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Time-Resolved Photoluminescence of Cubic GaN Grown by Metalorganic Vapor Phase Epitaxy

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Time-resolved photoluminescence of high quality cubic GaN was measured to clarify the origin of the emissions observed in cubic GaN. The 3.27 eV emission decay was fast and the intensity decreased with a lifetime of 260 ps at early decay and with a lifetime of 900 ps afterwards. The decay of the 3.18 eV emission was much slower than that of the 3.27 eV emission and the lifetime was 3.8 ns. In time-resolved spectra, the 3.18 eV emission had a broadening on the high-energy side at early times and the peak moved to lower energies with increasing time. These results can be explained in terms of the model for a donor–acceptor pair transition.

1. Introduction

The III–V nitride semiconductors have been attracting much attention because of their promising potential for light-emitting device applications in blue and ultraviolet wavelength ranges. Although the stable crystal structure of the III–V nitrides, such as AlN, GaN and InN, is the wurtzite structure, they have also the zincblende structure as a metastable phase when grown under particular conditions [1]. The epitaxially grown zincblende nitride films can be easily cleaved together with the substrates, which is of great advantage to preparing cavity mirrors for laser diodes [2]. Nevertheless, it strongly depends on the growth conditions whether the cubic or hexagonal phase is grown even if cubic substrates are used [1]. Consequently, the hexagonal phase often coexists with the cubic phase and the mixing of the hexagonal phase tends to result in the inferior quality of cubic nitrides. Thus, since it has been rather difficult to grow high quality cubic nitrides, not a few physical properties of cubic GaN still remain unclear. In this study, we have measured time-resolved photoluminescence (PL) of high quality cubic GaN grown by metalorganic vapor phase epitaxy (MOVPE) in order to clarify the origin of the emissions observed in cubic GaN.

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2. Experimental

The sample used in this study was cubic GaN grown on a GaAs(001) substrate by low pressure MOVPE using trimethylgallium and 1,1-dimethylhydrazine as the Ga and N sources, respectively [3]. A 20 nm GaN buffer layer was first grown on the substrate at 575 °C and the epitaxial growth of a 1 μ m GaN was carried out at 900 °C. Time-resolved PL measurements were performed at 9 K with the time-correlated single photon counting method. The excitation source was the second harmonic generated light ($\lambda = 300$ nm, pulse width < 5 ps) of a dye laser. The time resolution for the system was 100 ps. We have also performed cw PL measurements at 6 K using a He–Cd laser ($\lambda = 325$ nm) as the excitation source.

3. Results and Discussion

Figure 1 shows low-temperature (6 K) PL spectrum of MOVPE-grown cubic GaN on GaAs (001) under steady-state excitation using a He–Cd laser. The emissions are clearly observed at 3.27 eV and 3.18 eV. From the temperature and excitation intensity dependence, the PL peaks at 3.27 eV and 3.18 eV were assigned to an excitonic transition and a donor–acceptor recombination pair transition (DAP), respectively [3].

The decay of the 3.27 eV and the 3.18 eV emissions in cubic GaN at 9 K is shown in Fig. 2. The 3.27 eV luminescence intensity decreases with a lifetime of 260 ps at early decay and with a lifetime of 900 ps afterwards. As will be shown later in time-resolved PL spectra, the 3.27 eV emission at early decay is located at a little higher energy side than at later decay. This indicates that the origin of the fast decay can be assigned to the contribution of free exciton, considering that the energy difference between free exciton and bound exciton is as small as 7 meV [4]. On the other hand, the slow decay is considered to be due to bound exciton. It should be noted that these lifetimes are comparable to the values reported for high purity hexagonal GaN (free exciton: 440 ps, donor bound exciton: 740 ps at 10 K) [5].

The decay of the 3.18 eV emission was much slower than that of the 3.27 eV emission and the lifetime was 3.8 ns. This value is much larger than that for the excitonic transition at low temperature but smaller than those reported for DAP emissions in other

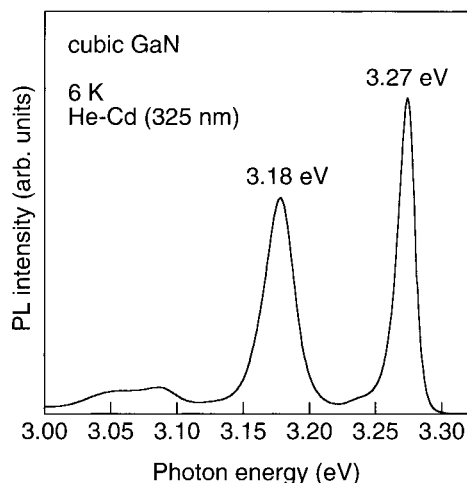


Fig. 1. Low-temperature photoluminescence spectrum of MOVPE-grown cubic GaN on GaAs(001) under steady-state excitation

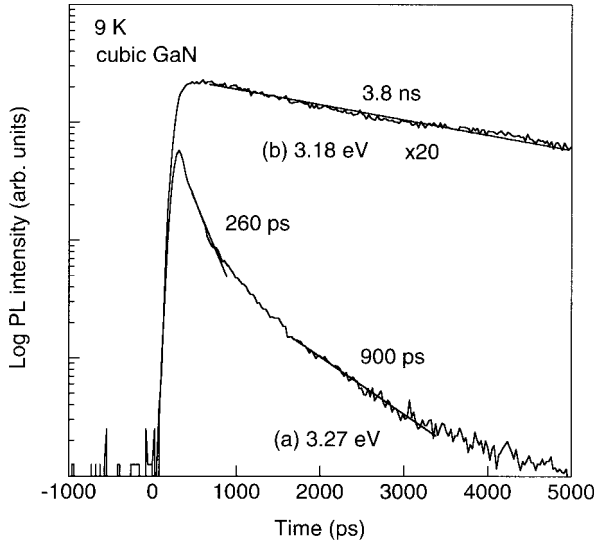


Fig. 2. Time-dependent luminescence intensity of a) the 3.27 eV emission and b) the 3.18 eV emission in cubic GaN

semiconductors [6,7]. However, this slow decay can be attributed to a DAP transition as explained later. This smaller value may be due to the influence of nonradiative recombination processes and rather high donor concentration.

Figure 3 shows time-resolved PL spectra taken from cubic GaN at 9 K. The spectra were integrally measured at the gate time of 0 to 0.5, 0.5 to 1.0, 1.0 to 1.5, 1.5 to 2.0 and 10 to 11 ns, respectively, corresponding to the curves from the upper to the lower side of the viewgraph. In the time-resolved spectra, immediately after the excitation of the pulsed laser (0 to 0.5 ns), the 3.27 eV emission is clearly observed, whereas the

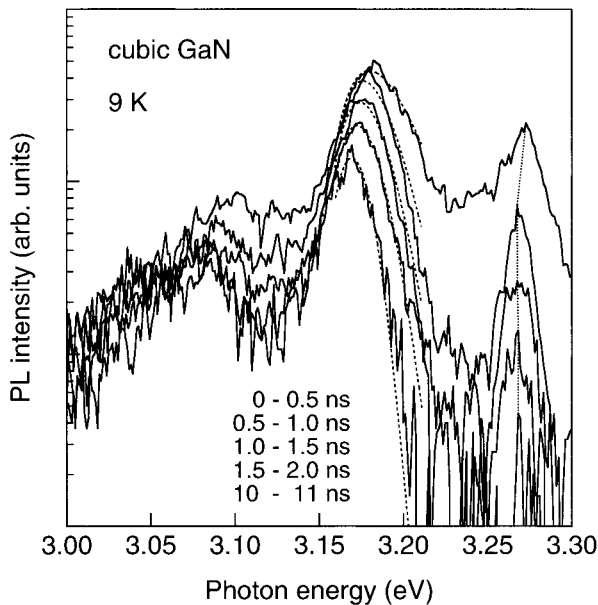


Fig. 3. Time-resolved photoluminescence spectra taken from cubic GaN

3.18 eV emission becomes dominant when the delay after the pulse excitation exceeds 1 ns. The 3.27 eV emission at the gate time from 0 to 0.5 ns is located at a little higher energy side than at later gate time, indicating that the fast decay comes from free exciton as described before. In addition, the 3.18 eV emission is found to have a broadening on the high-energy side at early times and the peak moves to lower energies with increasing time. We used the model proposed by Thomas et al. [7] to analyze the time evolution of the time-resolved PL spectra. The theoretical curves calculated based on the model were shown by dashed lines in Fig. 3. These curves agree well with the measured time-resolved PL spectra. This demonstrates that the 3.18 eV emission is due to a DAP transition.

4. Conclusions

We investigated time-resolved PL of high quality cubic GaN. The 3.27 eV emission intensity decreased with a lifetime of 260 ps at early decay and with a lifetime of 900 ps afterwards. The origin of the fast decay was assigned to the contribution of free exciton, while the slow decay was considered to be due to bound exciton. In the time-resolved spectra, the 3.18 eV emission had a broadening on the high-energy side at early times and the peak moved to lower energies with increasing time. The temporal evolution could be explained well in terms of the model proposed for a DAP transition. These results demonstrated that the 3.27 eV emission is due to excitonic transition and that the 3.18 eV emissions are due to a DAP transition.

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