Available online at www.sciencedirect.com
SCIENCE DIRECT®

ELSEVIER

Physica E 20 (2004) 308–312

PHYSICA E

www.elsevier.com/locate/physe

Temperature-dependent optical properties of wurtzite InN

Fei Chen[a], A.N. Cartwright[a,*], Hai Lu[b], William J. Schaff[b]

[a]*Department of Electrical Engineering, University at Buffalo, State University of New York, 332 Bonner Hall, Buffalo, NY 14260, USA*
[b]*Department of Electrical and Computer Engineering, Cornell University, Ithaca, NY 14853, USA*

Abstract

Optical properties and carrier recombination dynamics of a series of InN epilayers, with varying free electron concentrations, grown by molecular beam epitaxy were studied by steady-state photoluminescence (PL) and time-resolved differential transmission spectroscopy. At room temperature strong PL around 0.7 eV was observed. Temperature-dependent PL measurements show a redshift of the peak energy and a linear increase of the emission linewidth with temperature. Furthermore, our results demonstrate that room temperature carrier lifetimes are inversely proportional to the free electron concentrations for theses samples. Carrier lifetime as long as 1.3 ns was observed in the best quality sample, indicating a highly improved crystalline quality.
© 2003 Elsevier B.V. All rights reserved.

PACS: 78.47.+p; 71.35.−y

Keywords: Optical properties; Time-resolved spectroscopy

InN is the least studied of the III-nitride materials, which are currently attracting intense interests. Recent progress in growth techniques has led to the availability of high crystalline quality hexagonal InN layers with low electron concentrations [1–3]. The optical characterizations of these improved wurtzite InN crystals have provided convincing evidence that there is a bandedge around 0.7–0.9 eV [4–7], much narrower than ∼1.9 eV that was measured in earlier studies of InN [8,9]. This unexpected discovery opened a whole new range of opportunities for the fabrication of optoelectronic devices, such as infrared emitters. Even with all the significant recent developments in the measurement and calculation [10,11] of the physical and optical properties of InN, the luminescence mechanisms and carrier recombination dynamics of this semiconductor are still poorly understood.

In this paper, we report our comprehensive studies of optical properties and carrier dynamics of InN epilayers. In contrast to the anomalous blueshift of photoluminescence (PL) peak energy with temperature presented in recent literature [4], the typical redshift with temperature was observed. Furthermore, time-resolved differential transmission spectroscopy shows the room temperature carrier lifetimes are inversely proportional to the free electron concentrations for theses samples. Carrier lifetime as long as 1.3 ns was observed in the best quality sample, indicating a highly improved crystalline quality.

Three InN films were grown on (0 0 0 1) sapphire with different buffer layers by molecular beam epitaxy. The details of the growth technique have been reported elsewhere [2]. The structures consist of:

* Corresponding author. Tel.: +1-716-645-3115x1205.
E-mail address: anc@eng.buffalo.edu (A.N. Cartwright).

1386-9477/$ - see front matter © 2003 Elsevier B.V. All rights reserved.
doi:10.1016/j.physe.2003.08.024

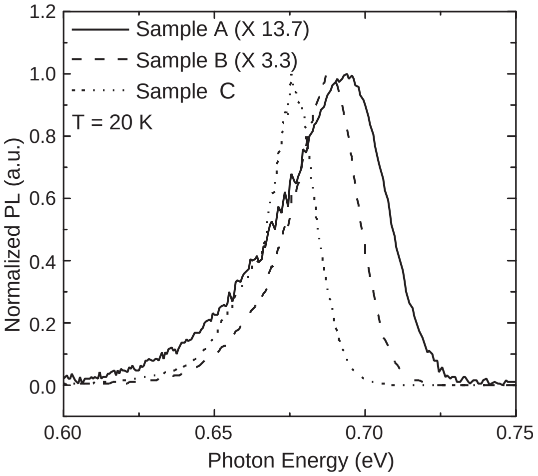

Fig. 1. PL spectra for sample A (solid line), sample B (dashed line) and sample C (dotted line) at 20 K.

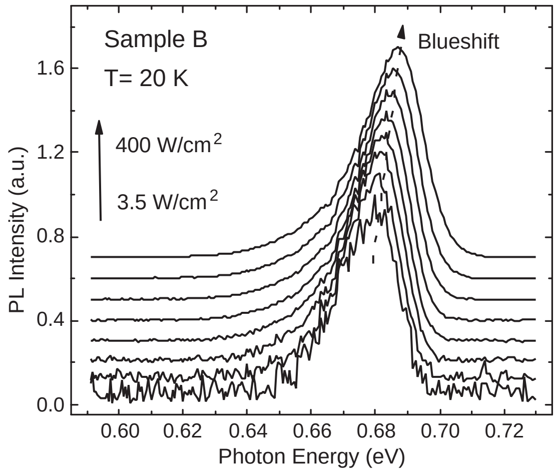

Fig. 2. PL spectra as a function of excitation density on sample B at 20 K.

(a) sample A, AlN(300 nm)/InN(350 nm); (b) sample B, GaN(220 nm)/InN(850 nm); and (c) sample C, GaN(300 nm)/$In_{0.65}Ga_{0.35}N$(110 nm)/InN(7500 nm). The mobility and free electron concentration are measured at room temperature as 826 cm^2/V s and 2.7×10^{18} cm^{-3} for sample A, 1340 cm^2/V s and 1.3×10^{18} cm^{-3} for sample B, and 2050 cm^2/V s and 0.4×10^{18} cm^{-3} for sample C, respectively.

Unintentionally doped InN has often been found to have very high electron densities. These donors have not yet been identified, but potential candidates are nitrogen vacancy [8,12], the nitrogen antisite [8,12], or the oxygen and silicon impurities [13]. Due to the small effective electron mass of InN, the Fermi surface in the conduction band has shown a strong dependence on the free electron concentration [7]. Thus, the free electrons can shift the absorption edge and PL peak energy to higher energy due to the bandfilling effects. Fig. 1 shows the typical PL spectra at 20 K excited by an Ar^+ laser at 3.41 eV. It is seen that both the PL peak energy and emission width increase with increasing free electron concentration, indicating that the transitions from high energy states in the conduction band contribute significantly to the PL spectra. Moreover, sample C exhibits the highest emission efficiency, which is 13.7 times and 3.3 times of samples A and B, respectively. The decreasing emission efficiency with increasing free electron concentration suggests the donorlike defects or impurities may stimulate the formation of the non-radiative recombination centers, which reduce the carrier lifetime and the internal quantum efficiency. This is further verified by the time-resolved measurements discussed later.

In order to clarify the bandfilling effects, we also performed the excitation density-dependent PL measurements on these samples. Fig. 2 shows PL spectra observed for different excitation powers ranging from 3.5 to 400 W/cm^2 at 20 K on sample B. When the excitation power was increased, the PL peak energy exhibited a pronounced blueshift, while the peak intensity varied in proportion to the excitation power. We consider that it shows a near-band-edge emission and the blueshift of PL peak energy with excitation power is attributed to the increase of quasi-electron Fermi energy in the conduction band.

Fig. 3(a) shows the detailed temperature-dependence of PL spectra on sample B. Consistent with the recent observations of other groups [4,5], strong PL emission near 0.7 eV was observed at room temperature. In contrast to the anomalous blueshift of PL peak energy with temperature, presented in recent literature [4], the typical redshift with temperature was observed. It is interesting to notice that the magnitude of the redshift of PL peak energy in sample C with temperature is significantly stronger than that in sample A. It is likely that more free electrons thermalized from defects or impurities in sample A can fill the conduction band and compensate the effects of the

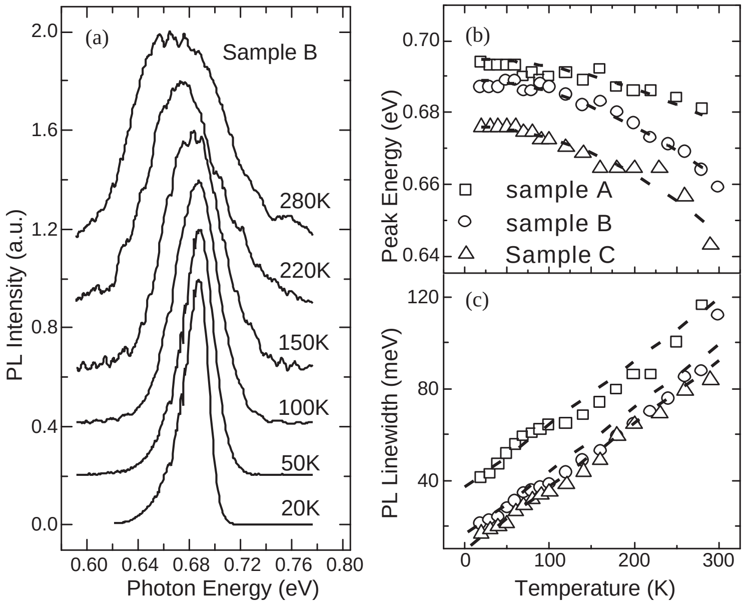

Fig. 3. (a) PL spectra as a function of temperature for sample B. (b) PL peak energy as a function of temperature. (c) Linewidth of PL vs. temperature. The dotted lines show the linear fits.

bandgap shrinkage with temperature. Furthermore, the linewidth of the PL increases linearly with temperature in Fig. 3(c) indicating that longitudinal acoustic phonon scattering dominates the emission broadening processes in these InN samples.

Subsequently, subpicosecond differential transmission measurements were performed to determine the carrier lifetimes and evaluate the material quality of these samples, by pumping with an 800 nm laser pulse and probing with a tunable infrared pulse (0.9–3.6 μm) with typical pulse widths of ∼300 fs. Fig. 4 shows the measured differential transmission signal as a function of time delay for a fixed pump fluence of 120 $\mu J/cm^2$ at room temperature. The probe energies were set corresponding to the PL peak energy for each sample. Notice that all the curves exhibit a single exponential decay with time. The recombination of the photogenerated carrier follows three different channels, non-radiative defect-related, bimolecular radiative interband, and non-radiative Auger. The carrier lifetime is given by

$$\frac{1}{\tau} = \frac{1}{\tau_{nr}} + B(n + n_0) + C(n + n_0)^2,$$

where B and C are interband and Auger recombination coefficient, n is the photogenerated carrier density and n_0 represents the free electron concentration. For radiative and Auger recombinations, the lifetimes show a carrier density-dependent characteristic. Under the pump fluence used in our experiments, the corresponding maximum average photoexcited carrier density is estimated as 2.8×10^{18} cm^{-3}, which is of the same order or higher than the free electron concentration in each sample. Thus, the single exponential decay and pump-fluence-independent decay time (ranging from 12 to 500 $\mu J/cm^2$) for all the samples indicate that the defect-related non-radiative recombination is the dominant mechanism at room temperature. The recombination lifetimes varying from 150 ps (sample A), 520 ps (sample B), to 1320 ps (sample C) can be extracted by an exponential fit. Compared with the recombination lifetime of InGaN epilayers (∼100 ps) at room temperature [14], this long carrier lifetime (1320 ps for sample C) suggests that the InN has high crystalline quality as a result of the improved growth technique. The inset graph of Fig. 4 displays the measured carrier lifetime as a function of free electron density. We found the lifetime is

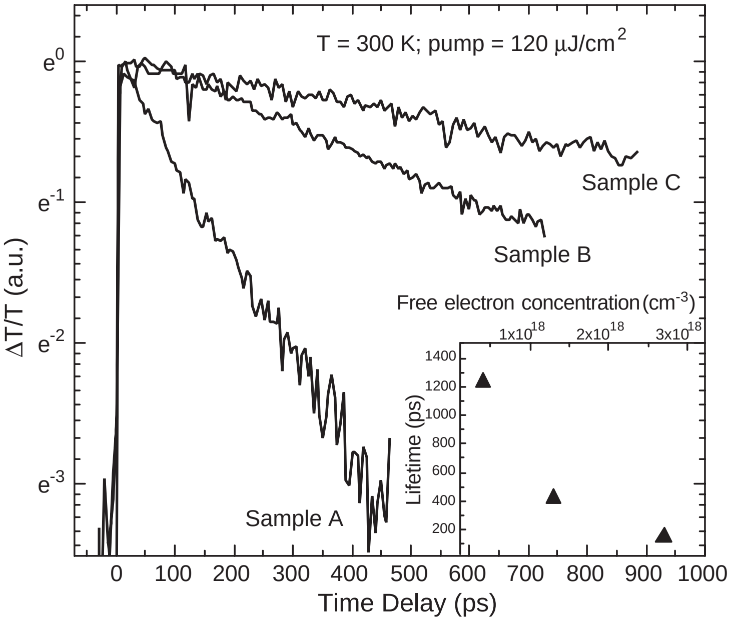

Fig. 4. Strength of the differential transmission signal vs. time delay under the pump fluence of 120 $\mu J/cm^2$ at room temperature. The inset graph shows the carrier lifetime as a function of free electron concentration.

inversely proportional to the free electron concentration. Let us remember the non-radiative lifetime τ_{nr} is related to the defect density N_{defect} by the following expression: $1/\tau_{nr} = \sigma \upsilon N_{defect}$, where σ is the carrier capture cross section and υ represents the carrier velocity. Thus, it is reasonable to assume that the donorlike defects or impurities may stimulate the formation of the non-radiative recombination centers, reducing the carrier lifetime.

In conclusion, temperature-dependent optical properties and carrier recombination dynamics in InN have been studied. The linewidth of the PL increases linearly with temperature indicating that longitudinal-acoustic phonon scattering dominates the emission broadening processes in these InN samples. We have found the room temperature carrier lifetimes are inversely proportional to the free electron concentrations. It is suggested that donor-like defects or impurities may stimulate the formation of non-radiative recombination centers, reducing the carrier lifetime. More importantly, the observed long carrier lifetime (1.3 ns in sample C) and strong luminescence at room temperature make this InN crystal an excellent candidate for the infrared emitters.

This work was supported by the National Science Foundation CAREER Award under Grant No. 9733720, and the Office of Naval Research Young Investigator Program under Award No. N00014-00-1-0508.

References

[1] V.V. Mamutin, A.A. Vekshin, V.Y. Davydov, V.V. Ratnikov, T.V. Shubina, S.V. Ivanov, P.S. Kopev, M. Karlsteen, U. Soderwall, M. Willander, Phys. Stat. Sol. (a) 176 (1999) 247.
[2] H. Lu, W.J. Schaff, J. Hwang, H. Wu, W. Yeo, A. Pharkya, L.F. Eastman, Appl. Phys. Lett. 77 (2000) 2548.
[3] Y. Saito, N. Teraguchi, A. Suzuki, T. Araki, Y. Nanishi, Jpn. J. Appl. Phys. Part 2 40 (2001) L91.
[4] J. Wu, W. Walukiewicz, K.M. Yu, J.W. Ager III, E.E. Haller, H. Lu, W.J. Schaff, Y. Saito, Y. Nanishi, Appl. Phys. Lett. 80 (2002) 3967.
[5] T. Matsuoka, H. Okamoto, M. Nakao, H. Harima, E. Kurimoto, Appl. Phys. Lett. 81 (2002) 1246.
[6] V.Y. Davydov, A.A. Klochikhin, R.P. Seisyan, V.V. Emtsev, S.V. Ivanov, F. Bechstedt, J. Furthmuller, H. Harima, A.V. Mudryi, J. Aderhold, O. Semchinova, J. Graul, Phys. Stat. Sol. B 229 (2002) R1.
[7] J. Wu, W. Walukiewicz, W. Shan, K.M. Yu, J.W. Ager III, E.E. Haller, H. Lu, W.J. Schaff, Phys. Rev. B 66 (2002) 201403.
[8] T.L. Tansley, C.P. Foley, J. Appl. Phys. 59 (1986) 3241.

[9] Q. Guo, A. Yoshida, Jpn. J. Appl. Phys. 33 (1994) 2453.
[10] F. Bechstedt, J. Furthmuller, J. Crystal Growth 246 (2002) 315.
[11] S. Wei, X. Nie, I.G. Batyrev, S.B. Zhang, Phys. Rev. B 67 (2003) 165209.
[12] M. Sato, Jpn. J. Appl. Phys. Part 2 36 (1997) L658.
[13] C. Stampfl, C.G. Van de Walle, D. Vogel, P. Kruger, J. Pollmann, Phys. Rev. B 61 (2000) R7846.
[14] M. Smith, G.D. Chen, J.Y. Lin, H.X. Jiang, M. Asif Khan, Q. Chen, Appl. Phys. Lett. 69 (1996) 2837.