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Suppression of the quantum-confined Stark effect in $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{Al}_y\text{Ga}_{1-y}\text{N}$ corrugated quantum wells

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We report comparative studies of 6-nm-thick $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{Al}_y\text{Ga}_{1-y}\text{N}$ pyroelectric quantum wells (QWs) grown by plasma-assisted molecular beam epitaxy on c-sapphire substrates with a thick AlN buffer deposited under different growth conditions. The Al-rich growth conditions result in a 2D growth mode and formation of a planar QW, whereas the N-rich conditions lead to a 3D growth mode and formation of a QW corrugated on the size scale of 200–300 nm. Time-resolved photoluminescence (PL) measurements reveal a strong quantum-confined Stark effect in the planar QW, manifested by a long PL lifetime and a red shift of the PL line. In the corrugated QW, the emission line emerges 200 meV higher in energy, the low-temperature PL lifetime is 40 times shorter, and the PL intensity is stronger (~ 4 times at 4.5 K and ~ 60 times at 300 K). The improved emission properties are explained by suppression of the quantum-confined Stark effect due to the reduction of the built-in electric field within the QW planes, which are not normal to the [0001] direction, enhanced carrier localization, and improved efficiency of light extraction. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4822155>]

I. INTRODUCTION

Heterostructures based on AlGaN solid alloys are currently used for ultraviolet (UV) light-emitting diodes (LEDs), which are crucial to many upcoming applications such as air/water purification, detection of different substances, and light-sensitive technologies in medicine. However the external quantum efficiency of the currently produced deep UV AlGaN LEDs with the emission wavelength shorter than 275 nm remains very low at 1%–2%,^{1–3} as compared to over 70% efficiencies of blue GaN/InGaN LEDs. The most important reasons for this are degradation of both internal quantum efficiency and light-extraction efficiency in the employed $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{Al}_y\text{Ga}_{1-y}\text{N}$ quantum wells (QWs) with high aluminum content.

The internal quantum efficiency suffers from a high density of threading dislocations generated in the buffer layers and from the quantum confined Stark effect (QCSE) induced by strong spontaneous polarization in Al-rich solid alloys grown on c-plane sapphire.⁴ Another disadvantage of the Al-rich AlGaN heterostructures is the phenomenon of switching the polarization of the emitted light from transverse electric (TE) polarization to transverse magnetic (TM) polarization as the Al content increases.⁵ The origin of this effect is the crossover of heavy-hole and split-off-hole bands in AlGaN at a certain Al composition. This circumstance defines an essential drop of the light-extraction efficiency in top-emitting c-plane UV LEDs because light propagating along the c-axis must have TE polarization. For the structures

grown on c-sapphire, the wavelength of crossover is at approximately 300 nm.⁵ In view of these problems, thorough design of the buffer, as well as of the whole heterostructure, is of particular importance.

Recently, close attention was focused on the growth of AlGaN QW structures along nonpolar and semipolar directions with diminished polarization-related electric fields inside the QWs.^{6,7} However the non-c-plane heterostructures still contain an enhanced density of nonradiative recombination centers that prevents their applications to efficient LEDs. In the polar c-plane AlGaN QWs the QCSE reduces the optical transition probability due to the polarization of electrons and holes confined in the QW. This results in an enhancement of the radiative lifetime and, hence, in a reduction of the internal quantum efficiency. The emission efficiency can be enhanced by decreasing the QW width below ~ 3 nm that partly suppresses the QCSE.⁴ On the other hand the possibility of using thicker QWs with deeper electron and hole levels could be especially advantageous in order to overcome the effect of efficiency droop in high-power LEDs.

It was previously shown that the QCSE in a GaN/ $\text{Al}_{0.2}\text{Ga}_{0.8}\text{N}$ QW can be largely suppressed by the growth on randomly textured GaN templates.⁸ In this paper, we use a similar technique to improve luminescence efficiency of $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{Al}_y\text{Ga}_{1-y}\text{N}$ QWs grown on c-sapphire, emitting near 300 nm. We demonstrate that the strength of the QCSE in such QWs can be controlled by varying growth conditions and, hence, morphology of a thick AlN buffer deposited on a c-sapphire substrate. By means of transmission electron microscopy (TEM), cathodoluminescence (CL) spectroscopy, and time-resolved (TR) photoluminescence (PL)

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spectroscopy we show that N-rich conditions during the growth of the AlN buffer result in the formation of a corrugated QW with the strongly suppressed QCSE and enhanced emission efficiency as compared to an identical reference QW structure grown with a planar buffer. In addition, it was found that the length-scale of the obtained corrugation is of the order of the emitting wavelength, which should facilitate the efficiency of light extraction.

II. SAMPLES AND EXPERIMENTAL TECHNIQUES

In this study we compare properties of two samples (A and B) with the same general design, grown by plasma-assisted molecular beam epitaxy (PA MBE) on c-sapphire substrates using different AlN buffer layers. In both samples the growth of the 2- μm -thick AlN layers was initiated at a low substrate temperature ($T_S \sim 550^\circ\text{C}$) under N-rich conditions with the group-III-to-nitrogen flux ratio ($F_{\text{III}}/F_{\text{N}}$) being about 0.6, which led to a three-dimensional (3D) growth mode. After 130 nm of growth, the substrate temperature in both samples was raised up to $T_S \sim 800^\circ\text{C}$. In sample A, the 3D growth mode was switched to the flat two-dimensional (2D) one by increasing the Al flux to ensure metal (Al) rich conditions with $F_{\text{III}}/F_{\text{N}} \sim 1.1$ – 1.2 . In sample B, the flux ratio was kept constant ($F_{\text{III}}/F_{\text{N}} \sim 0.6$) during growth of the whole AlN buffer layer. In both structures, the buffer was followed by 500-nm-thick $\text{Al}_x\text{Ga}_{1-x}\text{N}$ ($x \sim 0.45 \pm 0.03$) layers grown at a relatively low temperature $T_S \sim 700^\circ\text{C}$ and metal-rich conditions with similar values of metal and activated nitrogen fluxes. The flux ratio $F_{\text{III}}/F_{\text{N}} \sim 1.6$ corresponding to the transition from 3D to 2D growth mode was used for both samples.^{9,10} Both samples contain an AlGaIn QW with the nominal thickness of 6 nm and the aluminum content by 10 percent less than that in the barrier layer. The QW was grown by a submonolayer digital alloying technique providing high accuracy of composition control in the AlGaIn QW with respect to that in the barrier.⁹ The thickness of the capping AlGaIn barrier was 75 nm. Thus, the structures mainly differ in the growth regime of the AlN buffer. Surface morphology and growth rate of all layers were controlled *in situ* by using both reflected high energy electron diffraction (RHEED) and optical reflectometry.

High-resolution X-ray diffraction (HRXRD) measurements were performed on a D8Discover (Bruker AXS) set up with rotated anode using Cu $K\alpha$ radiation. Reciprocal space maps (RSM) were obtained with a four-bounce Ge [220] monochromator and a PSD detector LynxEye. The intrinsic crystal morphology of the samples was explored by taking cross-sectional transmission electron microscopy (TEM) images and room-temperature plan-view images of spatially resolved CL and scanning electron microscopy (SEM). Low-temperature PL spectra were measured under the conditions of cw excitation by emission of a Xe lamp, passed through a monochromator. The spectra of time-resolved PL were measured using a triple-frequency femtosecond Ti-sapphire laser (Coherent Mira 900) and a Hamamatsu streak camera. The width of the laser pulse was ~ 150 fs, the repetition rate was 76 MHz, and the typical average excitation power density was in the range 0.5 – 1.0 W/cm². The excitation wavelength was 260 nm, and the total time resolution of the PL set-up was estimated as ~ 15 ps.

III. RESULTS AND DISCUSSION

A. Structural studies and cathodoluminescence

Monitoring of the RHEED patterns revealed a striking difference between the samples: a streaky pattern was observed during the growth of the high-temperature AlN buffer layers and the AlGaIn layers in sample A, while a bright spotty pattern remained practically unchanged during the whole growth of sample B. Figure 1 shows the RHEED patterns as well as surface SEM and cross-sectional TEM images taken in samples A (Figs. 1(a) and 1(c)) and B (Figs. 1(b) and 1(d)), which evidence either 2D (sample A) or 3D (sample B) surface morphologies. These results confirm that the metal-rich stoichiometric conditions, different for the AlN and AlGaIn layers, enhance surface mobility of adatoms, leading to a 2D growth mode.⁹ One should note that the morphology of sample B remains 3D during the growth of the AlGaIn layers despite the metal-rich conditions used, which can be related to still insufficient metal-enrichment ($F_{\text{III}}/F_{\text{N}} \sim 1.6$) to flatten the AlGaIn surface for the relatively small thickness of the layers (<1 μm). The TEM images

FIG. 1. Cross-sectional TEM images of samples A (a) and B (b). Surface SEM images of samples A (c) and B (d) with insets illustrating corresponding RHEED patterns.

FIG. 2. Reciprocal space maps for the asymmetric $(11-24)$ reflections of samples A (a) and B (b).

exhibit the flat QW in the planar structure (sample A), while in sample B it is corrugated reflecting the 3D wavy morphology of the underlying layers.

Figure 2 shows the RSM's of the studied AlGaIn/AlN QW heterostructures. Analysis of these maps reveals Al contents close to $x \sim 0.45$ in the ternary barrier layers of both samples. However, these samples exhibit big difference in the degree of relaxation of the AlGaIn layers $R = 1 - (\varepsilon_{\text{meas}}/\varepsilon_{\text{coh}})$, where $\varepsilon_{\text{meas}}$ is the elastic strain value determined from the RSM analysis and ε_{coh} is the elastic strain in the fully coherent epitaxial layer.¹¹ Modeling in accordance with elastic strain theory revealed a relatively low degree of relaxation for sample A with the 2D morphology ($R = 0.25$), while sample B with the 3D morphology suffers from stronger strain relaxation ($R = 0.75$).

For both samples, the average density of screw threading dislocations in the neighborhood of the QW is in the range $(5 - 6) \times 10^9 \text{ cm}^{-2}$, while the density of edge and mixed dislocations reaches $(1 - 3) \times 10^{10} \text{ cm}^{-2}$ as counted from TEM images taken at $g = \{0002\}$ and $g = \{01-10\}$ two-beam imaging conditions, respectively.¹¹ It should be noted that in sample A the dislocations are distributed homogeneously, while in sample B they tend to bunch beneath the dips of the topographic relief rather than beneath the protrusions (see Fig. 1(b)).

This tendency is confirmed by plane-view images of spatially resolved CL (Fig. 3(a)) and SEM (Fig. 3(b)) measured at the same place of sample B. The CL wavelength corresponds to the peak of the QW emission. There is clear correlation between the CL and SEM images, confirming CL

FIG. 3. Plan-view CL (a), (c) and SEM (b), (d) images of the same place in samples B (a), (b) and A (c), (d).

enhancement when the electron-hole pairs are excited by the electron beam within protrusions (bright areas in the SEM image in Fig. 3(b)), where the dislocation density is reduced. One should note that the corresponding CL images measured in sample A are fairly uniform and featureless (Figs. 3(c) and 3(d)). To explain this observation we speculate that at the measurement temperature 300 K the spreading of excited carriers in this sample is larger than the average distance between the dislocations.

B. Continuous-wave and time-resolved photoluminescence

Figure 4(a) represents low-temperature PL spectra measured in both samples under the conditions of *cw* excitation. The spectra exhibit emission lines originating both from the QW and the barriers. In sample A, these two lines are relatively narrow and well separated, whereas in sample B the respective peaks are wider and merge together. The intensity at the QW emission line maximum in sample B at 20 K is nearly 4 times higher than in sample A. Besides, this peak in sample B is shifted toward higher energies by about 200 meV. Figure 4(b) shows the time-integrated PL spectra measured under pulsed excitation conditions at 300 K. At this temperature, the PL from the barriers is negligibly weak, whereas the integral QW PL intensity in sample B is about 60 times larger than in sample A.

The spectrally and time-integrated intensity of the QW PL line is plotted in Fig. 5 versus temperature. Below the temperatures about 120–140 K, the intensity decrease is relatively weak and similar in both samples. Above this temperature, the PL intensity in sample A falls much faster, being at 300 K about 140 times weaker than at 4.5 K. In sample B, the respective PL reduction is only seven-fold.

Figure 6 represents PL decay curves measured at the peaks of the QW emission in both A and B samples. At low

FIG. 4. PL spectra measured in samples A and B under *cw* excitation at 20 K (a) and under pulsed excitation at 300 K (b).

temperatures below ~ 140 K, the PL decay in sample A is rather long. One can see in Fig. 6(a) that a significant part of the PL signal does not decay during the period between the laser pulses (~ 13 ns). While precise determination of the decay parameters is hampered by the relatively high repetition rate of the laser pulses, one can be sure that the respective decay time constant is in the range of tens of nanoseconds.

FIG. 5. Time- and spectrally integrated QW PL intensity versus temperature in sample A (empty circles) and B (filled squares). The curves are normalized to the intensity obtained at the lowest temperature 4.5 K.

FIG. 6. Decay curves measured at the QW PL maximum in sample A (a) and B (b) at three selected temperatures: 4.5 K, 140 K, and 300 K.

Dashed lines in Fig. 6 show the results of fitting performed in the model of two-exponential decay, taking into account contributions from multiple laser pulses. According to this simulation nearly 30% of the population of photoexcited carriers decays at 4.5 K with the characteristic decay constant as large as ~ 20 ns. We believe that this decay constant represents at low temperature the radiative lifetime of localized excitons, which are responsible for the QW emission. When the temperature increases above ~ 140 K, the PL decay gradually becomes faster, and at 300 K the dominant decay time constant is as low as about 130 ps. Note that at all temperatures the decay curves cannot be fitted by a single decaying exponent and even two-exponential fitting is at some temperatures not satisfactory. In sample B the PL decay is much faster; at low temperatures most of the signal decays within one period of the excitation pulses with the dominant characteristic decay constant about 500 ps.

One can see in Fig. 6 that the decrease of the time-integrated PL intensity with temperature can be separated into two contributions. First, the temperature rise causes an increase of the non-radiative recombination rate $\Gamma_{nr} = \frac{1}{\tau_{nr}}$ and, hence, the reduction of the internal quantum yield $Y = \frac{\Gamma_r}{\Gamma_r + \Gamma_{nr}}$, where $\Gamma_r = \frac{1}{\tau_r}$ is the radiative recombination rate and τ_{nr} and τ_r are, respectively, the non-radiative and radiative decay time constants. Besides, the increase in temperature results in a certain quenching of the PL intensity immediately after excitation, which can be attributed to

temperature variation of the transport efficiency of photoexcited carriers through the thick AlGaIn cap layer. This interpretation correlates with the quenching behavior of the time-integrated PL shown by Fig. 5. Below ~ 140 K Γ_{nr} is much smaller than Γ_r for both samples, and the PL quenching is governed by the same mechanism connected to carrier losses during the transport through the AlGaIn barrier. At higher temperatures, Γ_{nr} becomes comparable to Γ_r (or less) that leads to the decrease in the internal quantum yield. The important factor defining faster quenching of the time-integrated PL in sample A is the longer radiative lifetime.

C. The quantum confined Stark effect and exciton localization

The observed long lifetime of photoexcited carriers in sample A can be explained by simultaneous action of two factors, namely, the QCSE in a relatively wide QW, and localization of carriers preventing their diffusion toward nonradiative recombination centers, at least at low temperatures. To estimate the radiative lifetime of excitons in the pyroelectric QW we adopt a variational approach with the trial envelope wave function of the ground s-like exciton state expressed as^{12,13}

$$\Psi_{ex}(\rho, z_e, z_h) = \frac{1}{\sqrt{N}} \varphi_e(z_e) \varphi_h(z_h) \times \exp\left(-\sqrt{\rho^2 + \sigma^2(z_e - z_h)^2}/\lambda\right). \quad (1)$$

Here z is the coordinate along the growth axis, $z_e(z_h)$ is the z coordinate of the electron (hole), $\rho = \sqrt{(x_e - x_h)^2 + (y_e - y_h)^2}$ is the coordinate of the relative motion of the electron and hole, φ_e and φ_h are single-particle wave functions formed by linear combinations of Airy functions, and N is a normalizing constant. Equation (1) includes two variational parameters, λ and σ ; λ corresponds to the exciton Bohr radius and σ is a measure of the exciton dimensionality. The radiative lifetime of the exciton is calculated as

$$\tau = \frac{1}{\pi a_B^3 k_0 \omega_{LT} |\Phi(0)|^2}, \quad (2)$$

where a_B and ω_{LT} are the exciton Bohr radius and longitudinal-transverse exciton splitting in the bulk QW material, k_0 is the wave vector of the emitted photon, and the electron-hole overlap $\Phi(0)$ is given by

$$\Phi(0) = \int dq \Psi(\rho = 0, z_e = q, z_h = q). \quad (3)$$

Material parameters in the calculation were taken from Refs. 14–16. The electric field was taken as homogeneous both in the well and barriers.

The calculation is complicated by the poor knowledge of many band parameters of AlGaIn ternary alloys with high content of Al, such as the deformation potentials and the band-gap bowing factor.¹⁴ Therefore, we preferred to

estimate the QW band line-ups from the experimental low-temperature PL spectra. In particular, the band gap energy $E_{g,b}$ of the $\text{Al}_y\text{Ga}_{1-y}\text{N}$ barriers was estimated from the exciton resonant energy as $E_{ex} = E_{PL} + 0.6\Delta E_{PL}$, where E_{PL} is the energy of the PL peak and ΔE_{PL} is its full width at half maximum.¹⁷ Then the band gap energy is calculated by adding the exciton binding energy obtained by linear interpolation between the known values for GaN and AlN for $y = 0.45$ that is the technologically intended composition. Furthermore, the difference in the band gap energy between the QW and the barriers, corresponding to the 10% difference in the compositions, was taken constant, since this quantity only weakly depends on the strain and composition variations. A small arbitrary value 10^3 V/cm was used in this calculation as the barrier electric field, since we assume that the polarization-induced electric field in the thick barriers is much weaker than in the thin QW.⁴ Actually, it was found that the exciton parameters only weakly depend on the barrier electric field in a wide range between zero and 10^4 V/cm.

According to the performed calculations, the exciton energy matching the position of the experimentally observed PL peak is obtained provided that the QW intrinsic electric field is as high as 7×10^5 V/cm. One should note that the estimation of the intrinsic electric field in this pyroelectric structure, performed in the simplest model of an undoped QW,⁴ provides a comparable value 7.5×10^5 V/cm. The respective calculated radiative lifetime of excitons was 15 ns. Hence, we conclude that both emission photon energy and lifetime of the QW PL in sample A is governed by the strong QCSE induced by the spontaneous and piezoelectric polarizations, in perfect agreement with the structure design.

In spite of the same nominal thickness of the QW and the same intended AlGaIn compositions in samples A and B, the PL lifetime in the latter sample is shorter, the PL intensity is larger, and the PL line position is shifted toward higher energies. All these observations are consistent with a considerable damping of the QCSE in this structure. Indeed, the energy of the QW PL line in sample B corresponds to the QW electric field $\sim 2.6 \times 10^5$ V/cm and to the exciton radiative lifetimes ~ 100 ps. This decay time constant is smaller than the experimentally obtained value ~ 500 ps. One should note nevertheless that the PL from barriers in both samples demonstrates very similar decay time, which implies that the dominant effect defining the decay of the QW PL in this sample is exciton localization by potential fluctuations rather than the QCSE in an ideal plane QW.^{18,19} Following Ref. 8 we tentatively attribute the observed reduction of the intrinsic electric field in the corrugated QW of sample B to deviation of the normal to the QW planes from the [0001] direction of the polarization vector. The respective suppression of the QCSE should be favorable for the internal quantum yield, especially at elevated temperatures facilitating nonradiative recombination.

One more specific feature of sample B is broadening of the PL peaks as well as emergence of a tail of emission and additional PL lines at lower energies (see Fig. 4). It could be explained by enhanced compositional inhomogeneity in both barrier and QW layers. Such inhomogeneities in the

$\text{Al}_x\text{Ga}_{1-x}\text{N}$ layers with the middle-range Al content ($0.25 < x < 0.5$), grown by PA MBE, can originate from a considerable difference in surface mobility of Ga and Al adatoms in the middle growth-temperature range ($700\text{--}750^\circ\text{C}$).^{20–22} However, the reduction of the growth temperature below 700°C and maintaining the slightly metal-rich growth conditions, realized for the AlGaIn growth in both A and B samples, should yield homogeneous alloys, as it occurred in the case of sample A. Thus, one can conclude that much stronger alloy inhomogeneity in sample B is generally induced by the 3D surface morphology inherited from the AlN buffer layer. The strong carrier localization caused by the enhanced compositional fluctuations is also advantageous for increasing the intrinsic quantum yield since it prevents the carriers from moving to the nonradiative recombination centers.³

In addition to the suppression of the QCSE and the enhanced carrier localization, the PL enhancement in the corrugated QW can be partly attributed to the increase of light extraction efficiency. Indeed, only less than 4% of the emitted light can be extracted from an AlGaIn structure with flat interfaces due to the effect of total internal reflection. Random surface corrugation occurring on the length scale of the emitting wavelength leads to escape-angle randomization increasing the photon escape probability.⁸ To estimate relative efficiencies of these three mechanisms of the PL enhancement we compare the QW PL intensity in both samples at different temperatures. The integrated PL from the corrugated QW is ~ 4 times stronger at 4.5 K and ~ 60 times stronger at 300 K as compared to that from the planar QW. Since the effect of the roughness-induced enhancement of the light extraction efficiency should not depend on temperature, we conclude that most of the PL enhancement at elevated temperatures occurs due to combined effects of the suppressed QCSE and the enhanced carrier localization.

IV. CONCLUSIONS

We have shown that a single $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{Al}_y\text{Ga}_{1-y}\text{N}$ QW grown by plasma-assisted MBE on a c-sapphire substrate with an AlN buffer replicates the morphology of the buffer. Al-rich growth conditions lead to 2D buffer morphology and formation of a planar QW. On the other hand, N-rich growth conditions facilitate realization of 3D buffer morphology and results in a corrugated QW.

This corrugated QW demonstrates significantly improved emission properties. In particular, the QW emission line in this structure is shifted toward higher energies and the emission intensity is enhanced compared to the planar QW by a factor of 4 at 4.5 K and a factor of 60 at 300 K. One reason behind the improved emitting characteristics is the effect of suppression of the polarization induced electric fields, unambiguously demonstrated by time-resolved PL measurements and supporting variational calculations of the exciton energy and radiative lifetime. Additionally, we spec-

ulate that the internal quantum efficiency of the corrugated QW can benefit from stronger carrier localization caused by the enhanced compositional fluctuations in the AlGaIn heterostructure with rough 3D morphology. Further work is needed to clarify relative contributions of the two enhancement mechanisms. We also deduced from the temperature dependent measurements that the enhancement of the light extraction efficiency expected for the structure with rough surface hardly exceeds a factor of four in the studied sample.

The observed increase in the PL intensity associated with the enhancement of the internal quantum efficiency makes the structures with corrugated wide AlGaIn QWs very promising for development of high-power deep UV LEDs.

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