PIEZOELECTRIC FIELD EFFECT ON OPTICAL PROPERTIES OF GaN/GaInN/AlGaN QUANTUM WELLS

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Abstract:

We designed and studied two sample groups: first, GaInN/AlGaN/GaN quantum wells with asymmetric barrier structure and secondly, GaInN/GaN quantum wells with asymmetrically doped barriers. Time-resolved measurements on the asymmetric structure reveal an enhanced oscillator strength when the AlGaN barrier is on top of the GaInN quantum well, indicating a better carrier confinement in such a structure. The photoluminescence emission energy of the GaInN/GaN quantum well with doped GaN barriers shifts towards higher energy than that of undoped samples due to screening, but only when the GaN barrier layer below the quantum well is doped. In contrast, the sample where only a GaN cap layer above the quantum well is doped, shows no blue-shift. These results, showing asymmetries in GaInN/GaN quantum wells, provide confirming evidence of the piezoelectric field effect and allow us to determine the sign of the piezoelectric field, which points towards the substrate in a compressively strained quantum well. Furthermore, we performed model calculations of the global band bending and the screening effect, which consistently explain our experimental findings.

INTRODUCTION

GaInN/GaN/AlGaN-based quantum wells (QW's) have played a key role in the rapid development of short-wavelength light emitters [1]. But the recombination mechanism in the the quantum wells has been unclear and strongly controversial. For some time, the puzzling optical properties have been explained in terms of localized excitons at potential minima due to composition fluctuations or quantum-dot-like states due to phase separation [2,3,4,5]. On the other hand, a strain-induced piezoelectric field has casted a fresh light on the interpretation of the optical transitions in GaN/GaInN/AlGaN QW's [6,7,8,9]. Now, in this paper, we approach these discussions from a new view point: the piezoelectric field breaks the inversion symmetry of the quantum well. Testing the lack of the symmetry is, therefore, unambiguously related to testing the existence of the piezoelectric field. Samples designed on this purpose are classified by two groups: one with asymmetric barriers and the other one with asymmetrically doped barriers.
Our results not only verified the piezoelectric field effect, but also revealed field screening due to doping and how to enhance carrier confinement related to the direction of the field.

EXPERIMENTAL

Our samples were grown on (0001)-oriented sapphire substrates using low-pressure metalorganic vapor phase epitaxy (LP-MOVPE) and employing an AlN nucleation layer. The GaInN layers were grown below temperatures of 800 °C with N_2 as a carrier gas. Reciprocal space mapping of

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X-ray diffraction intensity shows that GaInN grown on GaN buffers is coherently strained up to thicknesses of some 100 nm [10]. In this study, two groups of samples were designed and fabricated to introduce an asymmetry into quantum wells. First, a nominally undoped 7 nm GaInN QW was sandwiched between asymmetric barrier layers, which consist of a 300 nm GaN buffer, a 60 nm GaN cap layer, and an additional 20 nm AlGaN layer below or above the quantum well. The AlN and InN mole fraction of AlGaN and GaInN layers are estimated as 15 % and 6 %, respectively. The second sample group consists of 6 nm GaInN QW's sandwiched between doped or undoped GaN barrier layers: in one sample both the GaN buffer and the GaN cap layer are doped with Si, and in another one only the GaN cap but not the buffer layer is doped. The Si-doping level is estimated as $(1 -2) \times 10^{18}$ cm^{-3}. As reference samples, 6 nm and 3 nm GaInN QW's with nominally undoped GaN barrier layers were grown. Time-resolved photoluminescence (TRPL) spectroscopy with resonant excitation of the quantum wells was performed at 5 K using a setup already described elsewhere [7].

RESULTS AND DISCUSSION

Asymmetric barrier structure

This section focuses on GaInN/GaN QW's with an additional AlGaN barrier above or below the quantum well. Low-temperature photoluminescence spectra of these two quantum wells with asymmetric barrier structure are summarized in Fig. 1. To start with time-integrated spectra (doted curves), the sample with an AlGaN barrier below the quantum well has an emission maximum at 3.060 eV, and the other one, in contrast, exhibits a broad emission band with two maxima at 3.146 eV and 3.236 eV. A more detailed picture is given by time-resolved spectra (solid curves). At short delay times, both samples show an emission line near 3.245 eV, but their temporal behaviors are clearly different afterwards: in the sample with the AlGaN barrier on top of the quantum well, an additional lower-energy emission line emerges with increasing time and finally reaches 3.146 eV, i.e. 120 meV below the emission line at early times, but the other sample with the AlGaN-barrier below the quantum well shows a dramatic red-shift by about

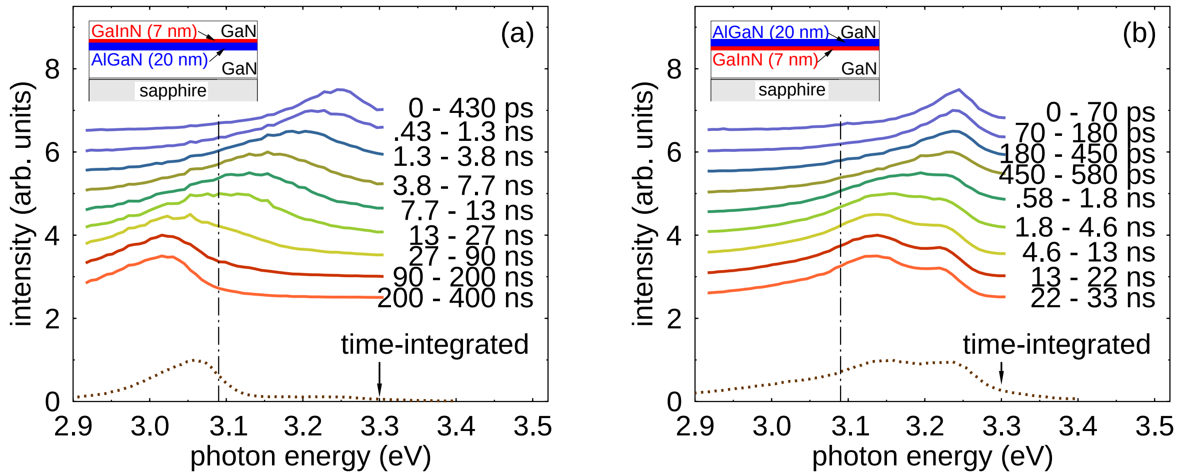

Figure 1: *Time-integrated (doted curves) and time-resolved (solid curves) low-temperature spectra of GaInN/GaN quantum wells with an additional AlGaN barrier below (a) or above (b) the quantum well. The dot-dashed lines indicate the emission energy of a simple GaInN/GaN quantum well. The insets show schematic pictures of the sample structures.*

220 meV, even 65 meV below the emission maximum of a simple GaInN/GaN QW (dot-dashed line).

Furthermore, we have a closer look at the emission lines which dominate at long delay times and measure the decay traces of each sample at the luminescence maxima in the late time interval. We find that the luminescence intensity of the sample with an AlGaN barrier above the quantum well decays much faster than that of the other one with an AlGaN barrier below the quantum well (see Fig. 2). At long delay times, we obtain a decay time of 77 ns for the former, which is about 500 times smaller than that of 34 µs for the latter.

The origin of these differences between samples can be well explained by a piezoelectric field in the quantum well: if there were no electric field in the quantum well, both samples would be identical in optical properties. The additional AlGaN barrier gives a rise to increased electron confinement and oscillator strength when it is placed where electrons are pushed by the electric field. Therefore, the shorter decay time of the sample with the AlGaN barrier on top of the quantum well indicates directly that the piezoelectric field points towards the substrate.

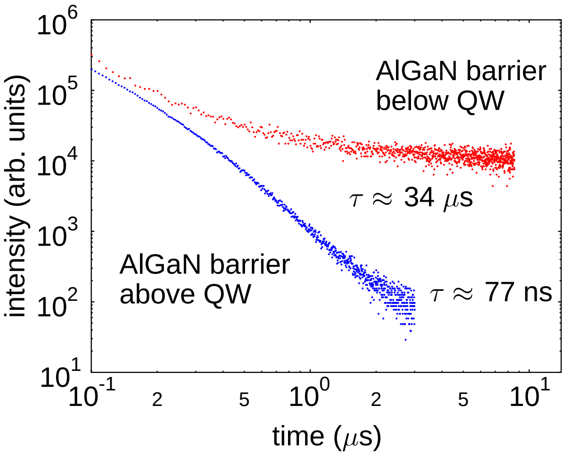

Figure 2: *Luminescence decay in asymmetric structures.*

Bearing in mind the direction of the field, we performed numerical calculations of the conduction bands to give a quantitative explanation. An electrostatic potential energy, induced by a spatially inhomogeneous electron distribution, is calculated using a solution of Poisson's equation and taken into account iteratively in solving the effective-mass Schrödinger equation. At first, we calculated the conduction band of a simple 7 nm GaInN/GaN quantum well with a piezoelectric field of 300 kV/cm. The GaN layers are assumed relaxed and have no piezoelectric field. A background carrier density of $5 \times 10^{16}\,\mathrm{cm}^{-3}$, which is estimated in nominally undoped samples, is applied. The result of the self-consistent calculation, which is depicted in Fig.3 , reveals a strong global band bending. The band in the GaN buffer layer is bent upwards, and electrons are depleted. The positive space-charge in the GaN buffer layer and electrons accumulated in the quantum well and the GaN cap layer build a electrostatic field (F_{scr}) which points in an opposite direction to that of the piezoelectric field (F_{piezo}) and screens it. Therefore, the effective field (F_{eff}) in the quantum well is smaller than the strain-induced piezoelectric field, and this calculation results in F_{eff} of 240 kV/cm, which is reduced by about 20 % of F_{piezo}.

The calculated band structures with an additional AlGaN barrier are more complex (see Fig. 4). The AlGaN and GaInN layer are assumed to be in tension and compression, respectively. Strain-induced piezoelectric fields are estimated as 350 kV/cm for the former and 300 kV/cm for the latter with an opposite direction. To take a glance at the conduction bands, the enhanced electron confinement with an AlGaN barrier above the GaInN quantum well are clearly recognizable as expected previously. A further comparison between the band structures reveals that the potential energy drop over the quantum wells is not equal. In case with an AlGaN barrier above the quantum well, a depletion region is built in the GaN buffer and cap layer. The electrons accumulated in the quantum well screen the piezoelectric fields both in the quantum well and in the AlGaN barrier. With an AlGaN barrier below the quantum well, electrons are accumulated in the GaN buffer layer, and the depletion region in the GaN cap layer screens the piezoelectric field in the AlGaN barrier but reinforces that in the quantum well. The screening effect,

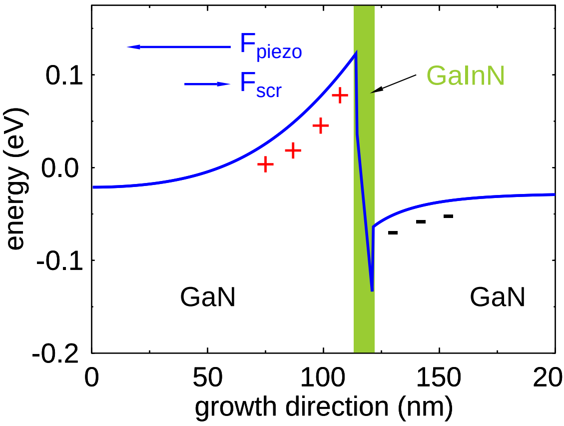

Figure 3: *Calculated conduction band structure of a GaInN/GaN single quantum well with a piezoelectric field.*

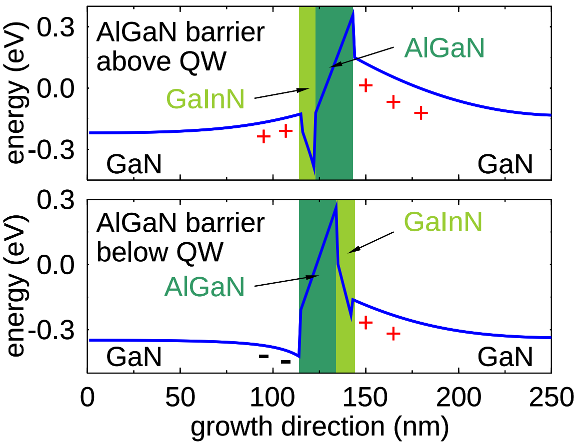

Figure 4: *Calculated conduction band structures of samples with an additional AlGaN barrier.*

therefore, depends on the placement of the AlGaN barrier: F_{eff} is calculated as 240 kV/cm and 330 kV/cm for the AlGaN barrier above and below the quantum well, respectively.

At this point, the energetic position of the emission lines can be well understood. The emission line of the sample with the AlGaN barrier below the quantum well lies about 65 meV below the emission line of the simple quantum well without AlGaN barrier. The different effective field (F_{eff}) accounts for this emission energy difference (ΔE): $\Delta E = \Delta F_{\mathrm{eff}} \times L_z$. The calculated effective field difference results in an energy shift of about 60 meV, which is well comparable with the experimental results if considering well unknown parameters, e.g. the electron concentration. On the other hand, the AlGaN barrier above the quantum well enhances the electron confinement, and therefore the increased confinement energy leads to an emission energy higher than that of the simple quantum well. The temporal red-shift of two the samples can be explained by the recovery of the piezoelectric field screened partially by carriers injected optically at early times.

Asymmetrically doped quantum well

To test in more detail the lack of the inversion symmetry due to the piezoelectric field and screening effects, we study GaInN/GaN quantum wells with varied doping in the GaN buffer and cap layer as described in the experimental section. The low-temperature photoluminescence spectra of these samples are shown in Fig. 5. The emission maximum of the 3 nm GaInN quantum well with nominally undoped GaN barrier layers lies at 2.875 eV and shifts toward 2.460 eV with increased well width of 6 nm. This energy difference allows us to estimate an effective field of about 1.4 MV/cm in the quantum well. The emission energy of the sample where only a GaN cap layer is doped is almost the same as that of the sample with undoped GaN barrier layers. In contrast, the sample doped in both the GaN buffer and cap layer shows a blue-shift of about 260 meV. We observed in addition that the stimulated emission of 6 nm quantum wells appears at almost the same energetic position, confirming that there is no significant variation of In content or well width among our samples [11].

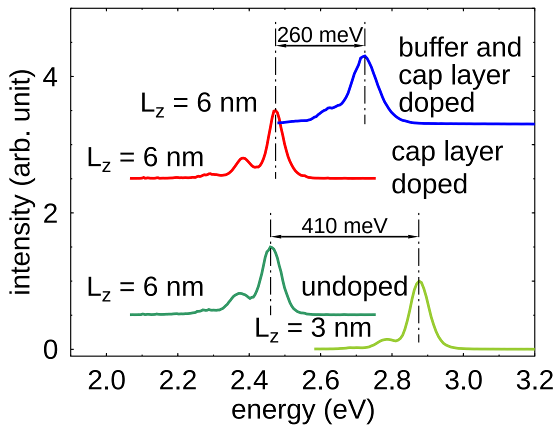

Figure 5: *Photoluminescence spectra of GaInN/GaN single QW's with and without Si-doping in the GaN barriers.*

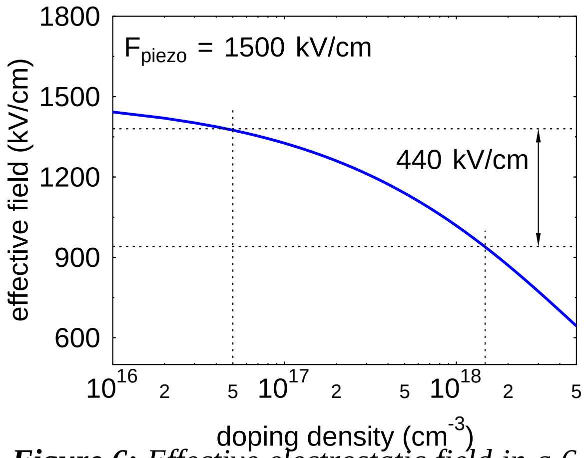

Figure 6: *Effective electrostatic field in a 6 nm quantum well versus doping density for a intrinsic piezoelectric field of 1500 kV/cm.*

These experimental results indicate that the piezoelectric field is screened only when the GaN buffer layer below the quantum well is doped. Considering the calculated band structure shown in Fig. 4, a space-charge region is built in the GaN buffer layer and induces an electrostatic field screening the piezoelectric field together with electrons accumulated in the hetero-interface between the quantum well and GaN cap layer due to the piezoelectric field. The doping level of the GaN buffer layer, therefore, has a dominant influence on the screening but not that of the GaN cap layer. It is interesting to note that this asymmetry confirms the direction of the piezoelectric field determined earlier: unless it were the case, the emission line of the sample where only the GaN cap layer is doped would shift toward lower energy.

To calculate the screening field analytically, we develop a simple model as follows. Under the abrupt approximation, the depletion-region width l below the quantum well can be expressed as $l = (2\varepsilon_0\varepsilon_r\Phi/eN_D)^{1/2}$ with a potential drop $\Phi = F_{eff}\times L_z$ over the quantum well. The doping density N_D is assumed as spatially homogeneous, and the electron concentration in the depletion region is neglected. Integrating over the depletion-region width gives the resultant electric screening field $F_{scr} = eN_D l/\varepsilon_0\varepsilon_r$. The effective field $F_{eff} = F_{piezo} - F_{scr}$ is therefore

$$F_{eff} = F_{piezo} + \frac{eN_D L_z}{\varepsilon_0\varepsilon_r}\left\{1-\sqrt{1+\frac{2\varepsilon_0\varepsilon_r F_{piezo}}{eN_D L_z}}\right\} \qquad (1)$$

where ε_0 and ε_r are the permittivity of the vacuum and the dielectric constant of GaN, respectively. For a doping density of 5×10^{16} cm^{-3}, an intrinsic piezoelectric field of 300 kV/cm, and a well width of 7 nm, an effective field is given by this equation as 243 kV/cm in good agreement with the previous result of numerical calculation. The effective fields can be therefore well calculated by this simple analytical model in a good approximation.

Now, we apply this model to the experimental results. A doping level of $(1 - 2)\times10^{18}$ cm^{-3} results in a blue-shift of about 260 meV in a 6 nm quantum well, which means a reduction of the effective field by 440 kV/cm (≈ 260 meV/6 nm) to 940 kV/cm. Fig. 6 shows the values of F_{eff} as a function of the doping density with a piezoelectric field of 1500 kV/cm according to Eq.1. Both the difference of the effective field strength and the corresponding doping density can be very well explained by the model.

SUMMARY

The asymmetric behavior of GaInN/AlGaN/GaN quantum wells provide confirming evidence of the piezoelectric field effect and allow us to determine the direction of the field which points towards the substrate. With reference to the direction, an additional AlGaN barrier on top of the GaInN quantum well enhances the electron confinement leading to increased oscillator strength. The doping level of the GaN buffer layer below the quantum well has a crucial role in screening the piezoelectric field, but not that of the cap layer above the quantum well.

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