Optical evidence for lack of polarization in $(11\bar{2}0)$ oriented GaN/(AlGa)N quantum structures

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We apply continuous and time resolved photoluminescence spectroscopy for studying GaN/AlGaN multiquantum wells structures grown on nonpolar *a*-plane GaN templates. We found that (a) the energy of the emission from the nonpolar samples decreases slightly with the quantum well width, in a manner explained by the quantum size effect only; (b) the energy differences between the absorption and the emission peaks are independent of the well width; and (c) the decay time of the photoluminescence is only slightly dependent on the quantum well width and is quite similar to that of bulk GaN. These observations are markedly different from measurements obtained from conventional polar [0001] oriented quantum well samples. They clearly demonstrate the absence of an electric field in the nonpolar samples. Our observations are favorably compared with an eight bands $k \cdot P$ model calculations. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1926406]

Nonpolar growth of III-nitrides (III-N) heterostructures is a promising means of circumventing the strong polarization-and piezoinduced electric fields that exist in conventionally grown wurtzite nitride semiconductors. The total polarization of a III-N film consists of spontaneous and piezoelectric polarization contributions which both originate from the single polar [0001] axis of the wurtzite nitride unit cell. The common growth direction of nitride layers is parallel to this polar axis, hence, polarization discontinuities exist along the growth direction that create fixed sheet charges at surfaces and interfaces. The resulting internal electrical field spatially separates electron and hole wave functions along the quantum wells (QWs) growth direction, thus reducing the energy associated with their radiative recombination and lowering its internal quantum efficiency. These effects are manifestations of the quantum confined Stark effect (QCSE) and have been thoroughly analyzed for GaN/(AlGa)N and GaN/InGaN quantum wells.[1–7] Conversely, polarization-induced electric fields should not affect III-N semiconductors grown in nonpolar directions (i.e., perpendicular to the [0001] axis) due to the absence of polarization discontinuities along these growth directions. For example, *m*-plane AlGaN/GaN quantum wells were recently grown on the (100) plane of $LiAlO_2$ substrates using both plasma assisted molecular beam epitaxy (MBE) and halide vapor phase epitaxy.[8,9] More recently, growth of *a*-plane QWs on *r*-plane sapphire substrates resulting in $(11\bar{2}0)$ oriented QWs was attempted by MBE (Ref. 10) as well as by metal-organic chemical vapor deposition (MOCVD).[11] These QWs indeed gave experimental indications for the absence of the internal, built-in electric fields.

In this article, we describe in detail, continuous wave (cw) and pulsed optical studies of nonpolar $(11\bar{2}0)$ *a*-plane GaN/(Al,Ga)N multiple quantum well (MQW) structures grown by MOCVD. The samples contained ten periods of $GaN/(Al_{0.2}Ga_{0.8})N$ QWs grown on a nonpolar $(11\bar{2}0)$ *a*-plane GaN template in a vertical close-spaced showerhead MOCVD reactor. The growth temperature was ~1000 °C and TMGa, TMA1, and NH_3 were used as precursors for the growth. The *a*-plane GaN templates were grown on single side polished *r*-plane sapphire [11]. Four different samples were grown with varying GaN well width (nominally from 4 to 9 nm) while holding the AlGaN barrier thickness (10 nm) and its Al composition constant (0.2). After growth, the structures were characterized by high-resolution x-ray diffraction (HRXRD) in conjunction with dynamical diffraction simulations.[12] Cross-section transmission electron microscopy and atomic force microscopy in tapping mode were used, in addition, in order to image the MQW microstructure and surface morphology, respectively.[12] The structural data obtained from the high-resolution x-ray diffraction measurements are summarized in Table I. The validation of coherence growth of similar MQW structures was confirmed by a reciprocal space mapping taken in the vicinity of asymmetric GaN $(20\bar{2}0)$ reflection.[12] For the optical measurements, the samples were mounted in a closed cycle helium flow optical cryostat. We used light from a xenon lamp, dispersed by 0.34 m monochromator, as an excitation source for the photoluminescence excitation (PLE) measurements. For time resolved spectroscopy, the samples were excited at normal incidence by a frequency-tripled, 260 nm, 120 fs-pulsed radiation from a Ti: sapphire laser. The collected PL light was dispersed by a 0.22-m monochromator, followed by ei-

TABLE I. HRXRD measured layer thickness and composition.

Sample ID	GaN QW thickness (nm)	AlGaN barrier thickness (nm)	Al composition
N021120B	4.1	10.3	0.2
N021120C	5.8	9.1	0.2
N021120D	7.9	9.0	0.2
N021120E	9.3	8.9	0.21

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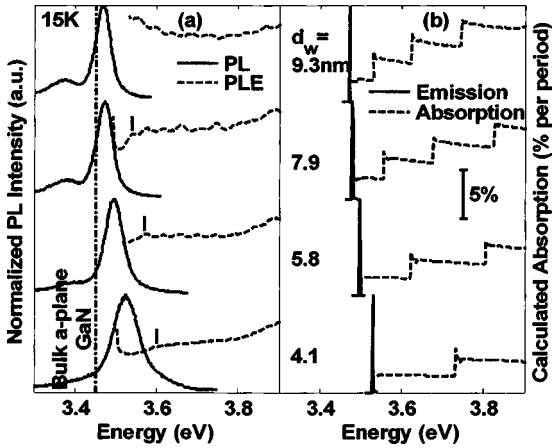

FIG. 1. Measured (a) and calculated (b) PL (solid line) and PLE (dashed line) spectra of four *a*-plane nonpolar AlGaN/GaN MQWs samples. The vertical bars in (a) indicate exciton peaks in PLE spectra.

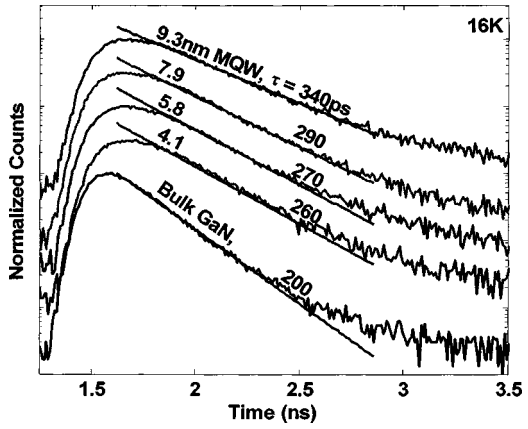

FIG. 2. Low temperature PL decay time measurements for the four *a*-plane nonpolar AlGaN/GaN MQWs samples.

ther a UV-enhanced liquid nitrogen-cooled charge coupled device array (cw mode), or by a multichannel plate photomultiplayer (pulsed mode). Conventional time-correlated, single-photon counting electronics were used for the time-resolved spectroscopy. The low temperature (15 K), PL spectra of four MQW samples are presented in Fig. 1(a). For the excitation, a 4.76 eV (260 nm) radiation was used with estimated average intensity of 400 w/cm^2. As can be seen in Fig. 1, the PL emission energy becomes lower as the QW widens. The energy of the PL from the widest QW samples (7.9 and 9.3 nm) is roughly the same (3.46–3.47 eV) and it is about 20–30 meV above the energy of the PL from the nonpolar relaxed GaN (not shown). This relatively small dependence of the PL energy on the QW width can be fully accounted for by the quantum size effect (see below). This observation is completely different from that observed in polar GaN/AlGaN or InGaN/GaN MQW (Refs. 5 and 6) samples grown on (0001) oriented sapphire substrates. In those samples, the PL energy dependence on the QW width is much stronger, since in addition to the quantum size effect, strong spontaneous and strain-induced piezoelectric fields significantly reduce the PL energy. In those samples, while the energy of the PL from narrow QWs is higher than the GaN band-gap energy, the energy of the PL from wide QWs fall well below (100–200 meV) the GaN band-gap energy. This is due to the quantum confined Stark effect,[5,6] in the presence of the large electric fields in those polar samples.

The small (20 meV) difference between the PL energy from the widest QW sample (9.3 nm) and that from the relaxed nonpolar GaN, is attributed, mainly, to residual compressive strain in the GaN QW layer. The compressive strain, which is absent in the relaxed GaN layer in the control sample, increases the PL from the MQW sample. We found that the PL energy from relaxed polar GaN is 3.48 eV, 30 meV higher than the PL from nonpolar GaN. This difference is probably due to differences in the thermal mismatch strain between the two different orientations of the sapphire substrates.

The PLE spectra of the MQW samples are shown in Fig. 1(a). For comparison, we present in Fig. 1(b) calculated emission and absorption spectra. The calculations are based on our $k \cdot P$ model, and they take into account the growth direction, the resulting lattice mismatch strain, and the two-dimensional confinement.[6] In order to facilitate quantitative comparison between the measurements and the calculations, the presented PLE spectra were normalized to unity at their maximum. We note in Fig. 1(a) that the energy difference between the absorption edge, as determined by the PLE spectra and the PL energy (Stokes shift) is nearly independent of the QW width. The shifts are comparable to the PL linewidth and completely absent in the calculated spectra. We attribute the shift to inhomogeneous broadening which is expected due to the samples' structure and composition nonuniformity. This is in marked contrast to polar samples, where it was shown (for [0001] oriented GaN/InGaN MQWs) that the Stokes shifts strongly increase with the QW width.[6] The Stokes shift dependence on the QW width was quantitatively attributed to the presence of strong electric fields in the polar samples.[6] Therefore, the absence of this dependence is yet another strong indication of the absence of electric fields in the nonpolar samples. The higher energy parts of the PLE spectra in Fig. 1(a) do not show the rich structures that the absorption calculations in Fig. 1(b) show. This discrepancy is also attributed to nonuniformity in the samples, which smears the higher energy spectral features. Indeed, lateral undulations in the quantum wells' thickness of order 10% can be deduced from the x-ray measurements reported in Ref. 12.

Another indication to this effect is the absence of a clear excitonic resonance in the PLE spectra of the nonpolar MQW samples. These resonances are commonly observed in narrow MQW and bulk GaN samples.[6] Here again, the uniformity of the nonpolar samples and their interface quality are lower than the uniformity and quality achieved in conventional, polar samples. As a result, the excitonic resonance is smeared in the non-polar samples.

PL decay time measurements for the nonpolar reference and MQW samples are shown in Fig. 2. All the PL transients show a nonexponential decay, which becomes slower at longer times. We fitted the initial, fastest part of each transient with a monoexponential decay model, shown by the dashed lines in Fig. 2. We note that for all the MQW samples, the fitted characteristic decay times are in the range of 200–400 ps and they are nearly the same as that of the bulk GaN control sample. This is vastly different from the PL decay times from polar GaN/AlGaN (Ref. 5) and InGaN/GaN (Ref. 6) MQW samples. In polar samples the PL decay times strongly depend on the MQW widths and can achieve a duration of 10–100 ns for similar well widths. This

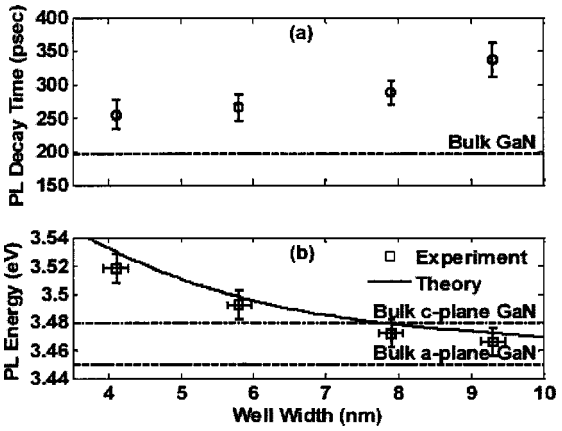

FIG. 3. Measured decay times (a) and PL energies (b) vs QW widths. The solid line represents the calculated band gap using eight bands $k \cdot P$ model.

observation has also been shown to be a manifestation of the presence of the strong electrostatic field in the polar MQWs samples. The electrostatic field spatially separates the electron and hole wave functions, thereby reducing the oscillator strength for their radiative recombination. At low temperatures, the measured decay time corresponds mainly to the radiative lifetime, which is inversely proportional to the oscillator strength.

The dependence of the measured and calculated PL energies and decay times on the non-polar MQW width is presented in Figs. 3(a) and 3(b), respectively. It is clear from Fig. 3 that the size quantization effect can approximately account for both the PL energy and decay time dependencies on the well width. Since the calculated PL energies do not include excitonic binding energies, there are systematic differences between the measured and calculated PL energies in Fig. 3(b).

The characteristically strong dependence of the PL energy and decay time, observed in polar MQW samples is absent in the nonpolar samples. The weak dependence of the decay times on the QW width is similar to that obtained from GaAs/AlGaAs MQW samples[13] with comparable width to the GaAs bulk exciton radius ($\sim$10 nm).[14] The results in Fig. 3(a) nicely demonstrate the same effect for nonpolar GaN/AlGaN QWs, which are comparable in size to the GaN bulk exciton radius ($\sim$3.6 nm).

In conclusion, we have studied nonpolar GaAlN/GaN MQW samples and have shown that

1. The PL energies slightly decrease with the increase in the QWs width.
2. The PL decay times increased to some extent with the increase in the QWs width.
3. The Stokes shifts between the exciton peaks in PLE and PL spectra are comparable to the PL line width and nearly QW width independent.

These dependencies are in quantitative agreement with considerations based on the quantum size effect in symmetric QWs. Therefore, we view these results as unambiguous manifestations of the absence of electrostatic field in the nonpolar samples.

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