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Electromodulation spectroscopy of the ground and excited state transitions in GaInN/AlInN multi-quantum wells

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ABSTRACT

Contactless electroreflectance (CER) has been applied to investigate energies and intensities of optical transitions in GaInN/AlInN multi-quantum wells (MQWs) with the various QW widths (1.55 and 1.8 nm) and barrier thicknesses (3 and 6 nm). In addition to the ground state transition, optical transitions related to excited states (2H–1C and 1H–2C transitions, where *k*H–*l*C denotes a transition between the *k*th valence and the *l*th conduction subbands) have been observed in CER spectra. It has been found that the intensities of 2H–1C and 1H–2C transitions (i.e., transitions which are forbidden for non-polar square-like QWs) are comparable with the intensity of the ground state transition. In addition, it has been observed that the relative intensities of QW transitions depend on both the QW width as well as the barrier thickness. This experimental observation has been confirmed by theoretical calculations performed in the framework of the electron effective mass approximation model taking into account polarization effects in wurtzite III-N alloys. The calculations clearly show that the intensity of QW transitions for GaInN/AlInN MQWs strongly varies with the QW width as well as the barrier thickness.

GaInN/AlInN quantum wells currently attract extensive attention due to their potential applications in blue/UV lasers [1,2] and optical modulators operating in telecommunication windows, i.e. at 1.3 and 1.55 μm [3–8]. Usually, these QWs are grown along the (0001) GaN direction (i.e., on *C*-plane-oriented GaN templates grown on sapphire substrates), which is the polar direction for wurtzite III-N nitrides [9,10]. Due to the strong spontaneous and piezoelectric polarization in this system, the envelope wave-function overlap varies significantly with the QW width and therefore the optical selection rules, which are typical of non-polar square-like QWs, cannot be applied to GaInN/AlInN QWs [11]. Recently, it has been observed in contactless electroreflectance, which is absorption-like technique, that the intensity of nominally forbidden transitions for non-polar square-like QWs (i.e., the 2H–1C and 1H–2C transitions, where *k*H–*l*C denotes a transition between the *k*th valence and the *l*th conduction subbands) is comparable with the intensity of the ground state transition (1H–1C transition) [11]. It proves that polarization effects significantly influence the envelope wave-function overlap in this QW system. In order to find the electron–hole overlap integral properly for a GaInN/AlInN QW, the Schrödinger equation has to be solved for the QW potential, which corresponds to the real situation in the QW structure. In the case of GaN-based QWs, the accuracy of the QW potential determination can be limited because of some scattering of material parameters in the literature [12], especially for In-containing III-N alloys (GaInN and AlInN in this case). In addition, the accuracy of the determination of QW width and In content is extremely important for thin GaInN/AlInN QWs ($L_w < 3$ nm, i.e., QWs which are interesting from the viewpoint of applications). It means that a direct comparison of theoretical calculations with experimental data can be a risk issue in some cases. However, the analysis of trends in theoretical calculations and their comparison with experimental data is very useful and fully justified. In this paper we have focused on the theoretical analysis of the electron–hole overlap integral for GaInN/AlInN QWs of various QW widths. The calculations have been compared with the experimental investigations of the ground and excited state transitions which were performed by using contactless electroreflectance spectroscopy.

CER spectroscopy, similarly as photoreflectance (PR), is known as an excellent, nondestructive and contactless tool to investigate optical transitions in semiconductor structures [13–16]. In contrast to PR spectroscopy, the CER technique does not use deep UV laser for photomodulation of built-in electric field inside the sample and therefore the problem of unwanted photoluminescence signal does not exist in CER technique. Due to the absorption-like character, CER probes the energy of the optical transition related to the ground and excited states in the QW, in contrast to photoluminescence, which probes mainly the ground state transition. The intensity of optical transitions observed in

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CER spectra is sensitive to the electron–hole overlap integral and its sensitivity to electric field.

So far, PR and CER have been successfully applied to study optical transitions in GaN-based QWs [11,17–20], but the intensity of CER resonances was not compared with the theoretical calculations. In this work, it is shown that the intensity of QW transitions is correlated with the electron–hole overlap integral derived from theoretical calculations. In addition, the theoretical analysis of electron–hole overlap integrals in a broad range of QW widths is discussed in this paper.

The samples used in this study were grown by plasma-assisted molecular beam epitaxy on Ga polar GaN/Al_2O_3 templates. After a 20 nm GaInN buffer layer, 40 (or 20) period AlInN/GaInN MQWs were grown and capped with undoped 100 nm GaInN. The nominal content for all GaInN layers is the same within the experimental error and equals ~11–13%. Two MQW samples with the same AlInN barrier thickness (3 nm) and various GaInN QW width (1.55 and 1.8 nm), and one MQW sample with the barrier thickness 6 nm and the QW width 1.55 nm were selected for this paper. The GaInN wells were uniformly doped with Si at a level of $10^{20}\,cm^{-3}$. The indium concentration in AlInN barriers is ~0.5%. High-resolution transmission electron microscopy (TEM) and X-ray diffraction (XRD) mapping techniques were used to study the structural quality of these samples. Both XRD and TEM results confirm smooth QW interfaces and high structural quality. More details on the growth procedure and structural characterization can be found in Refs. [8,22].

A conventional experimental set-up with a tungsten halogen lamp (250 W) as a probe light source, a 0.55 m monochromator and a photomultiplier was applied for obtaining contactless electroreflectance spectra. Samples were mounted in a capacitor with the top electrode made from a copper wire mesh which is semi-transparent for light. This electrode was kept at a distance of 0.1–0.3 mm from the sample surface while the sample itself was fixed on the bottom copper electrode. A maximum peak-to-peak alternating voltage of ~2.5 kV with frequency 285 Hz was applied. Phase sensitive detection of CER signal was made using a lock-in amplifier. Other relevant details of CER measurements are described in Ref. [21].

Figs. 1(a) and (b) show CER spectra for GaInN(L_w)/AlInN(3 nm) MQW samples with $L_w = 1.55$ and 1.8 nm, respectively. CER spectrum for the GaInN(1.5 nm)/AlInN(6 nm) MQW sample is shown in Fig. 2. The identification of CER resonances was possible due to theoretical calculations which are discussed in the next part of this paper. It is important to note that the barrier thickness for the first two MQW samples is twice smaller than for the last one. This difference significantly influences distribution of electric field inside the MQW (this issue is clarified in the next part of this paper). It means that CER spectra for samples with the same QW width and various barrier thicknesses can be significantly different as seen in Figs. 1(a) and 2. In addition, it is clearly visible that the intensity of 2H–1C transition, which is forbidden for non-polar square-like QWs, is significantly different from zero for all samples. Also, it is seen in Figs. 1 and 2 that the relative intensities of QW transitions vary with the QW width as well as the barrier thickness.

For all MQW samples CER resonances are very broad as it is expected for this material system. It is worth noting that the high broadening of CER resonance in In-containing III-N alloys is associated with indium incorporation and is attributed to In-related energy gap inhomogeneities in these alloys. In the case of narrow GaInN/AlInN QWs ($L_w < 3$ nm), the broadening of optical transitions increases additionally due to QW width fluctuations [20]. In a result, significant overlaps of CER resonances are observed in CER spectra. However, individual QW transitions can be extracted from CER spectra when the standard fitting

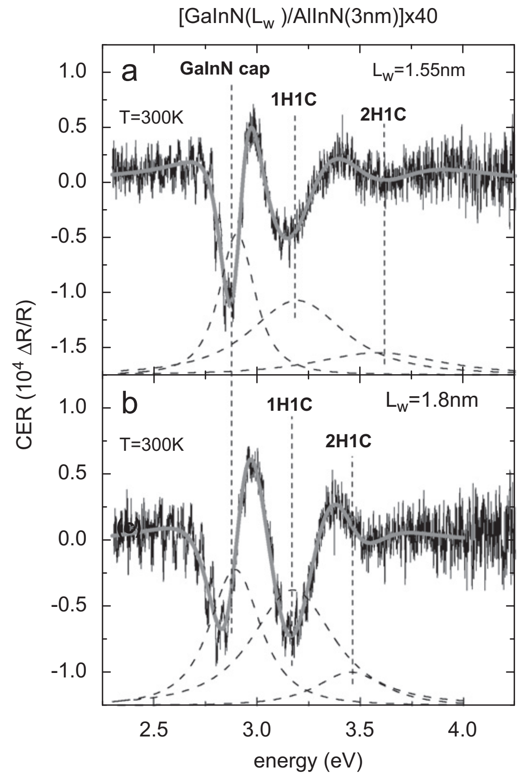

Fig. 1. Room temperature contactless electroreflectance spectra for (a) 1.55 nm and (b) 1.8 nm wide GaInN/AlInN multi-quantum wells (40 QWs).

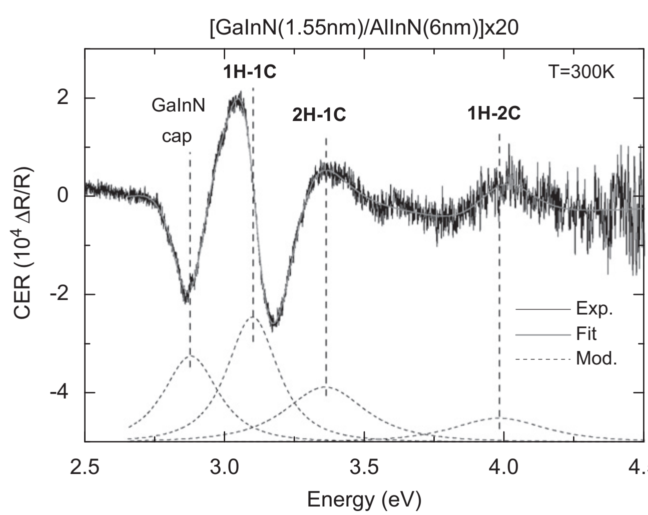

Fig. 2. Room temperature contactless electroreflectance spectra for 1.55-nm-wide GaInN/AlInN multi-quantum wells (20 QWs).

procedure is applied [11]. In our case, the Lorentzian lineshape model [23] has been used to describe the experimental data. According to this model CER spectrum can be fitted using the following formula:

$$\frac{\Delta R}{R}(E) = \mathrm{Re}\left[\sum_{j=1}^{n} C_j\, e^{i\vartheta_j}(E - E_j + i\Gamma_j)^{-m_j}\right], \qquad (1)$$

where n is the number of optical transitions and independent spectral functions used in the fitting procedure, C_j and ϑ_j are the amplitude and phase of the line, respectively, and E_j and Γ_j are the energy and the broadening parameter of the transition, respectively. In our case, it has been assumed that $m_j = 3$ for all optical