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EFFECTS OF PARABOLIC BARRIER DESIGN FOR MULTIPLE GaAsBi/AlGaAs QUANTUM WELL STRUCTURES

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The results of a comparative study on how the design of multiple quantum structures containing a parabolic barrier profile affects optical properties are presented. All quantum well (QW) structures were grown by molecular beam epitaxy (MBE) on semi-insulating GaAs substrates. The investigated samples consisted of (i) double parabolic quantum wells (type A) or (ii) multiple (two or three) rectangular quantum wells surrounded by parabolic barriers (type B). The optical quality of samples was characterized performing room-temperature (RT-PL) and temperature-dependent photoluminescence (TD-PL) measurements. The investigation aimed at the optimization of a multiple quantum well (MQW) structure design for application in the gain region of near infrared (NIR) laser diodes (LDs) revealed benefits of both double parabolic quantum wells and a mixed design (rectangular MQW with parabolic barriers). The PL band position for all samples was registered in the vicinity around 1.19 eV, which corresponds to the Bi content in QW of ~4.4%. It was shown that all structures of type A exhibit an intense emission, while the intensity of photoluminescence measured for the samples of type B depends on the number of QWs. The weaker intensity of the PL signal from two QWs inserted between parabolic barriers was explained by a larger point defect density at low temperature grown inner GaAs barriers. The room-temperature PL intensity of the structure with three GaAsBi QWs embedded in one parabolic AlGaAs barrier was the highest one. The shift of PL peak position to lower energies (1.16 eV) was attributed to the slightly higher bismuth concentration, 4.9%.

Keywords: GaAsBi, parabolic quantum wells, molecular beam epitaxy, photoluminescence, laser diode, near infrared

1. Introduction

Over the past decade, the near infrared region is extensively studied due to important and prospective environmental and medical applications. The so-called near-infrared–II (NIR-II) region (1000–1700 nm) is being exploited effectively by the telecommunications industry integrating the systems with various optoelectronic emitters and detectors. Lately, the NIR-II region in comparison to the NIR-I region (650–900 nm) is gaining more popularity in the biosensing scene as the longer wavelengths exhibit reduced scattering and attenuation by human tissue, greater penetration depths and improved signal-to-background ratio [1]. A great candidate for the improvement of the performances of already existing biosensing devices is gallium arsenide bismide (GaAsBi).

In recent years, GaAsBi material has been under investigation for its potential applications in various NIR optoelectronic devices. The incorporation of bismuth (Bi) into the classical semiconductor gallium arsenide (GaAs) has numerous desirable effects. Only a few percent of bismuth introduced into the lattice of GaAs significantly reduces the bandgap of bismide [2]. The reduction of bandgap in the bulk material by 88 meV/$\%_{Bi}$ was reported [3]. As a result, a redshift of the photoluminescence peak with an increasing concentration of bismuth is observed. The incorporation of Bi also affects the bandgap dependence on temperature. The GaAsBi bandgap showed increased resistance to temperature changes compared to GaAs and other conventional semiconductors, like InGaAs or GaAsSb [4, 5]. Moreover, with the increase of Bi concentration, the spin-orbit split-off energy (Δ_{SO}) of GaAsBi rapidly increases [6]. Furthermore, theoretical and experimental results showed that GaAsBi structures containing the Bi content above 10% undergo a large valence band spin-orbit splitting [2]. This in turn could result in the suppression of Auger recombination in structures containing bismuth.

Due to its desirable properties, such as a fast reduction of the bandgap, GaAsBi can be integrated as active media of light emitting diodes (LEDs) [4], photodetectors (PDs) [7], laser diodes (LDs) [8], solar cells [9], etc. The lower bandgap sensitivity on temperature in bismides allows for the reduction of additional cooling solutions for devices, consequently, reducing production costs and the size of the final optoelectronic device or a system. These properties make bismides very attractive for applications in telecommunication 'windows' II and III, especially, for LDs emitting at wavelengths of 1.3 and 1.55 μm [2]. For this purpose, GaAsBi quantum wells (QWs) can be used as an active medium in Fabry–Perot (FP) NIR laser diodes. Such laser diodes exhibit a narrow spectral emission, which is strictly required in sensors for the detection of selected materials and molecules. However, the manufacturing of emitters featuring GaAsBi QWs poses different technological and technical challenges. The specific epitaxy of GaAsBi – a very low temperature and the stoichiometric arsenic to gallium beam equivalent pressure ratio (BEPR) – must be optimized to employ multiple quantum wells (MQWs) of bismide in a fully operational device. Thus, scientists investigate different concepts to reach the technological repeatability and transfer to industry. One of possible approaches to achieve a working optoelectronic device is implementing various architectural designs of the active medium. A successful implementation of graded-index barriers while manufacturing a laser diode was already reported by Tsang in 1981 [10]. In 2019, our group showed that GaAsBi single quantum wells with parabolically graded barriers (PGBs) exhibited a boost in photoluminescence at room temperature at least by 50 times, compared to conventional rectangular QWs [11]. Such results were explained by an increase of carrier trapping efficiency due to the parabolically graded design of the barriers and carrier localization. Yet another benefit of a high carrier localization effect in PGBs due to the blurred well–barrier interface is more reproducible optical properties within a wider range of growth conditions. Despite the great progress in technology and fundamental investigation, several questions are still open, and a detailed study of the QW number influence on bismide-based LD performance is necessary. Meanwhile, different opinions about related AIII-BV LDs are under discussion. Nakamura et al. [12, 13] demonstrated the results obtained on InGaN MQW structure laser diodes. They pointed out that the lowest threshold current density of LD is related to the emission wavelength, and the optimal QW number varies. Moreover, the authors concluded that the substrate material significantly affects threshold current density.

Thus, the analogies of processes and behaviour in InGaN compound based LDs allows us to preview possible problematic scenarios. Taking into account the specifics of Bi atoms, namely, the size, the trend to segregate at high temperatures and surfactant features, the LD growth protocol must be planned precisely and the undesired degradation of GaAsBi well layer must be previewed and avoided. The lack of knowledge and practice could lead to an increase of the internal loss of the LD with a subsequent increase in the threshold current density.

In this work, two designs of parabolic quantum wells (PQWs), including a double parabolic quantum well (DPQW) and multiple rectangular quantum wells surrounded by parabolic barriers (2inPQW and 3inPQW), were grown and investigated. The impact of the design and QW number was considered. Both types of heterostructures were grown by molecular beam epitaxy (MBE). The optical quality of samples was characterized performing room-temperature and temperature-dependent photoluminescence measurements. The study was concentrated on the influence of MQW design on emission. A comparative study was aimed to reveal the benefits of both PQW based and mixed designs (rectangular QWs in PGB).

2. Growth of the structures

The GaAsBi/AlGaAs PQW structures were grown using a solid-source, *Veeco* GENxprlor R&D MBE system containing cells of high purity 7N5 metallic Ga, Al and Bi sources, and an adjustable valve As source equipped with a cracker to provide a dimeric arsenic flux. The structures were grown on semi-insulating GaAs (100) substrates, the temperature of which was monitored/adjusted using the heater thermocouple readings.

Prior to the growth, the substrate preparation procedure was carried out. The substrate was loaded into a load lock chamber and heated for 2 h at 200°C to start the degas process. After that, the substrate was heated for an hour in a buffer chamber at 300°C, and then transferred to a growth chamber for the last heating at 700°C for ~15 min with the maximum arsenic flux supplied. This thermal treating was performed to deoxidize the GaAs wafer.

The bismide quantum well growth conditions were determined by the optical characteristics, targeted to the emission wavelength interval of about 1000–1200 nm, characteristic of footprints of many organic molecules. The growth of bismides requires unique epitaxy conditions, namely, the low substrate temperature that was chosen to be 425°C in this work and BEPR of As/Ga close to the unity. The issue of a stable As/Ga BEPR becomes more significant during longer, several hour lasting growth procedures. It must be noted that Bi is prone to segregation, meaning that AlGaAs and GaAs barriers on top of the GaAsBi QWs had to be grown at the lowest possible temperature. However, the low barrier growth temperature introduces even more defects further reducing the optical properties of the sample. Those technological challenges defined two different designs of a parabolic barrier architecture in this work. Aiming both to study the comparable optical properties and to make the growth procedure easier, less time consumable, two different types of structures were designed: (i) two rectangular GaAsBi QWs each surrounded by parabolic AlGaAs barriers (type A, DPQW) and (ii) multiple (2 and 3) rectangular GaAsBi QWs separated by GaAs barriers and sandwiched between parabolic AlGaAs barriers (type B, 2inPQW and 3inPQW, respectively). The sketches of different designs of QW structures are presented in Fig. 1. For both types of samples, the outer parabolic profiles were grown by varying the Al content in the AlGaAs from 30 to 0% and *vice versa*; this design was described in our earlier articles [11, 14]. One can point out that both the inner parabolic AlGaAs barrier profile (type A) and standard rectangular GaAs barriers (type B) were grown at the temperature typical of bismide growth. To reduce bismuth segregation, the final parabolic barrier of type A and B structures was also done at low temperature, and only the capping layer was grown at high temperature. For particular growth conditions and temperature profiles of two types of MQW designs used in this work see Table 1. Quantum structures are very susceptible to the abovementioned issues: firstly, the low temperature growth of GaAsBi results in the highly disordered material in QWs, secondly, the extended sample growth time requires a precise control and stability of atomic fluxes. As a consequence, two undesirable effects could occur: the reduction of emission intensity (non-radiative point deffects) and the inhomogenous Bi incorporation into the GaAs lattice (change in BEPR of As to Ga), resulting in a wider emission spectrum. It was assumed that an alternative design introduced in this

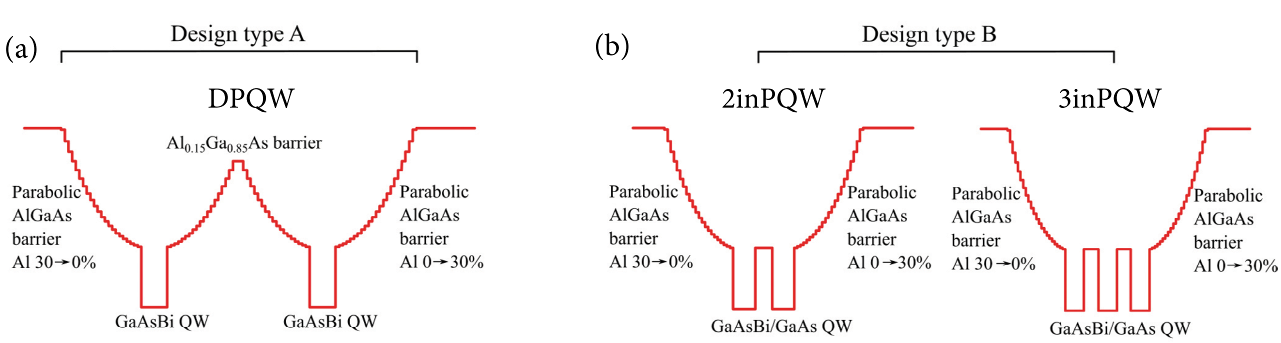

Fig. 1. Sketches of the designs of multiple GaAsBi QW with parabolically graded AlGaAs barriers. (a) Design type A: DPQW structure is two GaAsBi QWs with AlGaAs PGBs; (b) design type B: 2inPQW and 3inPQW structures are two or three GaAsBi QWs embedded together in one AlGaAs PQW.

Table 1. Growth protocols of the investigated structures starting from top of the sample to the substrate. d is the width of the layer (nm) and T_g is the temperature of growth (°C). PB means parabolic barrier.

DPQW (top → substrate)	d, nm	T_g, °C	2inPQW (top → substrate)	d, nm	T_g, °C	3inPQW (top → substrate)	d, nm	T_g, °C
GaAs cap	5.5	600	GaAs cap	5.5	600	GaAs cap	5.5	600
AlGaAs spacer	100	600	AlGaAs spacer	100	600	AlGaAs spacer	100	600
AlGaAs PB	32	425				AlGaAs PB	32	425
GaAsBi QW	10	425	AlGaAs PB	32	425	GaAsBi QW	10.2	425
AlGaAs PB	32	425	GaAsBi QW	10.2	425	GaAs barrier	10	425
AlGaAs barrier	30	425	GaAs barrier	10	425	GaAsBi QW	10.2	425
AlGaAs PB	32	425	GaAsBi QW	10.2	425	GaAs barrier	10	425
GaAsBi QW	10	425	AlGaAs PB	32	600	GaAsBi QW	10.2	425
AlGaAs PB	32	600				AlGaAs PB	32	600
AlGaAs spacer	100	600	AlGaAs spacer	100	600	AlGaAs spacer	100	600
			GaAs buffer	2	600	GaAs buffer	2	600
GaAs buffer	135	650	GaAs buffer	110	665	GaAs buffer	110	665
GaAs substrate								

work (type B) with all GaAsBi QW layers grown in close proximity could reduce the growth time by 25% and ensure the stability of element fluxes. Such architecture is expected to have a better crystalline quality and a more homogeneous composition of GaAsBi layers, resulting in a higher intensity and narrower emission spectra.

3. Optical characterization and discussion

Firstly, room-temperature photoluminescence (RT-PL) measurements were carried out for both types of structures, type A (DPQW) and type B (2inPQW and 3inPQW), to determine the spectral region of emission energy. Taking into account that bismides are highly disordered compounds, and using our achievements on the parabolic architecture of MQW, the pumping of carriers at the barriers of the structure was considered. For the experiment, a diode-pumped-solid-state (DPSS) laser with a wavelength of 532 nm and a power density of 5 kWcm^{-2} was used as an excitation source. The PL signal was registered by an InGaAs thermoelectrically cooled photodetector. The obtained RT-PL spectra are depicted in Fig. 2. One can see from the plot that all registered RT-PL spectra consist of one PL band at the investigated energy range, and the observed PL signal is assigned to be characteristic of optical transitions in GaAsBi QWs. Comparing the spectra of structures, containing 2 GaAsBi QWs, DPQW (type A) and 2inPQW (type B), we point out that the PL intensity of GaAsBi QWs embedded in separated parabolic AlGaAs barriers (DPQW) is almost two times in magnitude higher than the emission intensity from two standard rectangular shape GaAsBi/GaAs QWs grown between parabolic AlGaAs barriers. The numerical calculation demonstrated that the energy of the ground state transition (1e1hh) in GaAsBi QW is not affected by the design of parabolic AlGaAs barriers. The PL band position for both DPQW and 2inPQW structures is similar and registered in the vicinity

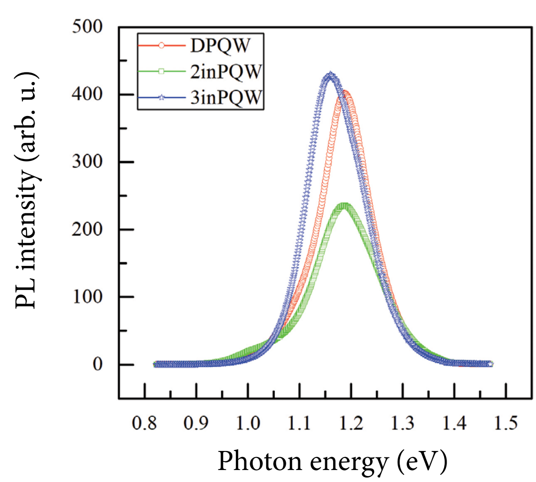

Fig. 2. PL spectra of the investigated DPQW, 2inPQW and 3inPQW structures measured at room temperature under excitation of 5 kWcm^{-2}.

around 1.19 eV. Since the width of QWs is the same for both DPQW and 2inPQW structures, the calculated Bi content in QW is around 4.4%. It is supposed that the lower intensity PL signal in 2inPQW is related to the low temperature inner GaAs barrier and the growth conditions could initiate a larger concentration of substitutional As_{Ga} defects, what means that As to Ga BEPR is very important and needs a more detailed investigation for the optimization of MBE growth conditions. The RT-PL intensity of the structure with 3 GaAsBi QWs embedded in one parabolic AlGaAs QW is the highest one. The PL peak position is shifted to lower energies (1.16 eV); thus, the calculated Bi content is slightly higher and reaches 4.9%. The analysis of registered spectral characteristics allows one to suppose that a slightly shifted peak position of DPQW and 2inPQW compared to that of 3inPQW could be attributed to the technical issue of stability of MBE sources and/or the poor quality of lower/higher (low for AlGaAs barrier and high for GaAs barrier) than necessary arsenic content in both low temperature barriers. An insignificant asymmetry in the 2inPQW spectrum could be originating from slightly different Bi contents (4.4 and 4.9%) in two GaAsBi QWs, and band to band transitions in the better quality QW are dominating. For this reason, two rectangular GaAsBi QWs could be an insufficient number of QWs needed for the efficient radiative carrier recombination in a LD. The enhanced PL intensity of 3inPQW could confirm our hypothesis, meanwhile the lower peak energy position needs future investigations. Possible explanations involving BEPR of As to Ga and the substrate temperature stability could be discussed.

Temperature-dependent photoluminescence (TD-PL) measurements were carried out using a low excitation intensity of 2 Wcm^{-2} in a temperature range of 3–300 K. The TD-PL spectra of DPQW, 2inPQW and 3inPQW structures are presented in Fig. 3. The spectra were normalized and shifted vertically for clarity. It can be seen from

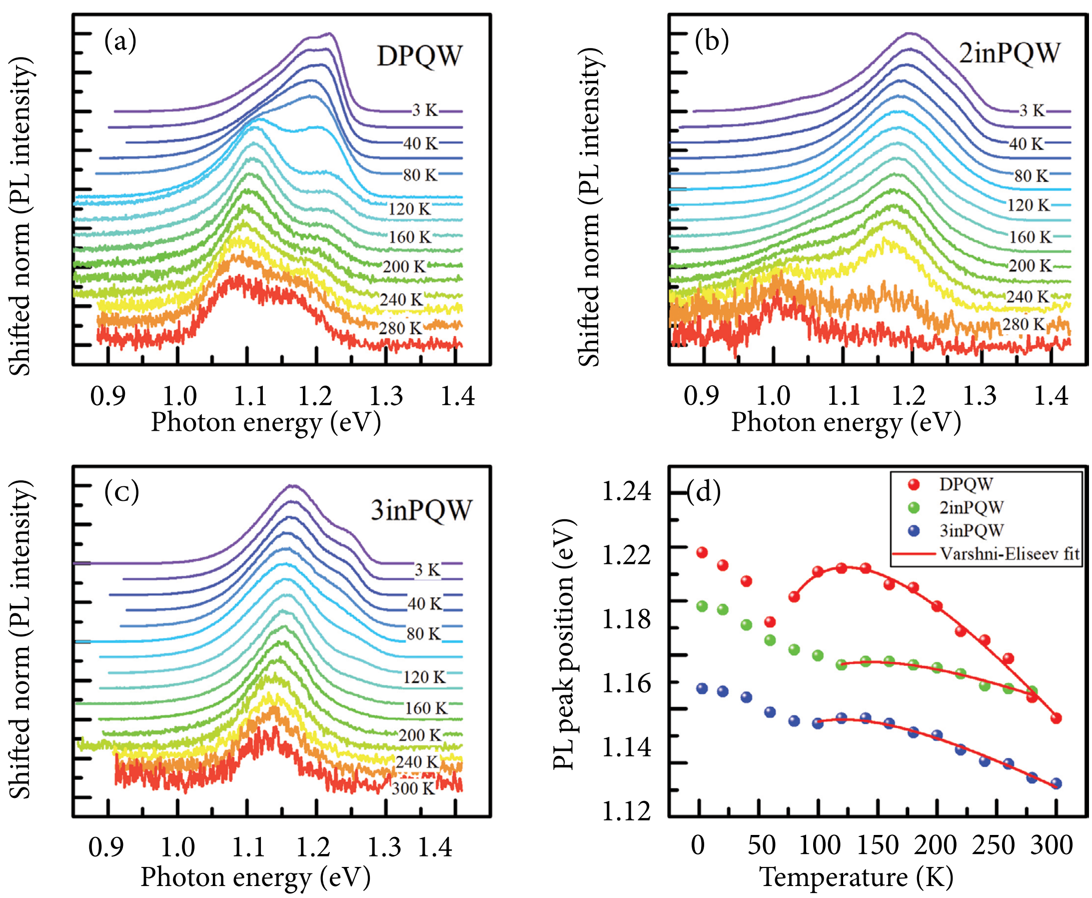

Fig. 3. Temperature-dependent PL spectra measured in a temperature range of 3–300 K of the DPQW (a), 2inPQW (b) and 3inPQW (c) structures and the PL component position dependence on temperature (d). Measurements were carried out under excitation of 2 W/cm^2.

Fig. 3 that the PL spectra of all structures measured in the low temperature range are broadened. This spectral change could be associated with the lack of homogeneity of the Bi content or even QW thickness in the structures containing more than one QW. Overall, the observed peak shifts with temperature following the S-shape, which indicates the localization of carriers in all three cases. These measurements support the discussion of RT-PL results.

The PL peak positions as a function of temperature are plotted in Fig. 3(d) for all investigated QW structures. The presence of red-blue-red shift (S-shape) in the PL peak positions of the structures under investigation is clearly observed and the origin of this dependence can be explained by the distribution of carriers between the localized states at different temperatures. At low temperatures, the electron–hole pairs are trapped in the shallow localized states because of their small thermal energy. With the increase of temperature the carriers gain enough energy to escape the shallow localized states and occupy deeper level localized states. This results in the redshift of PL peak positions. As the temperature further increases the electron–hole pairs obtain more energy and become capable of completing multiple hops between the states and occupy those which were empty at lower temperatures. In the PL peak position dependence on the temperature this corresponds to a blueshift. At temperatures close to RT, a usual redshift is present due to the fundamental decrease in the bandgap with increasing temperature.

In order to analyze the blueshift of the peak, the experimental data of the measurement was fitted by the Varshni–Eliseev function [15]

$$E_{\mathrm{p}}\left(T\right)=E_{0}-\frac{\alpha T^{2}}{\beta+T}-\frac{\sigma^{2}}{k_{\mathrm{B}}T}\,, \qquad (1)$$

where E_0 is the bandgap at 0 K, α is the Varshni fitting parameter, which describes the rate of bandgap reduction with temperature [meVK^{-1}], β is another Varshni fitting parameter, which in this work was fixed to 204 K – the β value for GaAs [15], k_B is the Boltzmann constant (8.617×10^{-5} eVK^{-1}), and σ is the Eliseev fitting parameter, which corresponds to the dispersion of the Gaussian density of localized states. The best fitting parameters are shown in Table 2.

The temperature dependence of the PL spectra of DPQW differs from the spectra of type B MQW structures. First, there are two well-distinguishable PL bands in all investigated temperature ranges, and the PL intensity of those bands changes differently with temperature. Such behaviour could indicate that those PL bands could correspond to different QWs in the structure. The scenario of QW with a different Bi content in the GaAsBi layer is possible: the lower energy PL band could be related to the Bi content of 6.3%, while the higher energy PL band to 4.9% of Bi. The different Bi concentration at different GaAsBi QWs in the DPQW structure is highly feasible since QWs are far apart from each other (almost 100 nm). The growth conditions (most likely As/Ga BEPR) could change over the time. Also, the substrate temperature due to the significant thickness of PQW could be lower during the growth of the second QW. It is known from the literature that the substrate temperature is very important for bismuth incorporation, thus the reduction of growth temperature by even tens of degrees could cause more favourable conditions for a larger Bi incorporation into the QW. Furthermore, there is a faster reduction of the type A sample bandgap energy with temperature, which could be related to a much higher carrier localization in the DPQW structure (23 meV) if compared to those of the type B structures 2inPQW (16 meV) and 3inPQW (15 meV). Note that the DPQW structure also shows more intense PL than 2inPQW at room temperature

Table 2. The best fitting parameters of the PL peak position versus temperature curve using the Varshni–Eliseev equation (1).

Type of structure	E_0, eV	α, meVK^{-1}	β, K (fixed)	σ, meV
DPQW	1.29±0.01	0.63±0.03	204	23±1
2inPQW	1.21±0.01	0.22±0.03	204	16±2
3inPQW	1.19±0.01	0.28±0.02	204	15±1
GaAs[18]	1.52	0.54	204	0

(Fig. 2). A relation between the highly expressed localization and the more intense PL spectrum at room temperature was shown in our previous work [11]. Since the thermal energy of carriers at RT is 26 meV, the localization effect is of great importance even at room temperature. The higher carrier localization in the DPQW design is likely caused by the blurred well–barrier interface due to the parabolic Al content decrease in the barrier before the GaAsBi QW. Meanwhile in the case of 2inPQW and 3inPQW designs, the inner rectangular GaAs barriers could ensure sharper well–barrier interfaces. Moreover, it was demonstrated in transmission electron microscopy (TEM) pictures pointing to sharp single or multiple rectangular GaAsBi QWs and low temperature GaAs barrier interfaces [16, 17]. However, further studies are required to confirm this explanation.

4. Conclusions

In this study, we present a comparative analysis of the impact of the parabolic AlGaAs barrier design on multiple GaAsBi quantum well (QW) structures. Two types of molecular beam epitaxy grown QW structures were examined: (i) double GaAsBi/AlGaAs parabolic quantum wells (referred to as type A, sample DPQW) and (ii) multiple GaAsBi/GaAs rectangular quantum wells surrounded by AlGaAs parabolic barriers (referred to as type B, samples 2inPQW and 3inPQW). Numerical calculations revealed that the design of the AlGaAs parabolic barrier profile does not affect the energy of optical transitions between the ground states of GaAsBi QWs embedded in AlGaAs parabolic quantum wells. Consequently, the PL band observed around 1.19 eV at room temperature for the investigated structures was attributed to optical transitions in GaAsBi QWs with the 4.4% Bi content. It was observed that the DPQW structure (type A) consistently exhibited an intense room temperature emission, likely due to a high carrier localization within those GaAsBi QWs. Although, temperature-dependent PL measurements revealed a lower QW homogeneity in that type A QW structure if compared to those of type B structures. The PL intensity measured for the type B structures depended on the number of QWs. The weaker PL intensity from two rectangular GaAsBi QWs embedded in one AlGaAs parabolic quantum well was attributed to the lower quality of the inner GaAs barrier grown at low temperature. However, the PL intensity of the structure with three rectangular GaAsBi QWs embedded in one parabolic AlGaAs quantum well was highest at room temperature. To conclude, our investigation demonstrated that the type B design, specifically 3inPQW, is more suitable for laser diode (LD) or light-emitting diode (LED) gain areas due to several technological advantages: (i) easier design, (ii) 25% faster epitaxy process and (iii) lower costs. However, further optimization and investigation of the number of QWs in B-type structures is needed.

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PARABOLINIO BARJERO DIZAINO POVEIKIS DAUGYBINĖMS GaAsBi/AlGaAs KVANTINĖMS DUOBĖMS

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Pristatomi daugybinių GaAsBi kvantinių duobių su skirtingais parabolinių barjerų dizainais tyrimų rezultatai. Molekulinių pluoštelių epitaksijos metodu ant pusiau izoliuojančių GaAs padėklų buvo užaugintos GaAsBi daugybinės kvantinės duobės su paraboliniais barjerais. Pastarasis dizainas turi tokius privalumus: geresnis optinių savybių atsikartojamumas, stipresnė krūvininkų lokalizacija bei geresnis krūvininkų sugavimo efektyvumas. Pasirinkti trys skirtingi parabolinių barjerų dizainai: dvi GaAsBi kvantinės duobės apgaubtos AlGaAs paraboliniais barjerais bei dvi arba trys GaAsBi stačiakampės duobės įterptos tarp išorinių parabolinių AlGaAs barjerų. Užaugintų struktūrų optinės savybės buvo tiriamos matuojant kambario temperatūros liuminescensiją ir fotoliuminescencijos priklausomybę nuo temperatūros. Tyrimų rezultatai parodė, kad kelios kvantinės duobės, apribotos paraboliniais barjerais, gali pasiekti panašų emisijos intensyvumą palyginti su daugybiniais paraboliniais barjerais, ribojančiais po vieną kvantinę duobę.