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Electron-hole plasma emission from $In_{0.3}Ga_{0.7}N/GaN$ multiple quantum wells

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$In_{0.3}Ga_{0.7}N/GaN$ multiple quantum wells have been grown on the epitaxial lateral overgrowth GaN successfully. The samples have been characterized by transmission electron microscopy and x-ray diffraction, which show the high quality of the samples. Photoluminescence measurements have been carried out at room temperature, with back scattering geometry. At a low excitation power, only exciton-related emission has been observed at 3.1935 eV. With increasing excitation power, a peak appears at the low-energy side of the exciton-related emission, and becomes dominant at high excitation power. This peak has been assigned to plasma emission, because the intensity of this peak increases with excitation power as $I_{ex}^{1.9}$.

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III-V nitrides are characterized by large direct band gaps which makes them suitable for optoelectronic applications covering most of the visible range of the electromagnetic spectrum. As a consequence, a great deal of effort has been devoted to the investigation of their optical properties. It was found that the optical properties of III-V nitrides are quite different from these of other III-V compound semiconductors in many aspects. The most interesting surprise was that a high density of dislocations, typically on the order of $10^8-10^{11}\,cm^{-2}$, has little effect on the emission efficiency of the III-V nitride semiconductors due to a very short minority carrier diffusion length in these materials.[1–3] The observed unusual phenomena in the $In_xGa_{1-x}N/GaN$ heterostructures include the abnormal photoluminescence (PL) temperature dependencies,[4,5] the large Stokes shift,[6] the blueshift of PL and electroluminescence during an increase of excitation power,[7] and ''S-shaped'' temperature-dependent emission.[8] Recombination from localized states resulting from In composition fluctuation was proposed to explain these phenomena.[9–13] It was pointed out that the large piezoelectric field in quantum wells (QW's) also plays a very important role.[14–17] Many-body effects are expected to be very important in III-V nitrides because of the large exciton binding energies in these systems compared to other III-V compound semiconductors.[18–21] Reports of plasma emission from GaN under high excitation can be found in the early literature.[22–24] Many-body effects in GaN epilayers and $In_xGa_{1-x}N/GaN$ heterostructures were investigated using different methods.[25–29] A redshift of the stimulated emission observed for optical pumped GaN and $In_xGa_{1-x}N$ epilayers was interpreted as the strong many-body effects overcoming the relatively small band-filling effect.[30–34]

However, to the best of our knowledge there are no reports on the plasma emission from $GaN/In_xGa_{1-x}N$ QW's because of the following two reasons: (1) the plasma emission can only be observed in QW's with good optical confinement; otherwise the band filling luminescence arises on the high-energy side of the first level transition with an increase of the excitation power.[35] (2) As pointed out above, besides many-body effect there are many other effects, such as localized states and the quantum-confined Stark effect, which can also make the luminescence band lower than the first-level transition in $In_xGa_{1-x}N/GaN$ QW's, so it is difficult to make an exclusive assignment. Theoretically, the exciton bleaches after the formation of electron-hole (e-h) plasma. Actually, e-h plasma and exciton emission can be observed simultaneously because of the different excitation levels across the excited spot. A systematic investigation of this phenomenon was carried out in $GaAs/Al_yGa_{1-y}As$ QW's at low temperature under the excitation of a high-power pulsed laser.[35] In this paper we present the observation of plasma emission in $In_{0.3}Ga_{0.7}N/GaN$ QW's, but at room temperature under the excitation of a cw He-Cd laser.

GaN and $In_{0.3}Ga_{0.7}N/GaN$ QW's growths were carried out in a low-pressure metal-organic chemical-vapor-deposition (MOCVD) system using epitaxial lateral overgrowth (ELO) technique. First, a 2-μm-thick GaN seed layer was grown on a c-plane sapphire substrate. A 100-nm SiN was patterned into stripes, oriented in the $\langle 1\bar{1}00\rangle_{GaN}$ direction, defining a 3-μm opening with a periodicity of 13 μm. Then the sample was reloaded into the MOCVD chamber. After 4 h of ELO GaN growth on the mask, the GaN stripes grew laterally and coalesced, forming a flat surface. The obtained continuous film is called an ELO GaN. X-ray (0002) rocking curves of ELO GaN with different thickness were measured with the scattering plane perpendicular to the stripe. Three peaks were observed for a just coalesced film (4 μm thick). This means there is a tilt between the window and wing parts of ELO GaN.[36] In our case the tilt is about 0.15°. When the ELO GaN becomes thicker, peak splitting due to the tilt is no longer evident. A single peak with a full width at half maximum of 330 arcsecs was obtained for a 7-μm-thick ELO GaN. Figure 1 shows a plan-view transmission electron microscopy (TEM) picture of an ELO GaN. In GaN, the threading dislocation is always perpendicular to the surface. So, in the plan-view TEM picture, the image of dislocation is a white and black lobe. Such a lobe can be found only in the window part. This means there is no threading dislocation in the wing part. A few dislocations parallel to the stripe were

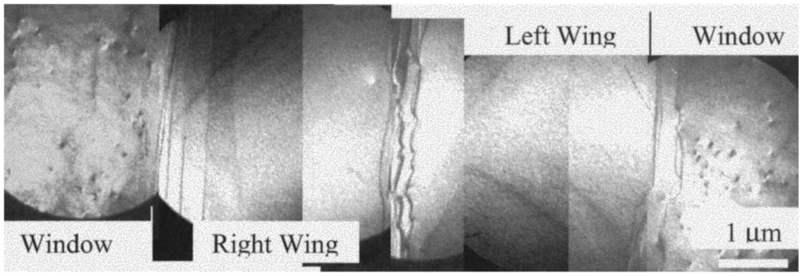

FIG. 1. Plan-view TEM picture of an ELO-GaN; $g = 11\bar{2}0$.

observed at the coalesced boundaries and window edges. These results prove the high quality of our ELO-GaN. $In_{0.3}Ga_{0.7}N$/GaN QW's used in this study were grown on a 10-μm-thick ELO GaN at 800 °C, with eight periods followed by a 20-nm-thick GaN cap layer. All layers were not intentionally doped.

$In_xGa_{1-x}N$/GaN QW's grown on ELO GaN have been characterized by x-ray-diffraction (XRD) measurement and TEM. Figure 2 presents the XRD (0002) diffraction of $In_{0.3}Ga_{0.7}N$/GaN QW's recorded in a ω-2θ mode. Besides GaN (0002) and QW's zero-order peaks, several satellite peaks can be observed. Both GaN and QW's zero-order peaks are very narrow, showing the high quality of the sample. From the position of a QW's zero-order peak, we estimated that the average In content in the QW's is about 6%.[37] This estimation was made under the assumption that an $In_xGa_{1-x}N$ layer is pseudomorphically grown on GaN, i.e., the in-plane lattice constant $a_{In_xGa_{1-x}N} = a_{GaN}$. If the $In_xGa_{1-x}N$ layer is partially relaxed, the actual value should be higher than 6%.[37] The period of the QW's measured from XRD is about 9.0 nm. Figure 3 shows a cross-section TEM picture of the sample. It is found that the $In_xGa_{1-x}N$/GaN interface is very sharp. The thicknesses of the well and barrier measured from TEM are about 1.7 and 6.9 nm, respectively, which are in a good agreement with the XRD. No indium was introduced during the barrier layer growth. Thus the In content in the well layer of this QW's is at least 30%.

Photoluminescence measurements were carried out using a micro-PL system at room temperature. The luminescence was excited by a He-Cd laser operating at an emission wavelength of 325 nm. The maximum power density obtained by tightly focusing the beam over a spot of 1-μm diameter was $I_0 = 2$ kW/cm^2. The measurements were performed under stationary condition because a cw laser was used to excite the luminescence.

The PL spectra of the QW's obtained at different excitation powers are shown in Fig. 4 on a semilogarithmic scale. The spectra were recorded in a backscattering configuration, and they were virtually unaffected by the reabsorption in the crystal. At the lowest excitation power density ($0.055I_0 = 0.11$ kW/cm^2), only an exciton-related peak corresponding to the $n=1$ heavy-hole exciton in the QW's (E_{11h}) is present around 3.194 eV. With increasing excitation power another band arises in the low-energy side of the E_{11h} peak, and becomes dominant at high excitation power, which is assigned to electron-hole plasma emission (EHP). This band shows a redshift of about 122 meV with an increase of the excitation power (I_{ex}) from $0.055I_0$ to I_0. The FWHM is of both peaks are about 100 meV, showing the high quality of the sample. The EHP band disappears when the excitation power is reduced. So any extrinsic or impurity-related emission mechanism can be unambiguously excluded. Clearly, any scattering processes cannot account for this band because of such a large redshift. The absolute value of the band-gap renormalization for the $In_xGa_{1-x}N$/GaN QW's was calculated to be 100 meV at a plasma density of 4.5×10^{12} cm^{-2} by Park and Chuang.[38] Our observation is consistent with their calculation. In addition, a weak peak from the GaN barrier and cap layer can be identified at 3.415 eV,

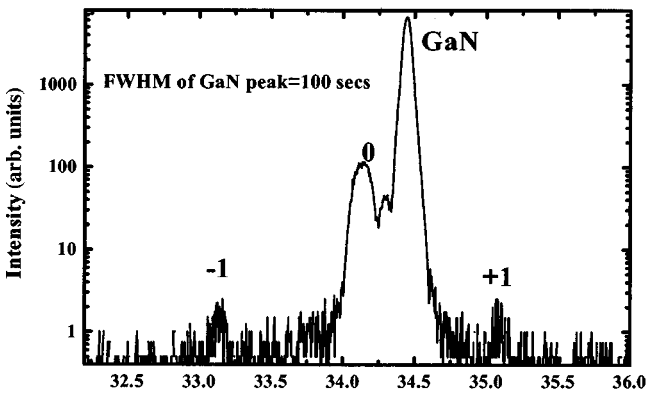

FIG. 2. XRD (0002) diffraction of $In_{0.3}Ga_{0.7}N$/GaN QW's recorded in the $\omega - 2\theta$ mode.

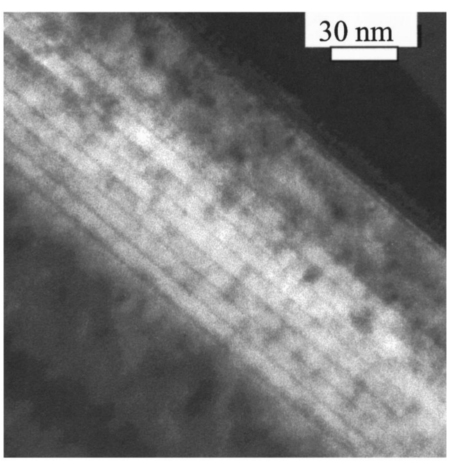

FIG. 3. Cross-section TEM picture of $In_{0.3}Ga_{0.7}N$/GaN QW's grown on an ELO GaN; $g = 0002$.

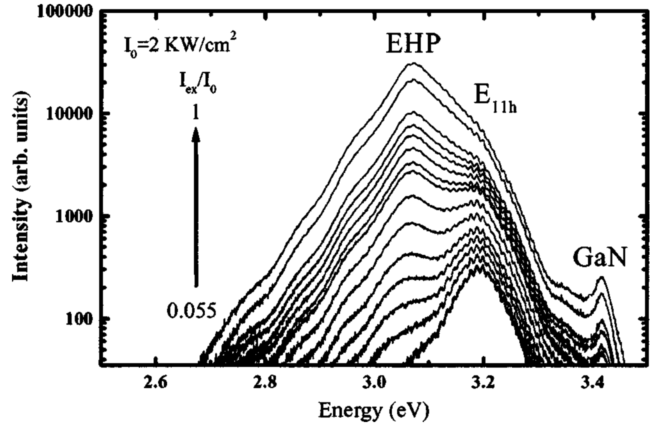

FIG. 4. PL spectra of $In_{0.3}Ga_{0.7}N/GaN$ QW's grown on an ELO GaN for various excitation powers.

indicating an efficient carrier confinement in the QW's active layer. Interferometric fringes also could be observed in the PL spectrum, which again shows good optical quality of the sample.

It is very difficult to deconvolute the measured spectra because there are so many factors that may contribute to the broadening of the emission band. The variation of the integrated emission intensity of the whole spectrum (include both EHP and E_{11h} bands) on excitation power is shown in Fig. 5. The integrated intensity increases superlinearly with excitation power. Moreover, one can find that the increased rate of the intensity also rises with the excitation power. The reason for this is that the exciton-related emission is dominated at low excitation power, whereas at the high excitation power the plasma emission band becomes dominant. The exciton-related emission is supposed to increase linearly with excitation power. The intensity of the plasma emission can be described as βI_{ex}^{γ}. We have found that the intensity of the plasma emission increases with excitation power as $I_{ex}^{1.9}$ by fitting the intensity curve with the function of $\alpha I_{ex}+\beta I_{ex}^{\gamma}$. Theoretically, for EHP emission the emission versus the excitation power should follow a power-2 law. However, the experimental result is always a little lower than 2 because of many other loss mechanisms. So the observation of power 1.9 is a strong evidence that the lower energy peak in our spectrum is due to EHP emission.

A further discussion of the position of the exciton-related emission is very necessary because of the existence of compressive stress in the $In_{0.3}Ga_{0.7}N$ well layer. It has been assumed that the 1.7-nm-thick $In_{0.3}Ga_{0.7}N$ well layers were pseudomorphically grown on GaN when the indium content was calculated from XRD measurement. This is quite reasonable, because it was recently reported that even thick (>100 nm) $In_xGa_{1-x}N$ epilayers grown on GaN are pseudomorphically strained for $x \leqslant 0.2$.[39,40] Although in the present study the indium content is a little higher, the thickness of the layer is much smaller. The energy gap of unstained $In_xGa_{1-x}N$ can be approximated by

$$E_{In_xGa_{1-x}N}(x)=(1-x)E_{GaN}+xE_{InN}-bx(1-x). \quad (1)$$

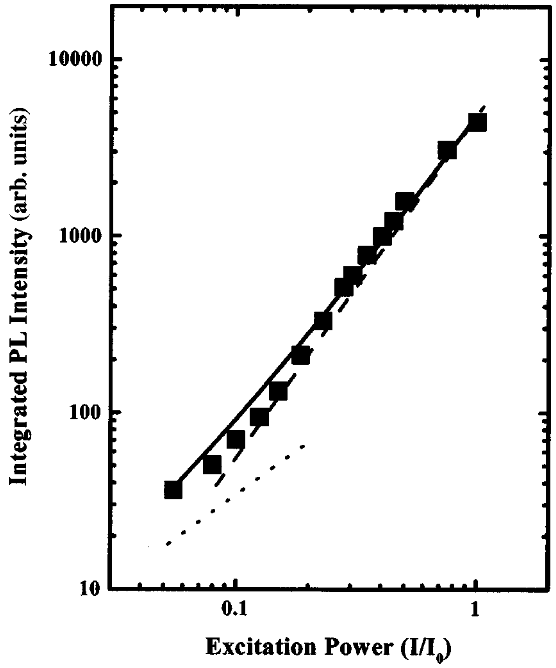

FIG. 5. Excitation power dependence of integrated emission intensity; solid squares are experiment results, the dotted line is a function of $350I_{ex}$, the dashed line is a function of $4505I_{ex}^{1.9}$, and the solid line is the function of $350I_{ex}+4505I_{ex}^{1.9}$, which is the best fit to the experimental result.

When the $In_xGa_{1-x}N$ layer's pseudomorphically grown on GaN, the blueshift induced by compressive strain is given by[40]

$$\Delta E=1.02x, \quad (2)$$

where ΔE is in units of eV. Thus the band-gap energy of $In_{0.3}Ga_{0.7}N$, pseudomorphically grown on GaN, is about 3.0478 eV, supposing $E_{GaN}=3.42$ eV, and $E_{InN}=1.89$ eV, and the bowing parameter b for $In_xGa_{1-x}N$ is 1.02 eV. The blueshift of the excitonic transition induced by quantum confinement is estimated to be 0.1637 eV for $In_{0.3}Ga_{0.7}N/GaN$ QW's, with well widths of 1.7 nm, by using the Kronig-Penny model. The quantum-confined Stark effect is weak for $In_xGa_{1-x}N$ QW's with well widths lower than 2 nm.[16] In this case, an excitonic transition around 3.2115 eV can be expected, which is in a good agreement with the present experiment results.

In conclusion, $In_{0.3}Ga_{0.7}N/GaN$ QW's with good optical confinement have been grown successfully on the ELO GaN. Both exciton-related emission and e-h plasma emission have been observed in a sample at room temperature under very low excitation power. This implies that EHP can easily arise in $In_xGa_{1-x}N$ structures, and plays a very important role in the emission. Currently, much attention and effort have been placed on the carrier localization and piezoelectric effect.[41] Our finding could probably change the way III nitride experts think about the $In_xGa_{1-x}N$ emission, and finally lead to a full understanding of the mechanism behind the emission of $In_xGa_{1-x}N$. Therefore, the observation of EHP PL in $In_xGa_{1-x}N$ multiple quantum wells is very important news to III-nitride experts, and is of broad interest.

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