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Photo- and cathodoluminescence investigations of piezoelectric GaN/AlGaN quantum wells

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ABSTRACT

We have examined multiple quantum well AlGaN/GaN structures with several quantum wells of varying widths. The structures had strain-free quantum wells and strained barriers. Strong piezoelectric fields in these structures led to a large red shift of the PL emission energies and long decay times were also observed. While the peak energies could be modelled using the effective mass approximation, the calculated free exciton radiative lifetimes were much shorter than those observed in experiments, indicating an alternative recombination mechanism, tentatively attributed to localised excitons. Cathodoluminescence depth profiling revealed an unusually small penetration range of electrons suggesting that electron-hole pairs preferentially remain within the multiple quantum well region due to the existing electric fields. Spatial fluctuations of the cathodoluminescence intensity were also observed.

INTRODUCTION

Advances in growth technologies of GaN/AlGaN quantum wells (QWs) made it possible to observe piezoelectricity in such structures [1-4]. In the present work the influence of the piezoelectric effects is studied in an Al_xGaN_{1-x}/GaN (with nominal content x=11.8 %) QW structure grown by MBE using a new generation RF plasma cell on sapphire covered with thick MOCVD-grown GaN. The structure contains 160 nm of GaN and then 100 nm of AlGaN was grown, followed by 2 nm of GaN (first QW), 10 nm AlGaN barrier, 4 nm of GaN (second QW), 10 nm AlGaN barrier and 6 nm wide third GaN QW with 10 nm wide upper AlGaN barrier. The structure has strain-free GaN QWs and strained AlGaN barriers.

RESULTS AND DISCUSSION

The steady-state photoluminescence (PL) is dominated by strong GaN emission from the quasi-substrate (Figure 1, bottom trace). The PL emission from GaN QWs is observed together with the yellow emission, with the maximum at 2.2 eV. The PL emission in the 2 nm QW appears above the GaN bandgap and thus shows the quantum confinement effect. The PL emission peaks from the 4 nm and 6 nm QWs are strongly red-shifted from their expected spectral positions due to a strong built-in electric field in the structure. The internal electric field is partially screened under high excitation conditions. This results in a small blue-shift of the 6 nm QW PL. This shift is more pronounced under high excitation density, under a nanosecond pulsed excitation, as is shown in Fig. 1 for excitons in the 6 nm and 4 nm wide QWs. We also observe the anomalously slow decay kinetics of excitons from the 4 nm and 6 nm QWs where the experimental PL decay curves can be fitted by two exponential curves with the two decay constants in the range of 10^{-8} – 10^{-6} s. Both time constants only weakly depend on the QW width.

THEORETICAL MODELING

The exciton characteristics in GaN/AlGaN MQWs are calculated using a simple variational approach within the envelope function approximation in a two-band model. The trial function of a quasi-2D exciton is used as

$$\varphi_{ex}(\rho,z) = f(\rho)\psi_e(z)\psi_h(z), f(\rho) = \sqrt{\frac{2}{\pi a^2}}e^{-\rho/a0}$$

ϕ with where $\psi_e(z)$ and $\psi_h(z)$ are single-particle electron and hole wave functions, and a_0 is a variational parameter.

The exciton radiative lifetime is estimated using the following equation:

$$\tau_0^{-1} = \pi a_b^3\, k_0\, \omega_{LT}\, f^2(0)\left(\int \varphi_e(z)\varphi_h(z)dz\right)^2$$

where a_B is the 3D Bohr radius, $k_0=n\omega/c$ and ω_{LT} is the exciton longitudinal-transverse splitting. We used the following values of the effective masses m_e=0.3m$_0$; m_h=1.55m$_0$, the conduction band offset was taken as 25% of the bandgap difference. The longitudinal-transverse exciton splitting was taken as 1 meV. We assumed homogeneous electric fields both in the GaN QW (F_{QW}) and in AlGaN barriers (F_b). This approach was shown to be accurate for relatively wide piezoelectric QWs [5]. The emission spectra at pulsed excitation conditions show a broad peak between 3.6 and 3.7 eV, attributed to the PL from the AlGaN barriers. This yields an independent estimate of the Al content in the barriers of 14-16%, and, according to this the value of the conduction band offset of 190 meV was used in the calculations of exciton energies. The electric field in the barrier can not be determined from the experimental data. In the calculations we kept the electric field in the barrier constant at 1x10^{6}V/cm because the results are not sensitive to the barrier field, and varied the field in the quantum wells.

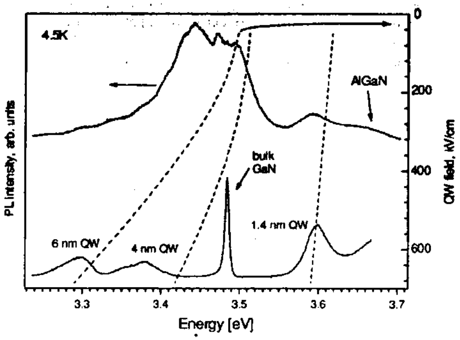

Figure 1 . Steady-state (bottom trace) and pulsed PL (top trace). Calculated exciton energies for varying electric field in QWs (broken ine, right scale).

We were able to match the low field exciton energy in the 2 nm QW to the PL peak observed with pulsed excitation, by adjusting the QW width. The best fit was obtained for the QW thickness of 1.4nm, this is about 2ML smaller than the nominal thickness and consistent with the anticipated range of thickness fluctuations. Exciton energies in wider QWs are not strongly affected by thickness fluctuations. The electric field dependence of exciton energies, is shown in Figure 1 (dashed curves). At steady-state excitation the electric field is almost the same in all the QWs of about 0.6-0.7 x 10^{6}V/cm, and possibly smaller in the narrowest QW. Under the pulsed excitation, at peak excitation intensity, the field seems to be almost completely screened. However, during the subsequent PL decay the electric field evolves around some intermediate values. This results in the PL emission peaking between 3.4 and 3.5eV.

Figure 2a-c shows the exciton intrinsic lifetimes calculated for different electric fields in the barrier F_b as a function of the field in the well, F_{QW}. A degree of correlation between the experiment and calculations is observed only for the widest QW. In this QW the electric field estimated from the excitonic energy (in the range 0.5-1x10^6 V/cm) covers the decay range 1x10^{-9} – 1x10^{-7} s, which approaches the experimental values. In the remaining QWs the calculation predicts much faster times than observed experimentally. For example, in the 2nm QW the calculated decay time is less than 1x10^{-9} s within the entire range of F_{QW}. We also note that the observed trends are opposite to those predicted by the calculations, as within the free exciton model we expect faster PL decay for narrower QWs, other parameters being equal. This suggests that the emission in the narrower QWs may have a different mechanism than recombination of nearly free excitons, such as recombination of excitons localised by thickness or composition fluctuations or some defects or impurities. In this case, the decay time can be longer compared to the calculated radiative lifetime.

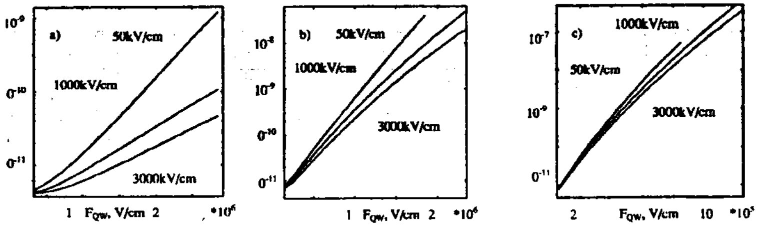

Figure 2. Calculated exciton lifetimes as a function of electric field in QWs, at several fields in the barrier in the 2nm Qw (a), 4 nm QW (b) and 6 nm QW (c).

The CL spectra shown in Figure 3 display the QW peaks coming from three QWs, as well as the emission from the substrate, clearly visible at the highest excitation conditions. We note that the excitonic emission of GaN that dominates the PL spectrum, is relatively weak in the spectra taken at lower voltages. It only begins to dominate at the accelerating voltage in excess of 20 kV and at high excitation densities (currents). The calculations of the CL depth profile and penetration depth of primary electrons under different accelerating voltages show that even at 10 kV the underlying GaN film should be penetrated by primary electrons and thus the GaN bandedge emission should be clearly observed, and, at voltages in excess of 20 kV a characteristic CL emission from sapphire should appear. These apparent deviations from the expected behaviour we relate to the presence of a strong built-in electric field in the structure strongly affecting the penetration depth of electrons.

Secondly, the emission from the deepest 2 nm wide QW first increases and than decreases with increasing voltage, which could be related to variation of depth penetration of primary electrons. If so, a similar dependence should be observed for the upper (closer to a surface) 4 nm and 6 nm QWs. However, the experimental spectra show a different behaviour. The emission intensity clearly increases with increasing excitation power (voltage x current), which we attribute to screening effects. At high excitation power, internal electric field is partly screened and the transition oscillator strength increases rapidly.

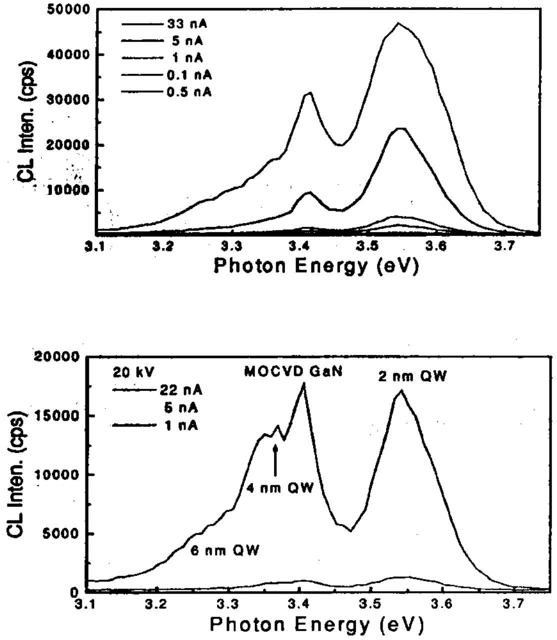

Figure 3. CL spectra at varying accelerating voltages:10 kV (top panel) and 20 kV (bottom panel)

The monochromatic CL images of the two QW emissions show an indication of small-scale grains. All CL emissions are reduced in the intensity at grain boundaries. More significant variations are observed in the spot mode CL experiment. In this study we compare relative of the QW emissions normalising the data to unity at the maximum of the 2 nm QW emission, in order to verify if the intensity fluctuations affect all three QW emissions in the same manner.

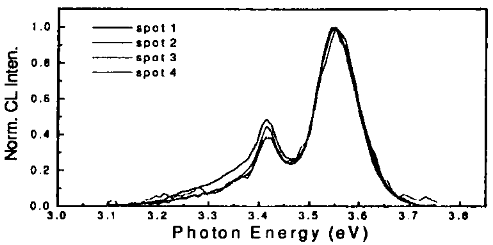

Figure 4. Normalised spot mode CL spectra at 1000x magnification.

The graphs in Fig. 4 indicate larger intensity fluctuations for 4 nm and 6 nm QW emissions. These two emissions are by far more sensitive to electric field effects and thus reflect electric field fluctuations in the plane. Such fluctuations may be due to dislocation charging. Small shifts of the emission energies are also observed.

CONCLUSIONS

In this study we have observed strong effects of the built-in electric field on the PL and CL emission from the piezoelectric GaN/AlGaN QWs. Large red shifts of the peak PL emissions are observed in the steady-state PL spectra and not in the spectra taken under pulsed excitation conditions. Long decay times of the PL emission are observed in the wider (4 nm and 6 m QWs). Theoretical calculations of excitonic energies based on the envelope function approximation match the experiment, however the observed decay times are much longer that the predicted ones. This effect is interpreted as due to exciton localisation. The depth-resolved CL spectra reveal that the penetration depth of electrons in the examined structures is much smaller than theoretically predicted in bulk materials. This suggests that the piezoelectric field affect strongly the generation and diffusion processes. Spatial fluctuations of CL intensity are more intense in wide QWs, these are attributed to in-plane fluctuations of electric field.

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