DYNAMICAL GAIN FORMATION PROCESSES IN
InGaN MULTIPLE QUANTUM WELLS

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Dynamical gain formation processes in InGaN multiple quantum wells were investigated by means of time-resolved pump-and-probe measurement at 2 K. The photoexcited excitons rapidly relaxed into the localized states. Above the stimulation threshold, the localized states were filled and the photoexcited excitons existed at the delocalized states. The two-dimensional excitons at the delocalized states relaxed into the localized states, from which the optical gain was formed in terms of the stimulated emission process.

Recently, there has been an amount of works to understand the fundamental optical properties of III-V nitride compound semiconductors. Especially, it is important to clarify the radiative recombination mechanism and the lasing mechanism in InGaN quantum wells, because InGaN quantum wells have been used as the active layer for commercialized blue/green light-emitting diodes and blue laser diodes [1]. It has been reported that the spontaneous emission from the InGaN layers is due to the radiative recombination of excitons localized at the potential minima in the InGaN layers [2–4]. Furthermore, it has been proposed that the localized excitons produce the stimulated emission process [2–4]. Dynamical gain formation processes should be clarified in the next step. In this work, we have investigated exciton relaxation dynamics in InGaN multiple quantum wells (MQWs) at 2 K. We have performed the time-resolved pump-and-probe measurement employing a white continuum as the probe pulses in the subpicosecond time domain.

The sample used in our study was grown on a sapphire substrate by a metal-organic chemical vapor deposition. The active region nominally consists of 25 periods of $In_{0.11}Ga_{0.89}N$ (2nm) / $In_{0.03}Ga_{0.97}N$ (4nm) MQWs. The sample was evaluated in both the average indium concentration of the MQWs and the period of the superlattice by the use of the x-ray diffraction measurement. The absorption edge was located at the higher photon energy than the band gap of the $In_{0.11}Ga_{0.89}N$ ternary alloy and was close to the lowest confinement energy level of the $In_{0.11}Ga_{0.89}N$ (2nm) / $In_{0.03}Ga_{0.97}N$ (4nm) MQWs obtained by the calculation, as is shown in Fig. 1 (a). Thus, the two-dimensional confinement effects are present in the sample.

For the time-resolved pump-and-probe measurement, the laser source was a mode-locked Ti:sapphire laser and a Ti:sapphire regenerative amplifier. A part of the amplified laser pulses was focussed in the water to produce the white-light pulses, which were used as the probe pulses. The second-harmonics of the amplified laser pulses were used as the pump pulses. The differential absorption spectra were recorded by a spectrometer and a liquid-nitrogen cooled charge-coupled device multichannel detector. This method is very useful for the investigation of the gain formation processes, because we can directly observe the energy distribution of the photoexcited carriers by the use of this method [5].

Figure 1 (a) shows the absorption spectrum (a dotted line) and the photoluminescence (PL) spectrum (a dashed lines) at 2 K. The excitation photon energy is 3.323 eV. The excitation energy density is 20 $\mu J/cm^2$. The lower-energy tail states (localized states) due to the compositional and structural disorder is observed in the absorption spectrum. The absorption spectrum at the tail states is fitted by $D(E) = 0.56 \exp[(E-3.178\ eV) / 84\ meV]$, as is shown by a solid line. The PL spectrum has a broad emission band and a sharp one. The broad emission is a spontaneous emission and its peak position is located at 3.06 eV with a full width
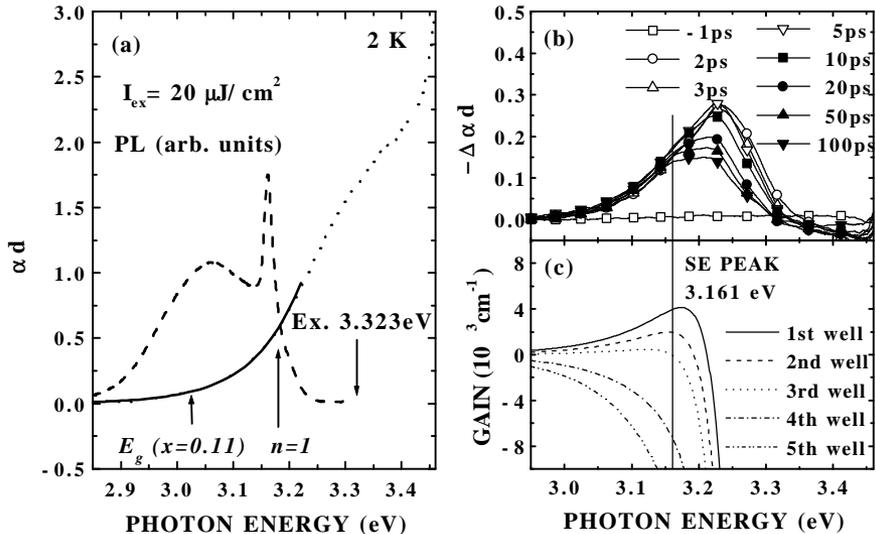
Figure 1. (a) Absorption spectrum and PL spectrum of the InGaN MQWs at 2 K. The excitation energy density is 20 $\mu J/cm^2$. The excitation photon energy is 3.323 eV. The absorption tail is fitted by $A \exp[(E-E_{me})/e]$. The arrows show the band gap of the $In_{0.11}Ga_{0.89}N$ alloy and the lowest confinement energy level of the $In_{0.11}Ga_{0.89}N$ (2nm) / $In_{0.03}Ga_{0.97}N$ (4nm) MQWs obtained by the calculation. (b) The differential absorption spectra under the excitation energy density of 20 $\mu J/cm^2$. (c) The calculated gain spectra at each quantum well in the MQWs under the excitation energy density of 20 $\mu J/cm^2$. They are calculated taking account of the depth profile of the carrier density.

at half maximum (FWHM) of about 200 meV. It shows a Stokes shift from the absorption edge. The decay time of the spontaneous emission increases with the decrease of the photon energy (0.4 2 ns), which indicates the dynamical features of the exciton localization [2,4]. The sharp emission is a stimulated emission and located at the higher-energy side of the spontaneous emission. Its peak position is located at 3.161 eV and a FWHM is 23 meV. Under the weak excitation, this emission can not be observed, while, above a stimulation threshold, its intensity increases superlinearly with the increase of the excitation energy density. The excitation intensity dependence of the PL intensity gives the stimulation threshold of 17 $\mu J/cm^2$. The decay time of this emission is shortened to be less than 30 ps due to the stimulated emission process.

The temporal changes of the absorption spectra were measured by the time-resolved pump-and-probe measurement employing the white-light pulses. Figure 1 (b) shows the differential absorption spectra as a function of time delay under the excitation energy density of 20 $\mu J/cm^2$. The differential absorption spectra show the negative signals, which means the bleaching of the absorption. Below the photon energy of 3.16 eV, the signal hardly changes because the energy relaxation at the localized states is very slow. At the higher-energy states, the signal has a drastic variety, especially within 20 ps corresponding to the decay time of the stimulated emission. Such a variety of the signal becomes conspicuous under the strong excitation, while it is small below the stimulation threshold. Thus, the variety of the signal is related to the stimulation process.

One of the interesting issues is where the optical gain is formed. The localized exciton gain model for an inhomogeneously broadened system is proposed in II-VI compound semiconductors [6]. In this model, the gain/absorption coefficient $g(E)$ can be written as

$$g(E) = D_i(E)(2f - 1).$$

Here D_i is the inhomogeneous line-shape function and f is the probability of the state being occupied. The calculated gain spectra under the excitation energy density of 20 $\mu J/cm^2$ are shown in Fig. 1 (c), in which we took account of the depth profile of the carrier density. Also we assumed the energy distribution function to be

$$f(E) = a / \{\exp[(E-b) / 0.025]+1\},$$

because the slower energy relaxation on the localized states than the stimulation process does not lead to the thermal distribution of the excitons. Here a and b are calculation parameters. We obtain the optical gain up to the third well. The energy position of the maximum gain is at the stimulated emission.

The results of the time-resolved pump-and-probe measurement clearly show the following processes. The carriers are initially formed at the delocalized states

under the interband excitation. The carriers have a high probability of the spatial migration and relax quickly into the localized states. The holes (or electrons) are captured at the trapping center due to the spatial potential fluctuation and the excitons are formed. Below the stimulation threshold, almost all the excitons go into the localized states. The energy relaxation at the localized states is very slow because of the strong exciton localization in InGaN quantum wells. On the other hand, above the stimulation threshold, the excitons exist not only at the localized states but also at the delocalized states, as a result of the state filling of the localized states. The optical gain from the localized states is formed at this stage successively. The excitons at the localized states recombine due to the stimulated emission process and the excitons at the delocalized states are supplied to the localized states successively.

In conclusion, we investigated the dynamical gain formation processes in the InGaN MQWs by means of the time-resolved pump-and-probe measurement. Based on the model of the localized exciton, we obtained the optical gain. Above the stimulation threshold, the photoexcited excitons rapidly relaxed into the localized states, and the other excitons are in the delocalized states due to the state filling of the localized states. At this stage, the optical gain from the localized states is formed, and the excitons at the delocalized states are supplied to the localized states. Thus, the stimulation mechanism in the InGaN MQWs is strongly related to the localized states.

References

1. Nakamura S. and Fasol G., *The Blue Laser Diode* (Springer-Verlag, Berlin, 1997) and references therein.
2. Satake A., Masumoto Y., Miyajima T., Asatsuma T., Nakamura F. and Ikeda M., *Phys. Rev.* B **57** (1998) pp. R2041−2044.
3. Chichibu S., Azuhata T., Sota T. and Nakamura S., *Appl. Phys. Lett.* **70** (1997) pp. 2822−2824.
4. Narukawa Y., Kawakami Y., Fujita Sz., Fujita Sg. and Nakamura S., *Phys. Rev.* B **55** (1997) pp. R1938−1941.
5. Sasaki F., Mishina T. and Masumoto Y., *Phys. Rev.* B **46** (1992) pp. 6750−6759.
6. Ding J., Hagerott M., Ishihara T., Jeon H. and Nurmikko A. V., *Phys. Rev.* B **47** (1993) pp. 10528−10542.