Comparison study of photoluminescence from InGaN/GaN multiple quantum wells and InGaN epitaxial layers under large hydrostatic pressure

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We report the results of a comparison study of photoluminescence (PL) from an $In_{0.15}Ga_{0.85}N$/GaN multiple-quantum-well (MQW) sample and an $In_{0.11}Ga_{0.89}N$ thick epitaxial-layer sample, which have very similar band-gap energies. Large hydrostatic pressures were used for our investigations. The PL emissions in both samples were found to shift linearly to higher energy with applied pressure. In the MQW sample, the pressure response of the InGaN is dominated by the GaN layers, which leads to a significantly weaker pressure dependence as compared to the epilayer sample. Our results yield a pressure coefficient of 2.8×10^{-3} eV/kbar for the $In_{0.15}Ga_{0.85}N$/GaN MQW sample and 4.0×10^{-3} eV/kbar for the $In_{0.11}Ga_{0.89}N$ epilayer. An abrupt decrease of PL intensity in both samples was observed at pressures above 100 kbar, indicating the carriers involved in the radiative recombination processes in the samples originate primarily from the adjacent GaN layers. © *1998 American Institute of Physics.* [S0003-6951(98)01638-6]

The $In_xGa_{1-x}N$ alloy system and related heterostructures such as quantum wells (QWs) are attracting much attention because of their importance in both scientific and technological aspects. This has been manifested by recent breakthroughs in the development of high-efficiency blue light-emitting diodes and laser diodes using $In_xGa_{1-x}N$/GaN QW structures as active media materials.[1–5] A large number of studies on the optical properties of InGaN epilayers and InGaN/GaN QW structures have been reported. In particular, recent pressure-dependent photoluminescence (PL) studies on bulk-like $In_xGa_{1-x}N$ epitaxial layers have found that the pressure coefficients of the PL emission from InGaN epilayers do not substantially differ from that of GaN.[6,7] In addition, the pressure coefficient of GaN grown epitaxially on sapphire does not differ from that of bulk GaN,[8] suggesting that the highly defective region near the GaN/sapphire interface plastically deforms under applied pressure. The pressure-dependent measurements are consistent with theoretical predictions.[9] However, pressure-dependent studies on the optical properties of $In_xGa_{1-x}N$/GaN QWs showed that the pressure coefficient of the PL emission is highly variable, depending on the QW structures and In concentrations.[7,10,11]

In this letter we provide evidence that the pressure response of InGaN QWs is dominated by the GaN layers, whereas thick InGaN epilayers behave like bulk material. A comparison study was performed, using two specially prepared samples [$In_{0.15}Ga_{0.85}N$/GaN multiple quantum wells (MQWs) and a thick $In_{0.11}Ga_{0.89}N$ epitaxial layer] with approximately the same band gaps. PL measurements were obtained as a function of hydrostatic pressure using a diamond anvil cell (DAC). The effects of applied pressure on the photoluminescence transitions in the MQW sample and the bulk-like InGaN epilayer were examined and compared.

The samples used in this work were grown on sapphire substrates by metalorganic chemical vapor deposition. The MQW sample has a ten-period InGaN/GaN superlattice on a 4-μm-thick GaN layer. The thicknesses of the well and barrier are 18 and 62 Å, respectively. These values were derived from the x-ray diffraction (XRD) measurements of the superlattice period (80 Å) and the ratio of the well/barrier growth times (35/120). Rutherford backscattering spectrometry (RBS) indicates an average In concentration of 3.4% in the MQW region. If one assumes that all the In resides in the wells, then the wells have an In concentration of 15%. The $In_{0.11}Ga_{0.89}N$ epilayer thickness is around 2500 Å, and is pseudomorphically strained to the underlying 4-μm-thick GaN.[12] Its In concentration was determined by RBS and XRD measurements.

PL signals resulted from excitation by the 3250-Å line of a HeCd laser and were dispersed by a 1-M double-grating monochromator. The pressure-dependent measurements were conducted using gasketed DACs. In order to accommodate the dimensions of the DAC, small sample chips with sizes of $\sim 200\times 200\ \mu m^2$ were prepared by mechanical polishing and cutting. A small ruby chip was mounted into the DAC for pressure calibration. All the spectra reported in this work were recorded at room temperature (295 K).

Photomodulated transmission (PT) spectra of the $In_{0.15}Ga_{0.89}N$/GaN MQW sample and the thick $In_{0.11}Ga_{0.89}N$ epilayer sample are shown in Fig. 1. The derivative-like spectral signatures denoted as E_0 in both spectra are associated with the optical transitions across the band gap of the

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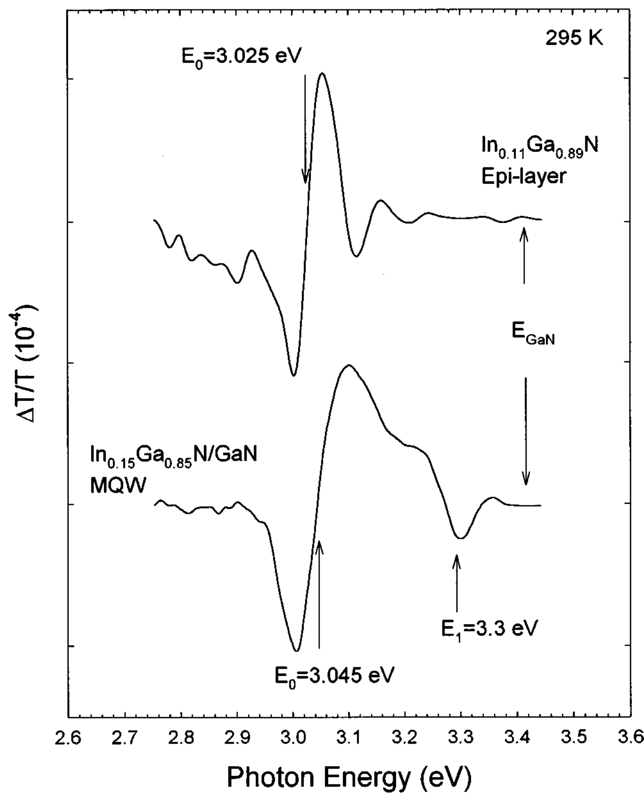

FIG. 1. Photomodulated transmission spectra of the MQW sample and the epilayer sample at room temperature.

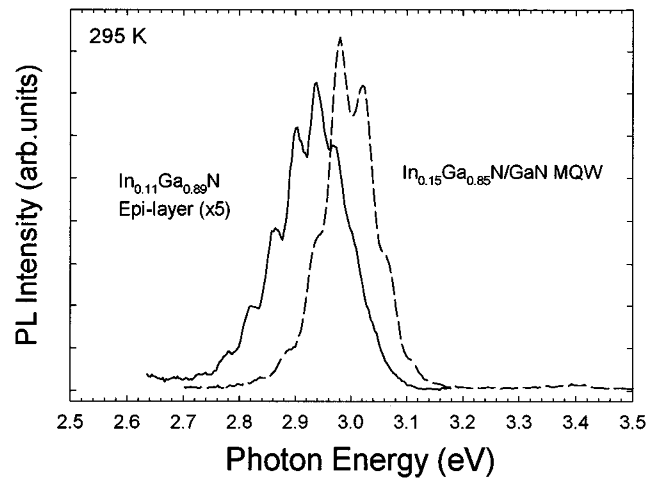

FIG. 2. Comparison of room-temperature PL spectra for the samples.

respective samples. Note that the quantum confinement effects on the electron and hole states in the MQW have shifted the band gap of $In_{0.15}Ga_{0.85}N$ to an energy approximately equal to that of bulk $In_{0.11}Ga_{0.89}N$. The second derivative-like spectral feature (denoted as E_1) in the PT spectrum of the MQW is due to transitions from ionized Mg acceptor states to the conduction-band edge in the *p*-type GaN cladding layer. The band gaps of these two samples at almost identical energy positions allow us to make an unambiguous comparison study of the PL emission and its pressure dependence from the samples.

Figure 2 shows PL spectra of the two samples at ambient pressure. The oscillatory features on the PL spectra are interference effects primarily caused by the heterointerface between the thick GaN layers and sapphire substrates. As compared to the thick InGaN epilayer, the PL emission from the MQW sample exhibits a much stronger intensity and smaller redshift (Stokes) relative to its band gap. This can be attributed to the merits of quantum confinement effects. Under hydrostatic pressure, the emissions from both samples shift to higher energy, but at different rates, as shown in Fig. 3. The inset of Fig. 3 shows the change of PL intensities with pressure. The energy position of the emission was determined by fitting its line shape to a Gaussian profile. The PL emission of the MQW sample shows a much weaker pressure dependence than that of the thick epilayer sample. From the pressure dependence of the energy change of the PL peaks (Fig. 3), we derived the pressure coefficients of the PL transitions for the samples. The solid lines in Fig. 3 are the least-squares fits to the experimental data using the linear-fit function

$$E(P)=E(0)+\alpha P, \quad (1)$$

where the energy E is in eV and the pressure P is in kbar. A best fit to the data yields a pressure coefficient of 4.0 $\times 10^{-3}$ eV/kbar for the emission from $In_{0.11}Ga_{0.89}N$. A much smaller pressure coefficient of 2.8×10^{-3} eV/kbar was determined for the $In_{0.15}Ga_{0.85}N$/GaN MQW. The result obtained from the thick epilayer sample again suggests that the pressure coefficient of the band gap of InGaN does not significantly differ from that of GaN, at least for alloys with low In concentrations.[6,7] Therefore, a drastic change of the pressure coefficient for $In_{0.15}Ga_{0.85}N$ would not be expected.

Several effects, including the increase of the electron effective mass with pressure, the mixing of QW and barrier wave functions, and the difference in the bulk pressure coefficients of the band gaps of the barriers and the wells, could influence the pressure dependence of transitions confined in QWs and give rise to smaller pressure coefficients compared to those of the well materials in bulk form.[13–19] However, a simple $\mathbf{k}\cdot\mathbf{p}$ calculation points out that the upper limit of

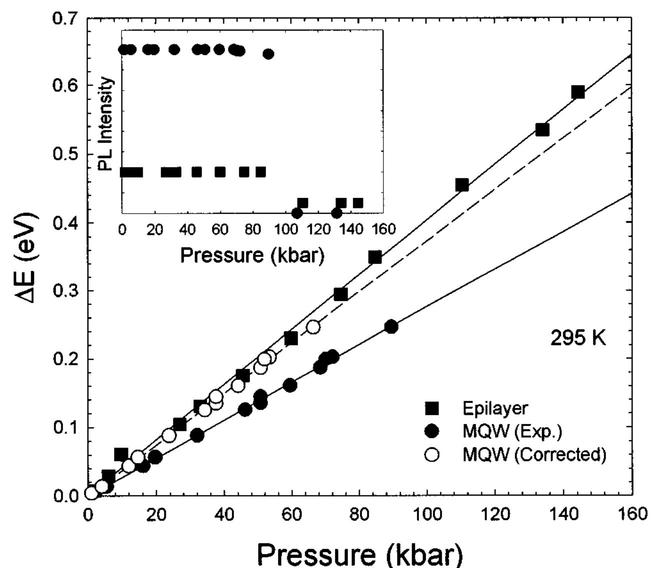

FIG. 3. Shift of the PL emission peak energies for the MQW sample (●) and the epilayer sample (■) as a function of applied pressure. The solid lines are the linear fits to the data. The open circles represent the energy shifts as a function of the effective pressure experienced by the $In_{0.15}Ga_{0.85}N$ well layers in the MQW sample. The inset shows the change of PL intensities with pressure.

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these effects can only result in an approximate 10% reduction, and cannot account for the nearly 30% difference between the observed pressure coefficients.

Application of hydrostatic pressure to strained QWs consisting of barrier and well materials with very different bulk moduli could produce uniaxial strains that make the barriers and the wells experience different effective hydrostatic and axial pressure components.[20,21] Since the MQW structure used in this work was grown on a thick GaN layer, and the GaN barriers are much thicker than the $In_{0.15}Ga_{0.85}N$ wells, the deformation of the well layers should be dominated by the compression of the stiffer GaN under hydrostatic pressure. The bulk modulus of InN ($\sim$125 GPa)[22] is approximately half of that of GaN ($\sim$210–237 GPa).[22–24] A linear interpolation gives a bulk modulus of $In_{0.15}Ga_{0.85}N$ about 7% smaller than that of GaN. Under hydrostatic pressure conditions, a tensile strain will be induced in the compressively strained $In_{0.15}Ga_{0.85}N$ well layers in the MQW structure to compensate the applied hydrostatic pressure because $In_{0.15}Ga_{0.85}N$ has a larger compressibility. As a result, the $In_{0.15}Ga_{0.85}N$ layers effectively experience a smaller hydrostatic pressure and an additional (0001) uniaxial stress. The effective pressure experienced by the well layers is estimated to be only about 74% of applied pressure. A detailed study on this will be reported elsewhere. The effect of the stress reduction caused by GaN barrier and cladding layers is also shown in Fig. 3. A pressure coefficient of 3.7×10^{-3} eV/kbar can be derived from this purely mechanical correction. Therefore, we attribute the difference in the compressibility of $In_{0.15}Ga_{0.85}N$ from that of GaN to be the major factor responsible for the significantly weaker pressure dependence of the PL transition in the MQW sample.

We note that both samples experienced an abrupt drop of PL intensity as the applied pressure rises above 100 kbar, whereas their luminescence intensities did not change much at pressures below 100 kbar (Fig. 3). While the PL intensity of the epilayer sample was found to decrease by a factor of 4, the PL signal from the MQW, in fact, was quenched completely at pressures beyond 100 kbar. This observation provides direct evidence that the PL emission from the MQW sample is dominated by contributions from the carriers photogenerated in the GaN cladding, barrier, and underlying layers. After being generated in the GaN layers, electrons and holes diffuse into the well regions and thermalize down to the lowest confined states, respectively, where they radiatively recombine to produce PL emissions. At pressures beyond 100 kbar, the band-gap energy of GaN becomes larger than the photon energy of the excitation laser line ($\sim$3.81 eV), so that very few carriers could be generated in the GaN layers. At that point, the PL emission from the MQW collapsed, indicating that the carriers created directly in the very-thin well layers do not contribute noticeably to the PL signal at room temperature. The residual PL emission in the epilayer sample above 100 kbar is primarily due to its large thickness.

In conclusion, a comparison study of photoluminescence as a function of pressure has been performed using an $In_{0.15}Ga_{0.85}N$/GaN multiple-quantum-well sample and an $In_{0.11}Ga_{0.89}N$ thick epitaxial-layer sample specially grown to have similar band-gap energies. The MQW sample was found to exhibit a significantly weaker pressure dependence as compared to the epilayer sample. An abrupt decrease of PL intensity in both samples was also observed at pressures above 100 kbar. The difference in the compressibility of $In_{0.15}Ga_{0.85}N$ and of GaN induces a tensile strain in the $In_{0.15}Ga_{0.85}N$ well layers compensating the applied hydrostatic pressure. This mechanical effect is found to be the major reason for the smaller pressure dependence of the PL transition in the MQW sample. The loss of the supply of photocarriers from GaN layers above 100 kbar is responsible for the sudden drop of the PL intensity in the epilayer sample and the quenching of the PL signal in the MQW sample at room temperature because the band gap of GaN exceeds the photon energy of the excitation light source.

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