Extended Abstracts of the 2006 International Conference on Solid State Devices and Materials, Yokohama, 2006, pp. 1086-1087

I-7-6L

The Observation of Anomalous Optical Berthelot-type Behaviors in Quaternary AlInGaN Semiconductor Heterosystems

Cheng-Wei Hung[1], Chih-Chun Ke[1], Tzer-En Nee[1], Chang-Cheng Chuo[2], and Zheng-Hong Lee[2]

[1] Department of Electronic Engineering, Chang Gung University,
259 Wen-Hwa 1st Road, Kwei-Shan, Tao-Yuan, Taiwan, Republic of China.
Tel: +886-3-2118800 ext 5791, Fax: +886-3-2118507, E-mail: neete@mail.cgu.edu.tw
[2] Electronics and Optoelectronics Research Laboratories,
Industrial Technology Research Institute, Hsin-Chu, Taiwan, Republic of China.

1. Introduction

The investigation about low-dimensional nanocrystalline materials and devices have extensively proposed in past decade, such as quantum dots (zero-dimensional system), quantum wires (one-dimensional system), and quantum well (two-dimensional system). These nanometer-sizes of optoelectronic devices can be expect to fabricate through various epitaxial technique. In recent years, some research teams have noticed the temperature dependence of the S-shaped emission peak position and W-shaped full width at half-maximum [1]. They attributed the abnormal PL emission peak blueshift to the incorporation of indium into AlInGaN multiple quantum wells. The indium-rich region originated from the solid phase immiscibility between InN and GaN. Even though the current epitaxial techniques are gradually developing, laying accurate quantity of In or Al molar fraction in the AlInGaN active layer still perplex most epitaxy engineers. In this work, we focus on the competition of transport dynamics induced anomalous optical properties in AlInGaN heterostructures by means of Berthelot-type model based on the disorder system. We investigated abnormal characteristics caused by indium incorporation from the angle of disorder like system.

We observed the thermodynamic fluctuations and extremely uneven distribution of indium in nitride-based nanostructures. By means of the statistical disorder like system, we elucidated some phenomena which similar to disorder system in quaternary AlInGaN. We attributed the anomalous optical properties to the mechanical competition between radiative and nonradiative decay dynamics. The combination effect of the two mechanisms, temperature related equation derived from the model can demonstrate that the critical turning-point was gradually raised with extending the pairs of wells. This significant finding indicates that the increase in the pairs of wells will augment the disorder-like level of the indium composition in the heterostructures. On other hand, carriers need higher thermal energy to escape from the localized minima energy state due to the larger entropy. As have been expected, the temperature dependence of emission energy was contingent on the Berthelot-type dynamical mechanisms in AlInGaN/GaN heterosystems. At last, the correlation between relaxation process and structural irregularity will be discussed in detail as well.

2. Experiments

The samples investigated in this work were grown by metal organic vapor phase epitaxy (MOVPE) on c-plane sapphire substrates with a 25-nm-thick low temperature GaN nucleation layer. The sample structure consisted of a 3-μm-thick *n*-type GaN layer doped, a 3-μm-thick *n*-type AlGaN layer, a three-, five-, and ten-period undoped quaternary AlInGaN MQWs active layer and followed by a 0.15-μm-thick *p*-AlGaN contact layer. SiH_4 and Cp_2Mg were the n-type and p-type dopants, respectively. The doping concentrations were 5×10^{18} and 1×10^{19} cm^{-3}, respectively. Each quaternary AlInGaN MQW pair consists of a 2.5-nm-thcik well layer and a 10-nm-thick barrier layer. For the temperature-dependent photoluminescence (PL) measurements, samples were mounted in a closed-cycle He cryostat, and excited by a continuous-wave He-Cd laser, covering a wide temperature range, from 20 K to 300 K. The average excitation intensity was 20 mW. The luminescence signal was dispersed through a 0.5-meter monochromator, and then detected by a Si photodiode, employing a standard lock-in amplification technique.

3. Results and Discussion

In Fig. 1, we observed the temperature-dependent PL emission peaks of quaternary AlInGaN structures with various pairs of quantum wells. It has been found the emission peak of PL spectra exhibited an obvious red-blue-red shift, i.e., S-shaped shift. Here, we observed the turning-point temperature (T_t) on the S-shaped shift shape of the emission peaks. It has seen the turning-point temperature increased gradually with increasing the pairs of quantum wells. In the disorder heterosystems, increasing the quantum well layers, it was implicated to not only enhance degree of the entropy, but also augment the static-barriers in quaternary AlInGaN heterostructures. Therefore, for the higher numbers of the quantum well, it would require larger thermal energy to carrier escape from indium fluctuation centers and accomplish radiative recombinations. In the disorder like systems, increasing the quantum well layers merely implicated to enhance degree of the entropy, but also augments the statistic microbarrier in AlInGaN heterostructures.

We cited Varshini empirical formula to fit and estimate the degree of localization effects within the AlInGaN epitaxial layers, presented as σ [1]. The magnitudes of σ are

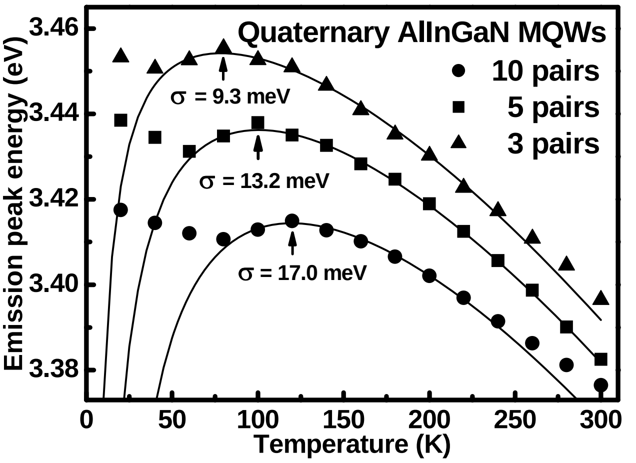

Figure 1 The photoluminescence emission peaks as a function of temperature for the AlInGaN samples with different pairs of quantum wells.

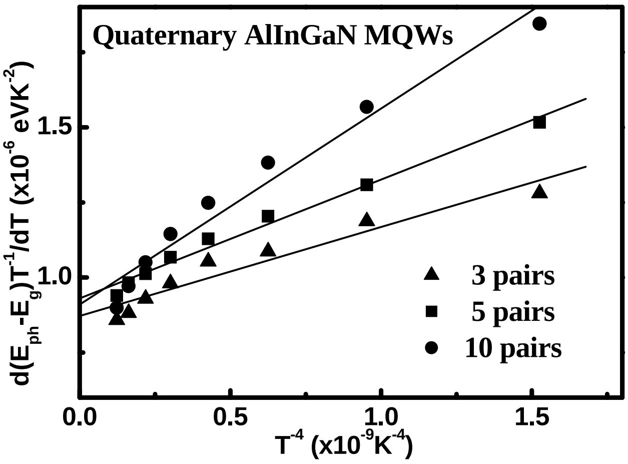

Figure 2 Plots of d(E_{ph}–E_g)T^{-1}/dT vs. T^{-4} for the positions of the temperature-dependent PL peak energies with different quantum well pairs. The slopes in plot indicate the magnitude of C_2.

9.3, 13.2 and 17 meV for 3QW, 5QW, and 10QW quaternary AlInGaN MQWs, respectively. As mentioned above, the higher quantum well pairs, the higher degree of disorder. It means that indium distribution more randomly and caused intensive localization effect.

As mentioned the competition of carrier transport dynamics, we considered the luminescence decay time formed with radiative rate and nonradiative rate can be express as

$$\frac{1}{\tau} = R_r + R_n = \upsilon_r \exp\left(\frac{-T_r}{T}\right) + \upsilon_n \exp\left(\frac{T}{T_n}\right) \qquad (1)$$

where $\{v_r, v_n, T_r, T_n\}$ are characteristic constants.

The Berthelot-type behavior is attributed to the carriers through nonradiative hopping mechanism across the statistic microbarrier width S with the vibrating frequency Ω. In the disorder like system, the Berthelot-type behaviors of the photon peak positions for these samples are shown in Fig. 2, employing the analytical formulations developed by V. A. Singh and G. C. John [2]. Under the assumption of the bandgap shrinkage with a linear thermal coefficient of C_1, the temperature dependence of the anomalous optical properties is uniquely characterized by

$$\frac{E_{ph} - E_g}{T} = -C_1 - \frac{C_2}{T^3} \qquad (2)$$

We supposed that the employed equation was linear system based on Berthelot-type model, by differentiate Eq. (2) and can be derived as Eg. (3)

$$\frac{d}{dT}\frac{(E_{ph} - E_g)}{T} = \frac{C_2}{T^4} \qquad (3)$$

where E_{ph} is photon energy, E_g is temperature dependent energy gap, C_1 is assumed to be constant irrespective of heterostructure in the device, C_2 is a variant to be of the opinion that correlated with different indium contents in the samples. From Eq.(3), we can derive the disorder like related parameter C_2 for the samples investigated in this work, the slopes indicate the magnitude of C_2 as shown in Fig. 2. Based on the Berthelot-type model, the turning temperature (T_t) can be extracted numerically by using the expression $T_t = (2C_2/C_1)^{1/3}$. It is correspond to we expected that the increase of the period of quantum wells result in the degree of disorder arise and the higher turning temperature as well.

4. Conclusion

We proposed that quaternary AlInGaN heterostructures can considered as disorder system because the indium fluctuation in the multiple quantum well epitaxial layers. Therefore, we cited the Berthelot type model to demonstrate the anomalous photoluminescence may result from the indium phase segregation. The In-rich phases were found to act as localized states and significant blueshift was observed with the increase of temperature. The entropy was arises with the pairs of quantum wells increased due to the indium phase segregation which would induced the so-called S-shaped emission luminescence spectra. We can observe the turning-point temperature (T_t) increased and the higher molar fraction of indium in active layer will have larger C_2, i.e., higher T_t. In summary, based on the Berthelot-type model, we can characterize the optical properties of quaternary AlInGaN from the viewpoint of disorder system.

Acknowledgments

This work was supported by the National Science Council of the Republic of China under Contract No. NSC 94-2215-E-182-007.

References

[1] K. Kazlauskas, G. Tamulaitis, A. Zukauskas, M. A. Khan, J. W. Yang, J. Zhang, E. Kuokstis, G. Simin, M. S. Shur, and R. Gaska, Appl. Phys. Lett. **82** (2003) 4501.

[2] M. Kapoor, V. A. Singh, and G. K. John, Phys. Rev. B. **61** (2000) 1941.