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Broadening of intersubband and interband transitions in InGaN/AlInN multi quantum wells

M. Gladysiewicz^{1, a)}, R. Kudrawiec¹, J. Misiewicz¹, G. Cywiński², M. Siekacz²,
and C. Skierbiszewski²

¹*Institute of Physics, Wrocław University of Technology, Wybrzeże Wyspiańskiego 27,
50-370 Wrocław, Poland*

²*Institute of High Pressure Physics, Polish Academy of Science,
Sokolowska 29/37, 01-142 Warsaw, Poland*

The authors have applied modulation spectroscopy to study intersubband (IS) and interband (IB) transitions in InGaN/AlInN multi quantum well (QW) structures with the QW width varying from 1.3 to 1.8 nm, and proposed a simple method to analyze the broadening of IS and IB transitions. Within this method the measured broadenings of IS and IB transitions have been compared with theoretical calculations performed within the effective mass approximation for a QW system with various QW width fluctuations. In the framework of this comparison it has been found that the QW width fluctuation in the investigated InGaN/AlInN QW system is close to 2 monolayers.

^{a)} Corresponding author: marta.gladysiewicz@pwr.wroc.pl

In order to achieve intersubband (IS) absorption at fiber-optics telecommunication wavelengths (i.e., 1.3 and 1.55 μm) in GaN-based quantum well (QW) structures, the QW width has to be smaller than 10 monolayers (MLs) [1-8]. In this regime of QW width, its small fluctuation (even 1 ML) significantly influences the energies of optical transitions. Recently, it has been shown that individual absorption peaks corresponding to an integer number of MLs can be observed for GaN/AlN quantum wells [7, 8]. The homogeneous broadening of IS absorption for this system (i.e., GaN/AlN QW with no QW width fluctuation) has been found to be ~ 67 meV [7]. In the case of indium containing QWs (i.e., InGaN/AlInN QWs), the broadening of optical transitions is expected to be higher because of the alloying effect which leads to higher inhomogeneities in this system, including indium segregation effects and, very likely, higher fluctuations of the QW width. However, the incorporation of indium atoms into GaN/AlN QWs is needed or/and profitable in many applications and therefore InGaN/AlInN QWs are the subject of many studies and applications in optoelectronic devices.

In this work we have applied modulation spectroscopy [9] to study energies and broadenings of IS and interband (IB) transitions in InGaN/AlInN multi QWs (MQWs) of various QW widths. So far the IS absorption in GaN/AlN QW structures was investigated by the transmission and/or photo induced absorption spectroscopy. In this paper we have applied photomodulated transmission (PT) [10] to investigate the energy and broadening of IS absorption instead of the standard transmission measurement. The broadening of IB transitions has been investigated by contactless electroreflectance (CER) [11, 12]. In order to evaluate the broadening of IS and IB transitions and determine the QW width fluctuation a simple theoretical model has been proposed.

For this study we grew InGaN(d)/AlInN(3nm) multi QWs (MQWs) samples of various QW widths (d=1.3, 1.6, and 1.8nm) by plasma-assisted molecular-beam epitaxy on

Ga polar GaN/Al₂O₃ templates. 40 period InGaN/AlInN MQWs were grown after a 20 nm In_{0.1}Ga_{0.9}N buffer layer and capped with an undoped 100 nm In_{0.1}Ga_{0.9}N layer. The InGaN wells were uniformly doped with Si at a level of 10^{20} cm⁻³. The structural quality of these samples were studied by high resolution transmission electron microscopy (TEM) and X-ray diffraction (XRD) mapping techniques. Both XRD and TEM results confirm good structural quality [13]. The indium concentration in InGaN QWs and AlInN barriers has been determined to be ~5 and ~0.5%, respectively.

A conventional experimental set-up with a tungsten halogen lamp (250 W) as a probe light source, a 0.55 m monochromator and an appropriate detector (PbS photodiode and photomultiplier for infrared and ultraviolet spectral range, respectively) was applied for obtaining photomodulated transmittance and contactless electroreflectance spectra. For optical measurements, samples were prepared in a way to form a multipass waveguide as shown in Ref. 6. For PT measurements the probe beam was sent through this waveguide and the photomodulation was provided by a laser beam (266 nm line from YAG laser) at the normal incidence on the sample surface. For CER experiments, samples were mounted in a capacitor with one semitransparent electrode. This electrode was kept at a distance of ~0.5 mm from the sample surface while the sample itself was fixed on the bottom copper electrode. A maximum peak-to-peak alternating voltage of ~3.0 kV with the frequency of 285 Hz was applied. CER spectra were measured at near normal incidence of the probing beam on the sample surface. Other relevant details of CER measurements were discussed in Ref. 12.

Figure 1(a), 1(b), and 1(c) show room temperature PT spectra in the infrared spectral range for InGaN/AlInN MQW structures with the nominal QW width of 1.3, 1.6 and 1.8 nm, respectively. A resonance-like transition, which is related to the intersubband absorption between the first and the second electron subband (1e-2e) in the InGaN/AlInN MQW, is

clearly observed in PT spectra of the three samples. This finding clearly shows that PT technique can be applied to study IS transitions in GaN-based QW structures.

The broadening of 1e-2e transition is a few times larger than the homogeneous broadening of IS absorption reported for indium free systems (~350 meV for InGaN/AlInN QWs versus 67 meV for GaN/AlN MQWs). In addition, it is visible that this broadening increases when the QW width decreases. In order to extract energies and broadenings for the intersubband absorption in the three samples, the standard fitting procedure assuming Lorentzian lineshape [14] has been applied. According to this model PT spectrum can be fitted using the following formula

$$\frac{\Delta T}{T}(E) = C \cdot e^{i\vartheta} (E - E_0 + i \cdot \Gamma)^{-m}, \quad (1)$$

where E_0 and Γ are the energy and the broadening parameter of the transition, and C and ϑ are the amplitude and phase of the line, respectively. The term m refers to the type of critical point for optical transitions. In our case, it has been assumed that $m = 3$ since according to our experience such a line is very similar to the third derivative Gaussian lineshape, i.e. the most appropriate lineshape for inhomogeneous system. The fitting curves are shown by thick grey lines together with the moduli of individual resonances (dashed lines) obtained according to Eq. 2

$$\Delta\rho(E) = \frac{|C|}{\left[(E - E_0)^2 + \Gamma^2 \right]^{\frac{m}{2}}} \quad (2)$$

with parameters taken from the fit. In order to analyze broadenings of intersubband absorption and to compare them with literature data for GaN/AlN systems, we have determined the full width at half maximum (FWHM) for the moduli peak.

Figure 1 (d) shows energies of 1e-2e transition obtained from PT measurements for the three MQW samples together with theoretical calculations, which were performed in the framework of the electron effective mass approximation with material parameters taken from Ref. 14. The built-in electric field inside the InGaN/AlInN MQW has been calculated from piezoelectric and spontaneous polarizations in this system [15] with appropriate periodic boundary conditions [16] assuming 20% screening of the electric field due to the Si doping.

In order to calculate the broadening of intersubband absorption, we made an assumption that it entirely originates from the QW width fluctuations. This is a “border case” neglecting the influence of In-segregation effects, which is expected to be small in these samples because of the low indium content. This broadening is defined as $\gamma^{IS} = \gamma_0^{IS} + \gamma_1^{IS}$, where γ_0^{IS} corresponds to a system without QW width fluctuations (we assume that $\gamma_0^{IS} = 67\text{meV}$ after Ref. 7) and γ_1^{IS} is a contribution which is associated with QW width fluctuations. This contribution can be calculated according to Eq. (3)

$$\gamma_1^{IS} = \left| \frac{\partial E^{1e2e}}{\partial w} \right| \Delta w, \quad (3)$$

where w is the QW width and Δw is its fluctuation. In order to determine the magnitude of QW width fluctuations, this parameter is treated as a free parameter.

Figure 1(e) shows theoretical calculations of the broadening parameter of 1e-2e transition for $\Delta w = 1, 2,$ and 3 ML together with experimental data, i.e., the FWHM obtained from PT measurements. A good agreement between experimental data and theoretical

calculation is observed for $\Delta w \approx 2$. This result is consistent with TEM measurements for these samples [13].

The broadening of the fundamental IB transition in these MQW has been obtained from CER measurements accompanied with the standard fitting procedure [12], see Fig. 2 (a), (b) and (c). The fitting curves are shown by thick grey lines together with the moduli of individual resonances (dashed lines). The strongest CER resonance at 2.9 eV is attributed to the absorption in the InGaN cap layer [12]. Remaining CER resonances are associated with the absorption in the MQW region. The notation $lHkC$ denotes the transition between l -th hole subband and k -th conduction subband. It is worth noting that the 1H1C transition is observed in these MQW structures in CER spectroscopy (absorptionlike technique) despite the silicon doping in the QW region. It means that electron concentration in some InGaN QWs can be smaller. Such a situation is expected close to the sample surface in the surface depletion region. On the other hand it is worth noting that the IB absorption at higher k states is possible for QWs with the occupied $k = 0$, but this absorption is much weaker than absorption at $k = 0$, especially in modulation spectroscopy.

Figure 2(d) shows the results of theoretical calculations of energy of 1H1C transition together with experimental data. The broadening of 1H1C transition, γ^{IB} , is defined like for IS transitions: $\gamma^{IB} = \gamma_0^{IB} + \gamma_1^{IB}$, where γ_0^{IB} corresponds to bulk alloy broadening (this broadening for InGaN cap layer equals 250-280 meV) and γ_1^{IB} is associated with the QW width fluctuations. The γ_1 component is calculated according to Eq. (4)

$$\gamma_1^{IB} = \left| \frac{\partial E^{1H1C}}{\partial w} \right| \Delta w, \quad (4)$$

where w is the QW width and Δw is its fluctuation.

Figure 2(e) shows theoretical calculations of the broadening of 1H1C transition for $\Delta w = 1, 2,$ and 3 ML together with experimental data, i.e., the FWHM for 1H1C transition obtained from CER measurements. Like for IS absorption a satisfactory agreement between experimental data and theoretical calculations is observed when the QW width fluctuation is close to 2 ML. It is worth underlining that the broadening of interband transitions is about ~ 1.5 -2 times larger than the broadening of intersubband absorption. This observation is very consistent with theoretical analysis, see Fig. 1(e) and Fig. 2(e), and agrees with expectations for the two types of optical transitions.

We expect that the magnitude of QW width fluctuation depends on the growth conditions (i.e., mostly on the growth temperature). In our case, the three MQW samples were obtained at exactly the same growth conditions and they differ between themselves by the nominal QW width only. In Fig. 3 we have compared our calculations with experimental data for InGaN/AlInN MQW samples obtained at very similar growth conditions [6, 13]. Like in the previous case [Fig. 1(e)] we have found that a good agreement between experimental data and theoretical calculations is observed when the QW width fluctuation is close to 2 ML. In addition, such a comparison has been performed for binary GaN/AlN MQWs from Ref. [7] where the authors have identified that the homogenous broadening of IS absorption for GaN/AlN system equals 67meV. Our analysis suggests that for some GaN/AlN MQWs the larger broadening of IS absorption appears because of a QW width fluctuation. This fluctuation has been estimated to be below 1 ML. It means that 1ML thick terraces can exist in these MQWs but the size of these terraces is rather small. For large terraces ($>1 \mu\text{m}$) individual peaks, which are related to the integer number of ML, are expected in absorption spectra. Such peaks were observed for some GaN/AlN MQWs in Ref. 7. In the case of In-containing MQWs, such peaks were not resolved in absorption spectra since the QW width

fluctuations are more smeared in this system due to the alloying effect. In addition, the incorporation of indium atoms into GaN/AlN QW enhances the QW width fluctuation (compare solid and open points in Fig.3). Indium also leads to the energy gap fluctuations but this effect is small in our InGaN/AlInN MQWs because of low indium concentration.

In conclusion, it has been clearly shown that energies and broadenings of IS and IB transitions in InGaN/AlInN MQW structures can be studied by modulation spectroscopy. In addition, a simple theoretical model to determine the QW width fluctuation from the broadening of IS and IB transitions has been presented in this work. Within this model the fluctuation of the QW width has been found to be 2 monolayers. It is worth noting that this model can be applied to the analysis of broadening parameters obtained from PT and CER (i.e. absorptionlike measurements). In the case of photoluminescence spectroscopy, the fluctuation of QW thickness cannot be determined within this theoretical model because of emissionlike character of photoluminescence, i.e., its high sensitivity to localized states. It means that the modulation spectroscopy is a very useful method to evaluate the quantum well width homogeneity in narrow QW systems. This finding is very important since optical measurements are much cheaper and easier than TEM measurements which need the time consuming sample preparation and are destructive for samples.

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Figure Captions:

FIG. 1. Room temperature photomodulated transmission spectra in the infrared spectral range for InGaN/AlInN MQW structures with the QW width of 1.3 nm (a), 1.6 nm (b), and 1.8 nm (c). Theoretical calculations of the energy of 1e-2e transition (a) and its broadening (b) for InGaN/AlInN MQW structure together with experimental data (solid squares).

FIG. 2. Room temperature contactless electroreflectance spectra in the ultraviolet spectral range for InGaN/AlInN MQW structures with the QW width of 1.3 nm (a), 1.6 nm (b), and 1.8 nm (c). Theoretical calculations of the energy of 1H1C transition (a) and its broadening (b) for InGaN/AlInN MQW structure together with experimental data (solid squares).

FIG. 3. Theoretical calculations of the broadening of 1e-2e transition in InGaN/AlInN MQWs (black lines) and GaN/AlN MQWs (gray lines) together with experimental data from this work and the literature (i.e., photoinduced absorption [6] and transmission [7, 13, 18]).

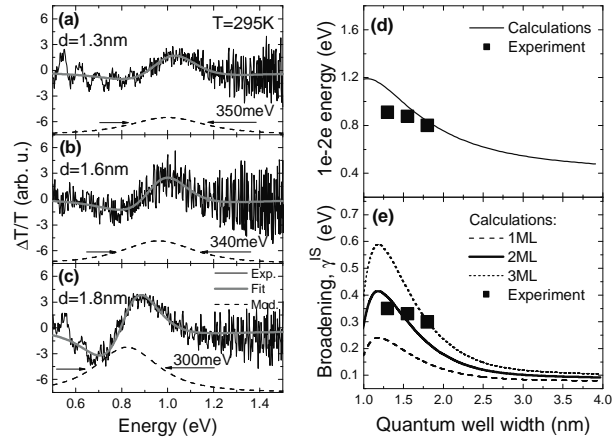


FIG. 1.

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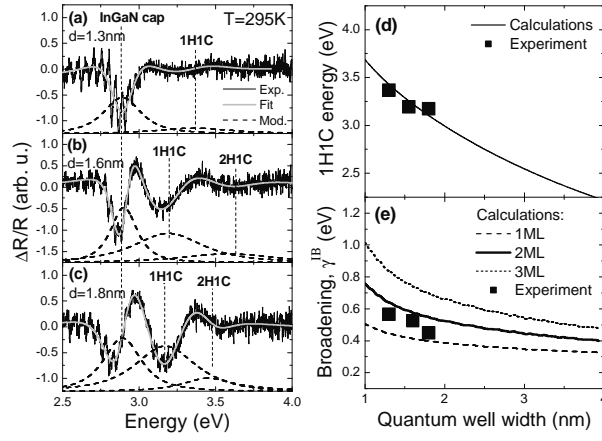


FIG. 2.

(R. Kudrawiec et al.)

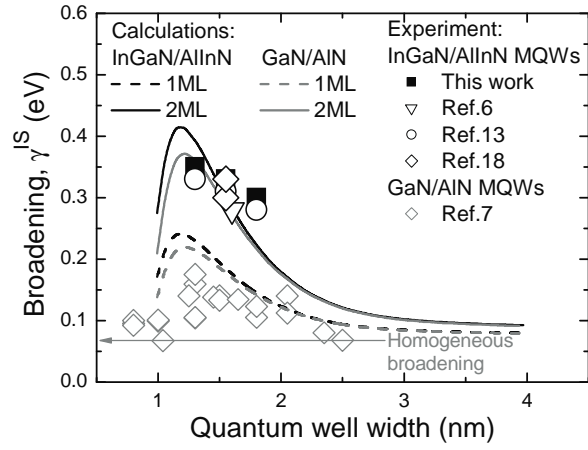


FIG. 3.

(R. Kudrawiec et al.)