Extended Abstracts of the 1999 International Conference on Solid State Devices and Materials, Tokyo, 1999, pp. 194–195

Recombination Dynamics of Self-Trapped Excitons in the High-Efficient Blue LEDs under Reverse Bias Condition

Tsunemasa Taguchi, Hiromitsu Kudo, Hiroki Ishibashi, Ruisheng Zheng and Yoichi Yamada

Faculty of Engineering, Yamaguchi University, 2-16-1 Tokiwadai, Ube, Yamaguchi 755-8611, Japan
Phone/Fax: +81-836-35-9941 e-mail: taguchi@po.cc.yamaguchi-u.ac.jp

1. Introduction

It is very important to reveal the recombination mechanism and related emission center in the 10 candela-class blue and green LEDs (having an external quantum efficiency of about 20 %) because of a useful application to the new LED lighting source instead of incandescent bulb and fluorescent tube.

There are several controversy arguments concerning the recombination centers in $In_xGa_{1-x}N$-based quantum well LEDs. We have proposed a particular type of the localised excitons (such as self-trapped bound excitons)[1,2] which is different from the feature of usual localised excitons under the composition fluctuations in the mixed alloys. This looked like a correct picture for interpreting the high-radiative recombination process. However, since our proposed model, many researchers[3,4,5] have followed up the classical exciton localisation model on the basis of the large Stokes shift and relatively longer decay time constant, in spite of the fact that the localisation energy is extremely larger than the exction binding energy. The classical localisation model can not be accounted for understanding the recombination mechanisms. On the other hand, a model due to excitons localised in the self-organized quantum dots[6] has been adopted to explain the efficient recombination process in spite of the large dislocation densities and strong exciton-LO phonon interaction energy using only the temperature-dependent luminescence decay measurement. Very recently, Im et al[7] and Takeuchi et al[8] have suggested a model in which the band-to-band (or free-carrier) recombination is predominant in the quantum wells under a large internal piezoelectric field.

At present, any kinds of the recombination models can not fully explain the present high-efficient recombination mechanisms and can not clarify the related recombination center.

We describe a new recombination center model using the time-resolved spectral measurements under reverse bias condition. The proposed high-efficient emission process including two individual emission components may be related to the extrinsic self-trapped exciton recombination.

2. Results and discussion

2.1. Abnormal temperature dependence and self-trapping of excitons

We have carried out the precise temperature dependence of the emission band at 3.23 eV observed in a single epitaxial layer of $In_xGa_{1-x}N$ (x=0.08) on a GaN buffer between 4.2 and 300 K, as shown in Fig. 1. At 10 K, the emission band shows an asymmetric lineshape having a tail towards lower energy side. With increasing temperature, the apparent peak position moves towards higher energy side and subsequently a tail at lower energy side becomes dominant in a temperature of 100 K. There appear clearly two emission components each other separated by about 40 meV. With further increasing temperature, the higher energy component is decreased in intensity between 100 and 140 K. Such behavior does not obey to the Boltzman law because of no thermal depopulation between two levels. Above 140 K, the higher emission component becomes dominant again and moves towards lower energy side with temperature. Considering the temperature dependence of the two emission components, the abnormal temperature dependence of S-shaped[5] and Snake-type[2] of the dominant band in InGaN epilayer and MQW structure can be fully understood.

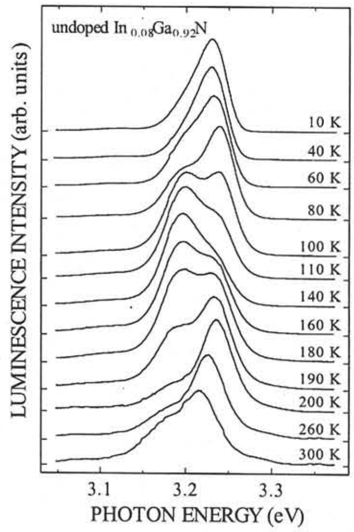

Fig. 1 Temperature dependence of the two emission components in an $In_{0.08}Ga_{0.92}N$ epilayer.

Two emission components were also clearly observed in the time-resolved emission sepectra at 4.5 K. The higher component at 3.23 eV shows a fast decay time of about 30 psec, whilst the lower component at 3.199 eV grows gradually after pulse excitation. This dynamic behavior indicates that the energy transfer between the two levels is taken place, and finally the low component appears after 450 psec, showing a longer decay time of about 500 psec.

Both components are derived from the self-trapped states, but the adiabatic potential curve in the C-C diagram of the high-emission component is related to the free state because of the shorter lifetime. The temperature dependence and decay time measurements suggest that the low-emission component may be ascribed to a typical recombination process of the self-trapped exciton as has been suggested in ZnSeTe[9]. This behavior, of course, depends on the optically injected carrier densities and impurity concentrations. The self-localisation of excitons can be accomplished by both the electronegativity difference between Ga and In atoms, and a lattice distortion around In atoms.

2.2. Dynamic behavior of ST exctions in LEDs

Two emission components have been detected in the electroluminescence (EL) spectra in a practical blue LED. Fig. 2 shows the PL spectra obtained in the super-bright SQW LED at 77 K as a function of reverse-bias voltage. The PL spectra from the active $In_{0.2}Ga_{0.8}N$ Layer are similar to those of EL. At zero bias, two components separated by about 40 meV appear. With increasing reverse-bias voltage, the peak position of the lower energy side slightly shifts to the higher energy side up to about 10 meV. Due to a small change in the 2D exction binding energy, the blue shift will occur. It is therefore thought that the effect of piezoelectric field on both EL and PL spectra is not important. Above –4.7 V, the higher energy component becomes dominant, and the peak position does not change at any bias voltages. However, even at –11.7 V (corresponding to an electric field of about 10^4 V/cm), both emission components were not completely deteriorated.

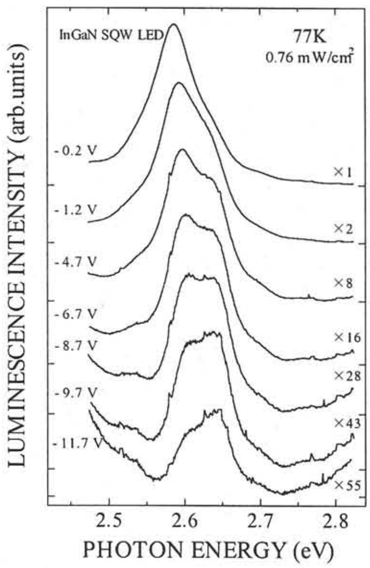

Fig. 2 Dependence of reverse-biased voltage on two emission components at 77 K.

At zero bias, the higher component shows a decay time of about 10 nsec. On the other hand, the lower component shows a decay time of 30 nsec, which is significantly affected by a band-bending of the active layer due to a compressive strain in the SQW, but not due to the piezoelectric field. By applying the reverse bias voltage, the higher component changes to a decay time of 3 nsec at –10V, and the lower component shows a decay time less than 2 nsec. This means that, by applying the reverse-bias voltage, the decay time of the lower emission component is particularly reduced by one order of magnitude as a result of the faster exciton transfer process from the lower emission component to the higher emission component as shown in Fig. 3.

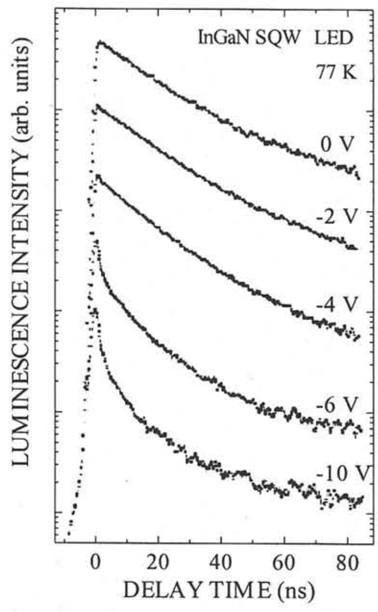

Fig. 3 Changes of the decay time at 77 K under the different reverse-bias voltages.

3. Summary

In conclusion, we have observed for the first time two predominant emission components speparated by about 40 meV in $In_xGa_{1-x}N$ films independent of composition x of In. These components may be stemmed from the extrinsic characters of excitons in InGaN mixed crystals having both a large eletronegativity and a lattice distortion around In atoms. We thus propose that the efficient recombination center can be related to the extrinsic self-trapped excitons which are strongly perturbed by the strained-quantum structures. This model will explain the small diffusion length of excitons, so that the high efficiency can be acquired by following the reduced transport of excitons to nonradiative recombination centers.

Acknowledgments

We acknowledge the financial support from the Japan National Project "The light for the 21st century" based on MITI/NEDO/JRCM system. One of the authors (T. T) would like to thank Dr. S. Nakamura and President E. Ogawa of Nichia Chemical Ind. for providing the samples used in the present study.

References

[1] T. Taguchi et al, Proc.23rd Int. Conf. on Physics of Semiconductors, eds. M. Scheffer and R. Zimmerman (1996) p.541.

[2] T. Taguchi et al. Proc. Int. Symp. on Blue Laser and Light Emitting Diodes (1996) p.372.

[3] S. Chichibu et al. Appl. Phys. Lett., **69** (1996) 14188.

[4] S. Bidnyk et al,. Appl. Phys. Lett., **72** (1998) 1623.

[5] Y. H. Cho et al, Appl. Phys. Lett., **73** (1998) 1370.

[6] Y. Narukawa et al, Phys. Rev., **B55** (1997) R1938.

[7] J. S. Im et al, Proc. 2nd Int. Symp. on Blue Laser and Light Emitting Diodes (1998)

[8] T. Takeuchi et al, Appl. Phys. Lett., **73** (1998) 691.

[9] D. Lee et al, Phys. Rev. Lett., **58** (1987) 1475.