average phonon energy of $GaAs_{1-x}N_x$ ($x = 2.3\%$) is estimated as $410\,\mathrm{cm}^{-1}$ in this study. This suggests that the interaction between electrons localized at N atoms interact preferentially with the localized Ga–N vibration mode with higher frequency. In fact, it is reported that GaN-type LO_2 phonon energy is $473\,\mathrm{cm}^{-1}$ [7] for $GaAs_{1-x}N_x$ alloys ($x = 2$–3%). Thus, the interaction between electrons localized at N atoms and the Ga–N localized vibration is considered to play an important role in the reduction of the temperature dependence of the band gap energy of GaAsN alloys.

4. Conclusions

We studied the temperature dependence of the photoluminescence of GaAsN alloys. The PL peak energy shift due to the temperature change decreases with increasing N concentration of GaAsN alloys. The decrease in the PL peak energy shift is partly due to the influence of the localized state emission which is dominant at low temperatures. In addition, the reduction in the temperature dependence of the band gap energy is responsible for the small PL peak energy shift. The Bose-Einstein statistical expression is used to analyze the temperature dependence of the band gap energy, and both the electron–phonon interaction strength and the average phonon energy are found to increase with increasing N concentration. The average phonon energy is much larger than that expected from the linear interpolation between the phonon energies of GaAs and GaN. This indicates that the preferential interaction of elec-

trons localized at N atoms interact with the localized Ga–N vibration mode with higher energy leads to the reduction of the temperature dependence of the band gap energy of GaAsN alloys.

Acknowledgements This work was partly supported by Nippon Sheet Glass Foundation for Materials Science and Engineering.

References

[1] M. KONDOW, T. KITATANI, K. NAKAHARA, and T. TANAKA, Jpn. J. Appl. Phys. **38**, L1355 (1999).
[2] K. UESUGI, I. SUEMUNE, T. HASEGAWA, T. AKUTAGAWA, and T. NAKAYAMA, Appl. Phys. Lett. **76**, 1285 (2000).
[3] I. SUEMUNE, K. UESUGI, and W. WAULUKIEWICZ, Appl. Phys. Lett. **77**, 3021 (2000).
[4] K. ONABE. D. AOKI, J. WU, H. YAGUCHI, and Y. SHIRAKI, phys. stat. sol. (a) **176**, 231 (1999).
[5] L. VINA, S. LOGOTHETIDIS, and M. CARDONA, Phys. Rev. B **30**, 1979 (1984).
[6] H. YAGUCHI, J. WU, B. ZHANG, Y. SEGAWA, H. NAGASAWA, K. ONABE, and Y. SHIRAKI, J. Cryst. Growth **195**, 323 (1998).
[7] A. M. MINTAIROV, P. A. BLAGNOV, V. G. MELEHIN, N. N. FALEEV, J. L. MERZ, Y. QIU, S. A. NIKISHIN, and H. TEMKIN, Phys. Rev. B **56**, 15836 (1997).