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Optical properties of GaN/AlGa_N QW nanostructures with different well and barrier widths

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Abstract

Optical properties of wurtzite AlGa_N/Ga_N quantum well (QW) structures grown by molecular-beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD) on *c*-plane sapphire substrates have been investigated by means of photoluminescence (PL) and time-resolved photoluminescence (TRPL) at low temperature. The PL spectra exhibit a blue-shifted emission of GaN/AlGa_N QW nanostructures by decreasing the barrier width, in contrast to the arsenide system (Pabla A S *et al* 1993 *Appl. Phys. Lett.* **63** 752). This behavior is attributed to a redistribution across the samples of the huge built-in electric field (several hundreds of kV cm⁻¹) induced by the polarization difference between wells and barriers. The trend of the barrier width dependence of the internal polarization field is reproduced by using simple electrostatic arguments. In addition, the effect of well width variation on the optical transition and decay time of GaN multiple quantum wells (MQWs) have been investigated, and it has been shown that the screening of the piezoelectric field and the electron-hole separation are strongly dependent on the well thickness and have a profound effect on the optical properties of the GaN/AlGa_N MQWs. The time-resolved PL spectra of 3 nm well MQWs reveal that the spectral peak position shifts toward lower energies as the decay time increases and becomes red-shifted at longer decay times.

(Some figures in this article are in colour only in the electronic version)

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1. Introduction

The group-III nitride compounds and their alloy systems are of great interest for optoelectronic applications such as wide band-gap light-emitting diodes (LEDs) and injection lasers as well as for high-frequency and high-temperature electronics [1]. The recent developments in the field of GaN-based blue–UV optoelectronic devices are stimulating several experimental and theoretical studies. The reason for this great interest depends either on the fact that a mature technology is still to come, or the lack of a complete comprehension of all the physical processes involved in the interaction between light and nitride semiconductors. Several recent experimental results have shown a strong dependence of the photoluminescence (PL) decay time and PL peak energy in GaN-based quantum wells (QWs) on the well and barrier thickness [2–8]. The common picture used to interpret such findings is based on the action of the electric built-in field that, separating the electron and hole wavefunctions, dramatically reduces the oscillator strength. However, nonexponential decays are observed at low T , and the PL efficiency strongly decreases as the temperature is raised from 10 K up to room temperature. These observations suggest that there is a complex interplay between radiative and nonradiative transitions. In particular, it must be noted that the built-in field causes a quantum-confined Stark effect which strongly decreases the radiative rate so that, even in state-of-the-art samples for wells larger than a few tens of angstroms, the nonradiative recombination rate becomes comparable to the radiative one.

In this paper, we present a combined theoretical and experimental analysis to describe the interplay between the polarization field, charge screening, and the effect of well and barrier widths on the optical emission of a set of GaN/AlGaIn nanostructures.

2. Samples and experimental set-up

We have studied two sets of GaN/AlGaIn multiple quantum wells (MQWs) and single quantum wells (SQWs) by means of the photoluminescence (PL) and time-resolved PL (TRPL) technique. In the first group, MQW structures were grown on (0001) sapphire substrates by metal-organic chemical vapor deposition (MOCVD) [9, 10]. On top of the substrate, a 20 nm thick low-temperature-grown AlN nucleation layer and a thick nominally undoped GaN buffer layer (about 2 μm) were grown, followed by a multiple QW structure with five periods of GaN QW layers with different widths in different samples (nominally 1.5, 3 and 4.5 nm wide, respectively) separated by 7 nm wide $\text{Al}_{0.07}\text{Ga}_{0.93}\text{N}$ barriers. No additional capping layer was present, i.e. the outermost QW barrier faces the surface. The second group consists of three $\text{Al}_{0.17}\text{Ga}_{0.83}\text{N}/\text{GaN}$ SQW and MQW structures with different barrier widths. The samples were grown on (0001) sapphire substrates by molecular-beam epitaxy (MBE) using NH_3 as a nitrogen precursor on a 3 mm thick MBE-grown GaN template. The structure and other information about all the samples discussed in this paper are given in table 1.

The PL measurements were performed at temperatures from 2 to 300 K in an Oxford bath cryostat. For optical excitation in the PL measurements we have used the continuous-wave (cw) 266 nm fourth harmonic of a Nd:vanadate laser. The PL signal was dispersed by a single-grating monochromator and detected by a liquid-nitrogen-cooled charge-coupled device. For the transient data the third harmonic ($\lambda_{\text{exc}} = 266$ nm) of a femtosecond Ti:sapphire laser was used as an excitation source. A Hamamatsu syncroscan streak camera detected the signal with a time resolution better than 10 ps.

3. Results and discussion

The PL spectra for a set of GaN/ $\text{Al}_{0.17}\text{Ga}_{0.83}\text{N}$ MQWs samples with different barrier width are illustrated in figure 1. Figure 1(a) shows the PL spectrum of a 4.1 nm wide GaN SQW

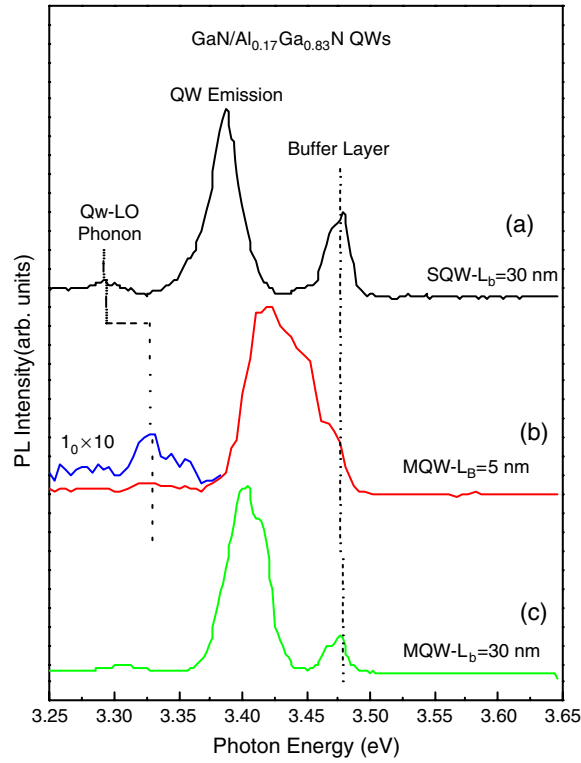


Figure 1. PL spectra at 2 K of an $\text{Al}_{0.17}\text{Ga}_{0.83}\text{N}/\text{GaN}$ single quantum well (a) and $\text{Al}_{0.17}\text{Ga}_{0.83}\text{N}/\text{GaN}$ multiple quantum wells with barrier widths of 5 nm (b) and 30 nm (c). All these structures have the same well widths of 4.1 nm.

Table 1. Detailed description of the samples studied.

Sample type	Number of QWs	Well width (nm)	Barrier width (nm)	Al composition, x
$\text{GaN}/\text{Al}_x\text{Ga}_{1-x}\text{N}$ SQW	1	4.1	30	0.17
$\text{GaN}/\text{Al}_x\text{Ga}_{1-x}\text{N}$ MQW	4	4.1	30	0.17
$\text{GaN}/\text{Al}_x\text{Ga}_{1-x}\text{N}$ MQW	4	4.1	5	0.17
$\text{GaN}/\text{Al}_x\text{Ga}_{1-x}\text{N}$ MQW	5	1.5	7.2	0.07
$\text{GaN}/\text{Al}_x\text{Ga}_{1-x}\text{N}$ MQW	5	3.0	7.2	0.07
$\text{GaN}/\text{Al}_x\text{Ga}_{1-x}\text{N}$ MQW	5	4.5	7.2	0.07

embedded in 30 nm $\text{Al}_{0.17}\text{Ga}_{0.83}\text{N}$ barriers. The PL intensity maximum peaks at 3.39 eV. Though the well is rather narrow (4.1 nm), its energy is significantly lower than the excitonic band gap of GaN, typically located at 3.478 eV in our samples.

This large red-shift (about 90 meV) between the SQW emission and the buffer layer in figure 1(a) is the first hint of the strength of the electric field in the QW, and is also related to quantum-confined Stark effect. Now, when considering an MQW structure which contains four wells of the same width separated by 5 nm wide barriers, one observes a strong blue-shift, 35 meV, of the PL energy (figure 1(b)). On the other hand, when increasing the barrier width up to 30 nm in the MQW structure, the PL energy red-shifts again (figure 1(c)). This behavior is the opposite of what is observed in standard semiconductor systems such as (Al, Ga)As/GaAs

MQWs. In this latter case, when decreasing the barrier width the QW ground-state energy of both holes and electrons decreases due to the interwell coupling of the confined wavefunctions. However, such a blue-shift related to the barrier width decrease has already been reported in the case of strained grown (Ga, In)As/GaAs MQWs [11]. As shown below, it is related to the presence of polarization fields that are very strong in wurtzite nitride heterostructures, distributed among wells and barriers that cause an efficient quantum-confinement Stark effect (QCSE). Straightforward electrostatic arguments account well for the behavior observed in figure 1. From the conservation of the electric displacement vector across the heterostructure

$$\vec{\nabla} \cdot \vec{D} = 0 \quad (1)$$

one gets the relation

$$\varepsilon_w E_w - \varepsilon_b E_b = P_b - P_w \quad (2)$$

where $\varepsilon_w(\varepsilon_b)$ is the well (barrier) static dielectric constant, $E_w(E_b)$ is the electric field in the well (barrier), and $P_w(P_b)$ is the zero-field polarization of the well (barrier) material [12]. There are two extreme cases where simple relations are obtained. The first one is that of an SQW in infinite barriers: the fields due to interfacial charges cancel each other in the barriers and

$$E_w = (P_b - P_w)/\varepsilon_w. \quad (3)$$

The second case is that of a periodic structure. The periodic boundary condition $L_w E_w + L_b E_b = 0$, where L_w and L_b are the well and barrier widths, leads to [12]

$$E_w = L_b(P_b - P_w)/(L_b + L_w)\varepsilon\varepsilon_0 \quad (4)$$

and similarly for the field in the barrier (in the (Al, Ga)N/GaN system, the dielectric constant mismatch is negligible) [13] we find

$$E_b = L_w(P_w - P_b)/(L_w + L_b)\varepsilon\varepsilon_0. \quad (5)$$

The arguments given above qualitatively explain the results in figure 1: on comparing equations (3) and (4) we discover that the electric field is larger in an SQW (figure 1(a)) than in an MQW with thin barriers; therefore, MQW PL spectra show a blue-shifted emission peak in contrast to SQW emission due to a smaller quantum-confinement stark effect (QCSE) (figure 1(b)), and in MQWs the larger the barrier, the larger the field in the well, following equation (4) (figures 1(b) and (c)). So we observe a clear red-shift in the MQW emission peak with 30 nm barrier width in comparison to an MQW structure with 5 nm barrier width due to increased internal electric field that causes a stronger quantum-confinement Stark effect (QCSE).

On the other hand, in this paper we have investigated another set of GaN/AlGaIn MQWs with different well widths to measure the effect of the well widths on the PL spectra of these structures. In the 1.5 nm MQWs sample the excitonic doublet transition energy peak position at 2 K (3.57 and 3.58 eV) is blue-shifted with respect to GaN buffer layer by an amount of 90 and 100 meV, which is due to well-known effect of quantum confinement of electrons and holes (figure 2). We tentatively interpret this peak splitting as evidence of two QW widths dominating the PL spectrum, consistent with the idea that the interface roughness is of the order one or two monolayers (2 ML = 0.52 nm) [14]. The temperature dependence of the PL intensity of the two peaks also supports this idea: at higher temperatures a thermal excitation of the lower-energy peak into the higher-energy one is observed (figure is not shown).

The assignment of the various peaks to specific well widths is assisted by a computation of the exciton energy for these thicknesses using the explicit expressions in [15], as shown in figure 3. In semiconductor heterostructures and QWs, e.g. GaN/AlGaIn, the band discontinuities that are the origin of QWs for the valence and conduction band are at most of the

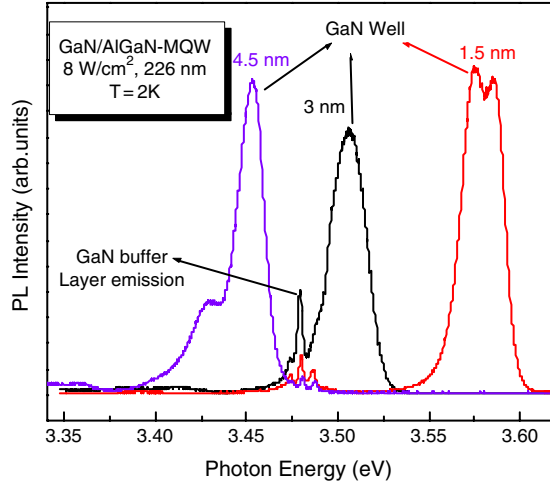


Figure 2. PL spectra of the undoped GaN/Al_{0.07}Ga_{0.93}N MQWs with different well widths measured at 2 K.

order of few tenths of a meV, and the infinite well is not a good approximation. In this paper, the QW structures consist of 7% Al in the barriers, which means that the conduction and valence band offsets are about 116 and 62 meV respectively. Then the infinite-well approximation should be modified. For instance, the well width can be replaced by the effective length, L_w^{eff} , of the QW. Singh employed the nearly free electron approach in periodic structures to provide accurate energy level expressions for the electronic ground state in a square quantum well with and without an applied transverse electric field [15]. He obtained analytical results, which are quite accurate for shallow wells where the infinite barrier solution is invalid.

In this work, we used the same analytical results by considering a very shallow conduction band well corresponding to GaN/Al_{0.07}Ga_{0.93}N QW structures. The first energy level of a very shallow square quantum well under an applied transverse electric field is given by

$$E_1 = \frac{5\Delta E^2}{2\pi^2} - \frac{\pi^2\hbar^2}{4m^*L_w^2} \left(\left\{ 1 + 32 \left[\frac{5\Delta E^2}{2\pi^2} + \left(\frac{eE_w L_w}{\pi} \right)^2 \right] \frac{L_w^4 m^{*2}}{\hbar^4 \pi^4} \right\}^{1/2} - 1 \right) \quad (6)$$

where ΔE is the band offset. By using appropriate values for these structures, the PL peak energy dependence on the well widths can be obtained.

This computation shows that the energy shift between different exciton peaks corresponding to well widths of full c -vectors (0.52 nm) vary strongly with width, between 10 and 25 meV in the range of well widths (1.5–4.5 nm) shown in figure 3. This strong spectral sensitivity to well width fluctuations is due to the internal electric field in addition to the quantum well width. According to this figure there is a reasonable agreement between calculated and experimental results for recombination energy between the hole and electron ground state in the triangular quantum well approximation. In addition, it is obvious that with increasing well width the PL peak energy of QWs has been red-shifted.

For the case of 3.0 nm well width in the MQW the PL peak is significantly red-shifted with respect to a 1.5 nm QW (figure 2); this is evidence of the influence of the internal electric field in the QW besides that of the quantum well width. The full width at half maximum (FWHM) of this peak is a bit more than 25 meV, indicating that it contains an unresolved overlap of contributions from different well widths. For the 4.5 nm MQW the spectral red-shift

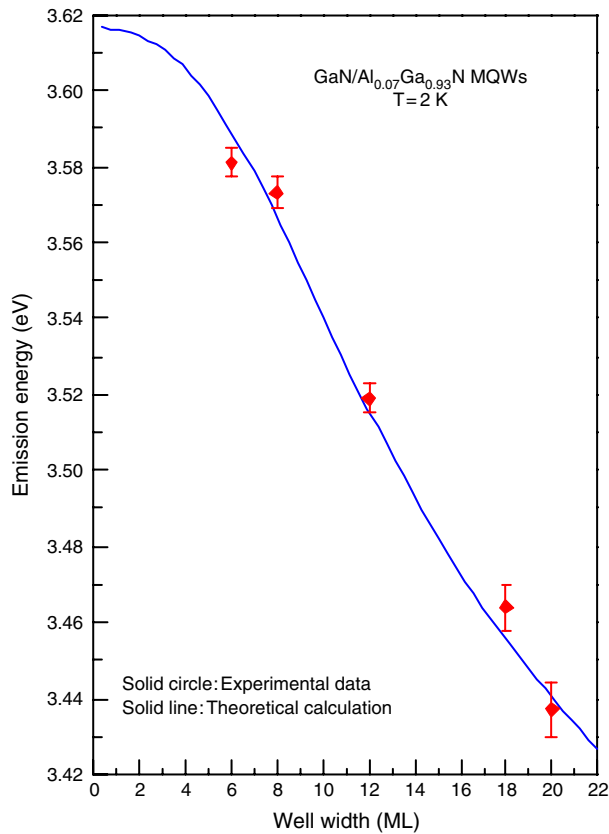


Figure 3. Well width dependence of excitonic peak energies for triangular GaN/Al_{0.07}Ga_{0.93}N MQWs.

is more evident (figure 2); the peak position at about 3.45 eV is here well below the GaN band gap. For the 4.5 nm wide QW, the FWHM of the low-energy PL peak is about 25 meV, while for the high-energy one it is less than 20 meV. Narrower peaks are observed for the 1.5 nm wide QW sample, where the FWHM of the low-energy and high-energy PL peaks are about 20 and 15 meV, respectively. This is consistent with the theoretical prediction of decreasing PL bandwidth broadening for very thin QW layers, as a result of an increasing extension of the exciton wavefunction into the barrier layer [16].

Time-resolved spectroscopy was employed to study the dynamics of optical transitions in GaN/Al_xGa_{1-x}N MQWs. Figure 4 shows the decay times of the main emission lines of the second set of MQW samples (i.e. QWs with 1.5, 3.0, 4.5 nm wells widths) measured at $T = 2$ K. As can be seen from figure 4, the recombination time is a growing function of the well widths, and with increasing well width from 1.5 to 3 nm the decay time increases by around one order of magnitude. To explain such behavior we present two reasons. First, the radiative lifetime increases almost exponentially with L_w , because of the separation of electrons and holes by the electric field [17]. Second, a nonradiative carrier transfer can occur from the narrower wells towards the wider ones, as recently shown by time-resolved PL measurements on similar samples [16]. This transfer is responsible for the collapse of PL intensity for the narrow wells, when the barriers are narrow. This effect is particularly dramatic for the 1.5 nm wide QW (see figure 7).

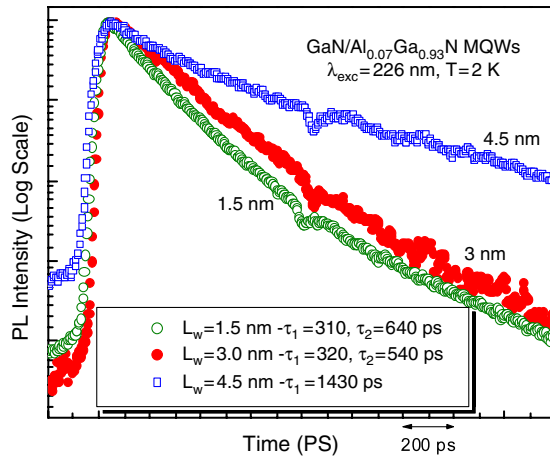


Figure 4. PL decay time at the peak position of the PL spectra for a set of GaN/Al_{0.07}Ga_{0.93}N MQW samples with different well width.

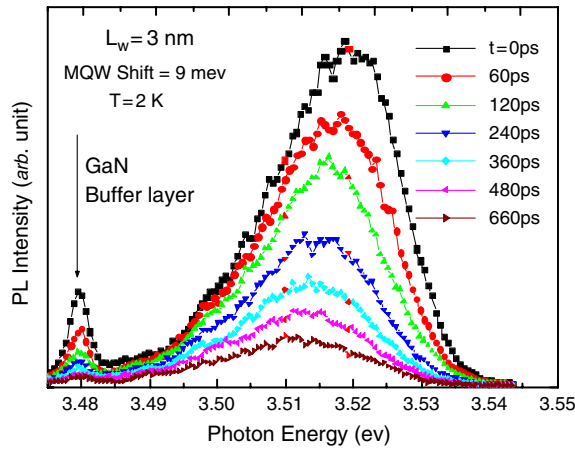


Figure 5. Time-resolved PL spectra of the main emission lines measured at $T = 2$ K for 3 nm well MQWs for several representative decay times.

The efficiency in the screening of the polarization fields by the extrinsic carriers can in principle be verified by time-resolved PL measurements. Figure 5 shows the TRPL spectra of the main emission lines of the 3 nm well MQW samples measured at $T = 2$ K, for several representative decay times. The arrows in figure 5 indicate the spectral peak positions at different decay times. The solid line indicates the position of the excitonic transition peak in the GaN buffer layer grown under similar conditions. Several features can be observed for the 3 nm well MQW sample, as shown in figure 5. First, in similarity to the cw PL spectra shown in figure 2, the spectral peak position at decay time $t_d = 0$ (3.519 eV) is blue-shifted with respect to the emission line in the GaN buffer layer (3.478 eV). Second, the line width of the emission line decreases with decay time. Third, the peak position of the emission line markedly shifts toward lower energies with an increase of decay time. For comparison with different well thicknesses, we have plotted in figure 6 the decay time dependence of the peak position E_p of different MQWs. The TRPL results can be explained well by the collective

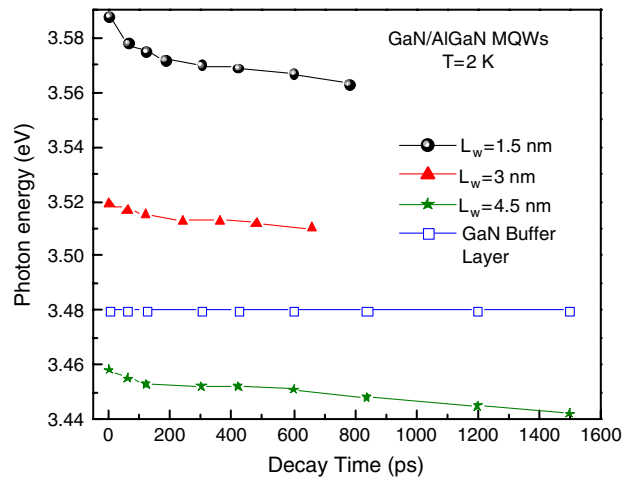


Figure 6. The peak position E_p of the main emission line as a function of decay time t_d measured at $T = 2$ K for the 1.5, 3, and 4.5 nm well GaN/Al_{0.07}Ga_{0.93}N MQW samples.

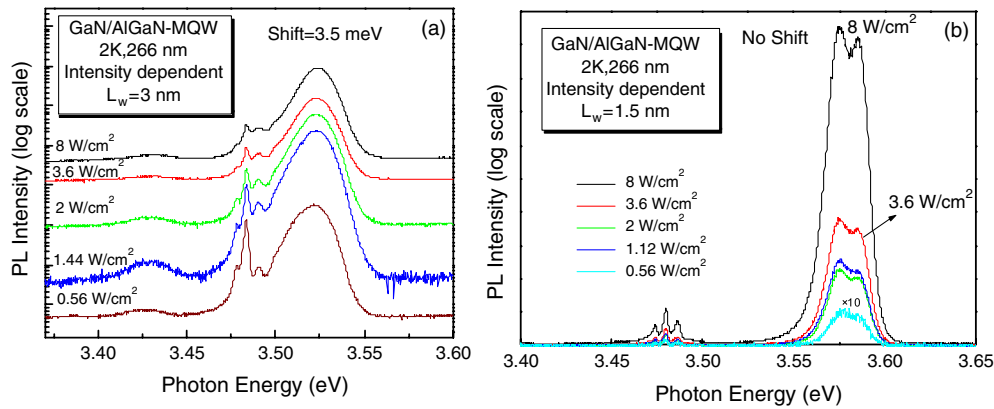


Figure 7. Excitation intensity dependence of the PL spectrum for the undoped GaN/Al_{0.07}Ga_{0.93}N MQW sample at 2 K (a) 3 nm and (b) 1.5 nm.

effects of piezoelectric field and photo-excited carrier screening. As mentioned above, it is well established that polarization fields (piezoelectric and spontaneous fields) are present in the well regions of the GaN/AlGaIn MQWs and heterojunctions due to the lattice mismatch between GaN and AlGaIn as well as the large value of the piezoelectric constant in GaN. Under the influence of the polarization field, optically excited carriers drift apart. The electrons (holes) move toward the direction opposed to (along) the polarization field and the field induced by these spatially separated charge carriers will screen the polarization field. On the other hand, the screening field due to the spatially separated charge carriers decreases with decay time because of the radiative recombination of electrons and holes. At $t_d = 0$, the screening field induced by the photo-excited electrons and holes is strongest, which reduces or partially balances out the polarization field. As the decay time increases, the carriers recombine radiatively and the screening field gradually diminishes and the original piezoelectric field is restored. Thus the total amount of shift from $t_d = 0$ to $t \rightarrow \infty$ effectively corresponds to the variation of the electron and hole energy levels in the presence of the polarization field with and without carrier

screening, respectively. From this the piezoelectric and spontaneous fields strength can also be estimated. By comparing experimental and calculation results, a lower limit value of the polarization field strength of about 48 MV m^{-1} in GaN/Al_{0.07}Ga_{0.93}N MQWs (as well as the electron and hole wavefunctions) has been obtained for a 3 nm well MQW. This value is in agreement with the results of other authors, who also studied GaN/AlGaIn QWs with similar barrier compositions. For example, Grandjean *et al* [18] have shown that an Al content in the barrier in the range of 10–15% cause a field in the range of 40–60 MV m^{-1} in the active layer [5, 19–23].

Finally, we have checked the possible screening of the internal electric field due to injected carriers by decreasing the laser intensity by around two decades in figure 7. This did not change the PL energy of narrow wells (1.5 nm wide), but could induce a weak (<5 meV) red-shift for the wider ones. The different behavior between thin and wide wells is simply due to much larger lifetimes in wider QWs, yielding a larger accumulation of carriers than in narrower wells; in the continuous regime (cw-PL) [19], this cause more effective screening of electric fields in wider QWs; i.e. figure 7 confirms the result that was obtained before in figure 4.

4. Conclusions

In conclusion, we have shown that electrostatic effects which take place in group-III nitrides in their wurtzite crystallographic phase have important consequences on the optical properties of GaN/AlGaIn multiple quantum wells and also, contrary to what happens in MQW structures fabricated from other semiconductor systems, such as (Al, Ga)As/GaAs, the PL energy of GaN/AlGaIn MQWs blue-shifts on decreasing the barrier width. This is assigned to the redistribution among wells and barriers of the strong electrostatic fields induced by the polarization differences between the well and barrier materials. This effect is of crucial importance in nitrides because of their huge piezoelectric and spontaneous polarization constants. From cw PL spectra at low temperature (2 K), we observe that the exciton transitions for 1.5 and 3 nm MQWs are blue-shifted with respect to the GaN buffer layer and that the PL emission peak position for 4.5 nm well MQWs is red-shifted with respect to the GaN buffer layer. On the other hand, the time-resolved PL spectra of the 1.5, 3 and 4.5 nm well MQWs reveal that the excitonic transition is in fact blue-shifted at early decay times due to quantum confinement of carriers, and the spectral peak position shifts toward lower energies as the decay time increases, and becomes red-shifted at longer decay times.

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