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PHOTOLUMINESCENCE PROPERTIES OF GaAsBi SINGLE QUANTUM WELLS WITH 10% OF Bi

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A set of single quantum well (SQW) samples of $GaAs_{1-x}Bi_x$ with $x \sim 0.1$ and *p*-doped GaAs barriers grown by molecular beam epitaxy was investigated by the temperature-dependent photoluminescence (PL) spectroscopy. Those GaAsBi SQW structures showed a high crystalline quality, a smooth surface and sharp interfaces between the layers and exhibited a high PL intensity and a lower than 100 meV PL linewidth of QW structures. Temperature dependence of the optical transition energy was S-shape-free for all investigated structures and it was weaker than that of GaAs. An analysis of the carrier recombination mechanism was also carried out indicating that the radiative recombination is dominant even at room temperature. Moreover, numerical calculations revealed that a higher Be doping concentration leads to an increased overlap of the electron and heavy hole wave functions and determines a higher PL intensity.

Keywords: molecular beam epitaxy, quantum wells, bismides, photoluminescence

1. Introduction

Development of new semiconductor materials has often led to important technological transformations. A recent example is commercial advances in solid-state lighting related to group III-nitride compounds. Before Nobel-prize winning discoveries, nitrides were a relatively insignificant area in the materials research. Their success raised a wide interest in the group III–V compounds from the opposite end of the periodic table, namely, group III-bismides.

Bismuth is the heaviest nonradioactive atom in the V group, therefore, its introduction into the III–V semiconductor alloy lattice leads to a large reduction of the energy band-gap. Bismides can be considered as a good alternative to dilute nitrides, because Bi introduction into GaAs results in a lower lattice mismatch than that for N. Also, the ternary compound GaAsBi is very attractive due to a faster band-gap reduction than in classical InGaAs and GaAsSb materials, the most widely used for fabrication of infrared (IR) sources. It was demonstrated previously that the incorporation of 1% of Bi into the GaAs lattice decreases the band-gap of GaAsBi up to 88 meV [1]. Moreover, dilute bismide alloys exhibit a low temperature band-gap coefficient [2], therefore, bismide-based IR sources could operate without additional cooling. Finally, it is worth noting that the spin–orbit split-off energy (Δ_{SO}) increases for higher Bi compositions in GaAs [3]. In particular, GaAsBi alloys with Bi content larger than 10% experience a large valence band spin–orbit splitting that exceeds the energy band-gap (E_g). As a result, inter-valence-band absorption and dominant Auger recombination processes in bismide-based components can be suppressed. These features force an investigation of GaAsBi as

an active region for IR light emitting diodes (LEDs) and lasers, resulting in reduced power consumption for a wide range of applications.

It has been already demonstrated that GaAsBi multiple quantum wells (MQW) with 10% of Bi exhibit room temperature photoluminescence at the wavelengths as long as 1.43 μm [4]. A proper design of the structures can enable enhancement of the luminescence up to a factor of 50 [5]. A complex study revealed that the technological parameters of molecular beam epitaxy (MBE) growth such as Bi flux and the group III to V pressure ratio play an essential role in obtaining high quality bismide quantum well (QW) structures and that appropriate conditions allow one to introduce a larger content of Bi while still keeping pseudomorphic compressively strained QWs.

Up to now, LEDs and laser diodes (LDs) have been realized only using GaAsBi as an active layer. The first edge-emmiting Fabry–Perot LD with GaAsBi bulk layer (4% of Bi) was demonstrated by the Kyoto group in 2010 [6]. Ludewig et al. realized the first electrically pumped room temperature GaAsBi/GaAs single quantum well (SQW) LD at a wavelength of 947 nm [7]. The developed technology of GaAsBi/GaAs MQW with 6% of Bi allowed one to fabricate LDs emitting at 1060 nm [8]. In later experiments, those Fabry–Perot type devices exhibited the lasing wavelength increase up to 1200 nm at low temperatures. However, the further progress was blocked by the appearance of a large number of voids traversing the active layers with a higher Bi content resulting in increased optical losses and disappearance of lasing. Currently, the longest wavelength of 1141 nm at room temperature was shown for electrically injected GaAs/GaAsBi single quantum well laser diodes [2].

As a rule, the MBE growth of GaAs and other related to AIII–BV family compounds requires the high substrate temperatures reaching $T \sim 540$–615°C and the arsenic over-supply keeping ratio of As/group-III from 7 to 60 times depending on the compound (characteristic ratio for GaAs epitaxy is about 7–10, for AlAs it is starting from 30; depends on the growth temperature). Meanwhile, bismide growth at low temperatures and nearly stoichiometric As_2/Ga flux ratios, required for Bi introduction, results in a significantly reduced crystalline quality. Thus, the introduction of more than 5% of Bi to the GaAs lattice, while avoiding defect formation (Bi-clusters) and accumulation of Bi droplets on the surface, is still challenging [9, 10]. Consequently, the enhancement of the quality of GaAsBi through the optimization of growth conditions is absolutely necessary in order to achieve effective IR sources and improve their performance.

In this work, we present the growth and study of optical properties of GaAsBi SQW with 10% of Bi (composition is close to a window, where spin–orbit splitting Δso value approaches E_g), which will be exploited for the determination of the main technological parameters and their influence on luminescent properties. The main criteria for GaAsBi QWs were the following: (i) Bi content around 10% in GaAs crystal host lattice, (ii) high crystalline structure quality QWs, which are pseudomorphic with respect to barrier layers, (iii) surface roughness less than 1 nm, and (iv) intense photoluminescence (PL) within the wavelength range from 1.0 to 1.3 μm.

2. Growth and structural characterization of quantum wells

The samples were grown using a solid source MBE system SVT-A. The reactor was equipped with conventional Knudsen effusion cells for metallic Ga and Bi as well as a unique design source for arsenic containing two independently controlled thermal zones for the bulk evaporator and cracking head for generating pure As_2 flux. The 'epi-ready' semi-insulating (SI) GaAs wafers oriented in the (001) crystallographic plane were heated at temperatures up to 300°C for 3 h in the load-lock and buffer chambers to desorb water vapour from the substrates, then they were transferred to the growth chamber. The base pressure in the growth chamber was in a range of $2–5 \times 10^{-10}$ Torr. The growth temperature was monitored by thermocouple readings with an accuracy of 1°C. Beam equivalent pressures (BEP) of Ga, Bi and As_2 were measured by a retractable ion gauge. Prior to the GaAsBi QW growth, the native oxide from SI-GaAs substrate was outgassed at 700°C for 30 min under the maximum arsenic flux. Cleaning of wafers and the surface during growth was monitored *in situ* by reflection high-energy electron diffraction (RHEED). A weak (2 × 4) RHEED pattern evidenced that the native oxide was desorbed.

Usually, MBE grown background doping of GaAs is of *p*-type and carrier density is in

a range of 10^{13}–10^{14} cm^{-3}. GaAs layers grown in a SVT-A reactor exhibit *n*-type with carrier density higher by more than one order of magnitude, ~5×10^{15} cm^{-3}. In order to have *p*-type barrier layers beryllium was used. The concentration of Be doping varied from 5×10^{17} to 3×10^{18} cm^{-3} using a growth rate of about 330 nm/h. The RHEED pattern following this procedure usually showed a strong 2 × 4 reconstruction indicating a smooth surface and layer-by-layer growth. Barriers of QW were grown at 450°C. To increase Bi incorporation and to avoid Bi segregation to the QW surface, the substrate temperature was decreased down to temperatures of 300–307°C and exposed under Bi flux for 10 s. The flux ratio of As_2/Ga was kept in an interval from 1.074 to 1.084 BEP. The QW width varied from 6 to 7 nm and the width of barrier layers was set to 30 nm. The summary of technological growth conditions and characterization parameters for the investigated GaAsBi SQWs is presented in Table 1.

The surface roughness of GaAsBi SQWs has been characterized *ex situ* by atomic force microscopy (AFM) measurements using a Dimension 3100 SPM system with a Nanoscope IVa controller (*Veeco Instruments Inc.*, USA). The high-resolution measurements of microstructure were carried out by a FEI Tecnai G2 F20 X-TWIN transmission electron microscope (TEM) with the scanning transmission electron microscopy (STEM) module, equipped with an X-ray energy dispersive spectroscopy (EDS) detector for elemental mapping and a high angle annular dark-field (HAADF) detector for Z-contrast imaging. In addition, a FEI *Helios* Nanolab 650 dual beam microscope equipped with an Omniprobe manipulator was used to prepare specimens in order to analyse the cross-sectional microstructure of SQWs.

The surface images of GaAsBi SQW structures are shown in Fig. 1. The AFM investigations revealed that the surface roughness varied from 0.15 to 0.5 nm depending on the As_2 to Ga flux ratio

Table 1. Main technological parameters of GaAsBi/GaAs:Be SQWs. Bi, the content evaluated from PL measurements (%); As/Ga, As to Ga BEPR; T_{QW}, the growth temperature of quantum well (°C); d_{QW}, the thickness of quantum well (nm); N_{Be}, the concentration of Be doping (cm^{-3}); E_p and I_{PL}, the room temperature peak position of the GaAsBi QW PL band (eV) and PL intensity at the peak position (a.u.), respectively.

Sample No.	Bi, %	As/Ga BEPR	T_{QW}, °C	d_{QW}, nm	N_{Be}, cm^{-3}	E_p, eV	I_{PL}, arb. u.
B367	10.1	1.080	300	7	5E17	0.911	3.5
B369	11.0	1.076	300	7	1E18	0.867	1.9
B372	10.2	1.084	307	6	3E18	0.919	20.9

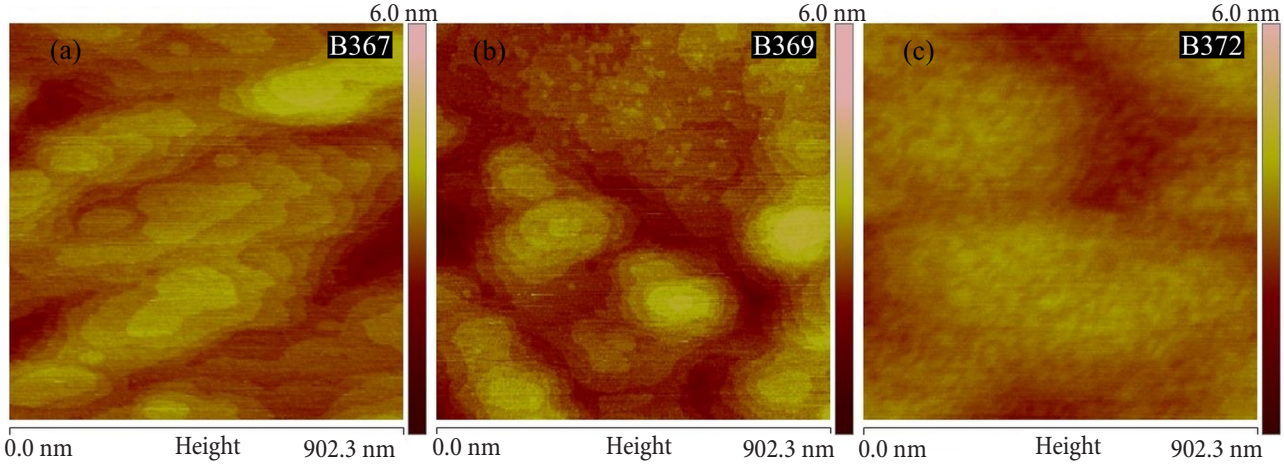

Fig. 1. (Coloured online) The surface morphology images of three GaAsBi SQW samples, grown varying the As_2/Ga pressure ratio in a range of 1.076–1.084 and the substrate temperature from 300°C (a, b) to 307°C (c), obtained by an atomic force microscope. The colour scale of 6 nm resolution demonstrates the surface roughness measured from peak (min) to peak (max).

and Bi flux. It should be noted that the smoothest surface with atomical terraces was observed for sample B367 grown at the lowest temperature (see Fig. 1(a)). Both the larger Bi content (sample B369) in GaAsBi SQW and the higher structure growth temperature (sample B371) lead to an increase in surface roughness (Figs. 1(b, c)).

Figures 2(a–c) represent the cross-section TEM images of all investigated GaAsBi/GaAs:Be QWs structures. The interfaces of QWs are quite smooth and the distribution of Bi within all QWs is homogenous. The atomic columns distinguished in the zoomed TEM image of sample B372 (Fig. 2(c)) demonstrate a high crystalline quality of both GaAs:Be barriers and GaAsBi (SQW) grown at such low temperatures. Moreover, the interfaces are free from Bi or Bi-Ga complex droplets.

3. Photoluminescence measurements and discussion

Complex study was performed to clarify the effect of technological parameters on the optical properties of bismide SQWs. PL measurements were carried out in a temperature range of 3–300 K. In order to change temperature, the samples were mounted on the cold finger of a closed-cycle He cryostat coupled with a temperature controller. A diode-pumped solid-state laser emitting at a wavelength of 532 nm was used as an excitation source at different excitation powers. PL signal was detected with a thermoelectrically cooled InGaAs photodetector using a conventional lock-in system.

Firstly, the content of bismuth in the QWs was estimated by fitting the calculated optical transition energies to the measured PL spectra at room temperature. Simulations were performed with the *nextnano*[3] software [11] within a single-band effective-mass approximation by solving self-consistent Schrodinger–Poisson equations. Material parameters for GaAsBi were taken from Ref. [12].

The temperature-dependent PL spectra of GaAsBi/GaAs:Be SQW structures with Bi-content of 10.1, 11 and 10.2% are shown in Figs. 3(a–c). As seen, all three samples emit light at around 1300 nm (0.95 eV). Low temperature PL spectra of the samples B367 (10.1% Bi) and B369 (11% Bi) consist of two PL bands. The difference between the maxima positions of these PL bands are of 77.1 meV (B367) and 70.0 meV (B369) at 3 K. These values are comparable with the numerically calculated difference between the states of heavy-hole (*hh*) and light-hole (*lh*): 80 meV for sample B367 and 65 meV for sample B369. The excitation-dependent PL measurements at low temperatures (not presented here) also support the assumption that the higher energy PL band can be assigned to the emission from higher QW states (optical transition 11l). Lower PL intensity and energy of the PL peak of the sample B369 can be associated with a higher Bi concentration in GaAsBi QW (11%). The photoluminescence spectra of sample B372 display the highest PL intensity (20.9 arb.u. at room temperature) and has only one PL band (0.994–0.919 eV) in the whole temperature range. The calculated splitting of *hh* and *lh* states is 55 meV for sample B372. One can note that it is difficult to distinguish emission from

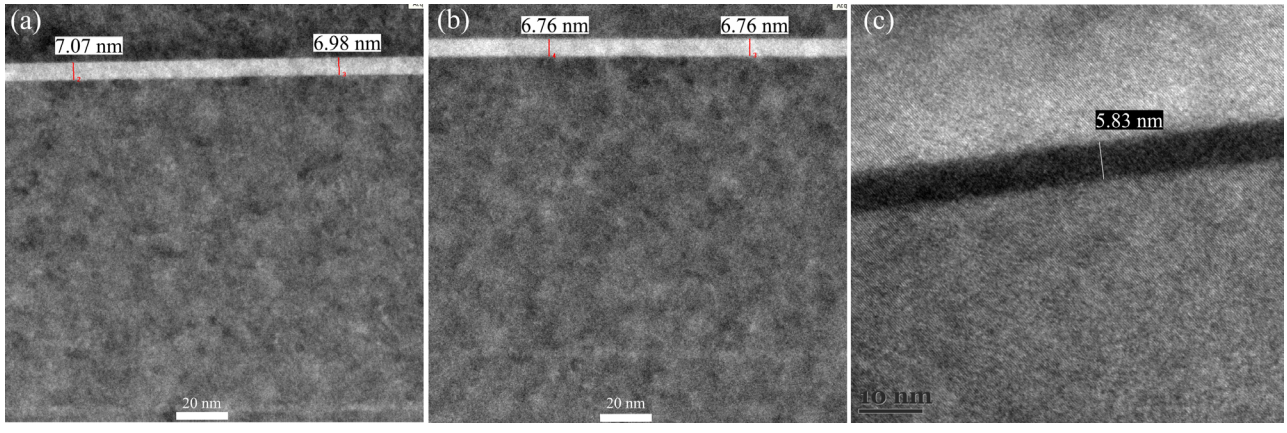

Fig. 2. The cross-section TEM image of three GaAsBi SQW containing samples, grown varying the As_2/Ga pressure ratio in a range of 1.076–1.084 and the substrate temperature from 300°C (a, b) to 307°C (c). The scale bars in pictures (a) and (b) are of 20 nm, while the scale bar (c) is 10 nm.

higher states, because *hh–lh* splitting is smaller and the PL intensity of the ground-state is much higher than for the other samples. Much higher PL intensity of sample B372 could be related to a higher growth temperature of QW (307°C) due to a lower density of Ga and/or As-related defects.

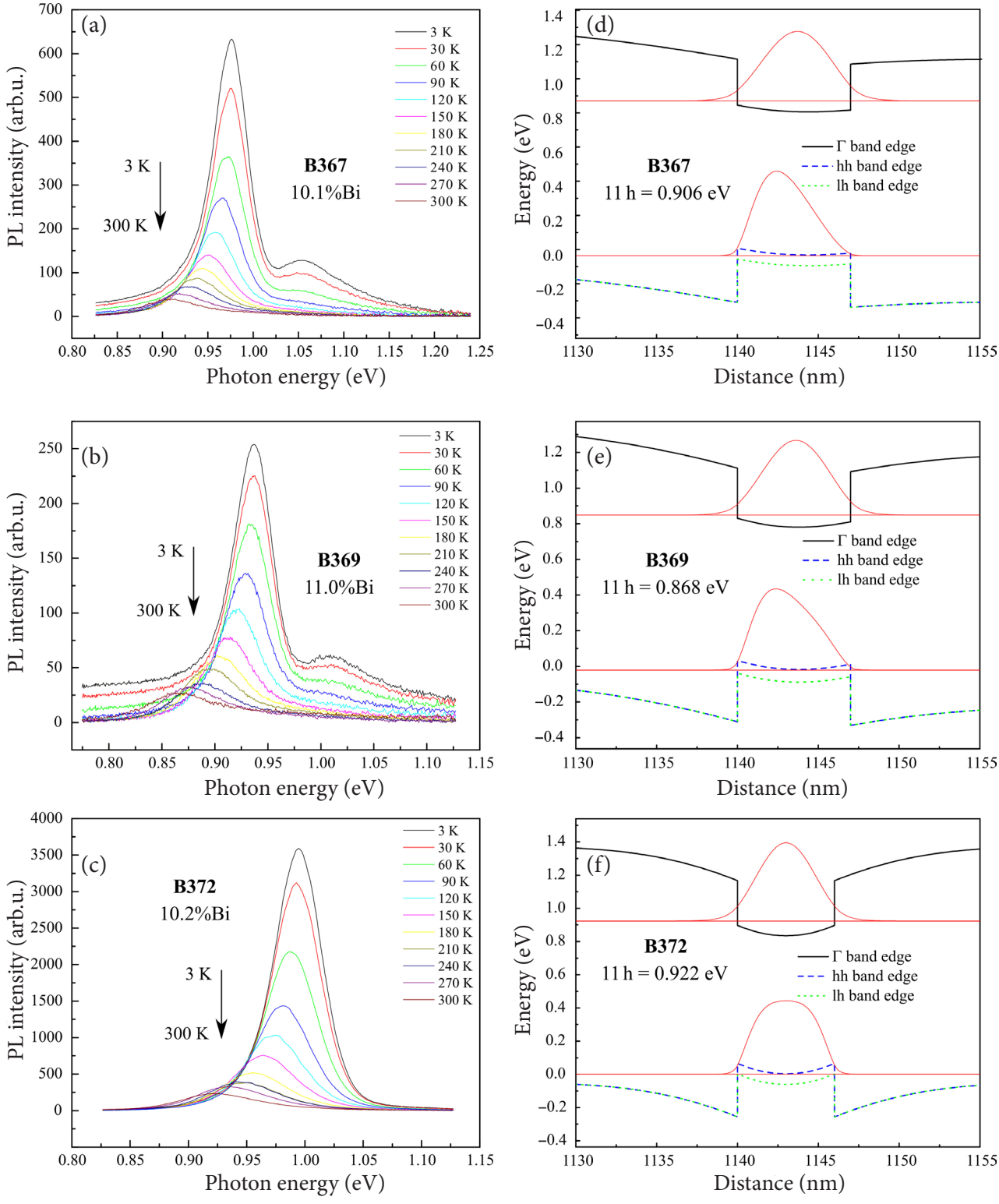

Fig. 3. (Coloured online) Temperature-dependent PL spectra of GaAsBi/GaAs:Be SQWs grown at 300°C (a, b) and 307°C (c) temperature. The band structure of $GaAs_{0.899}Bi_{0.101}$/GaAs:Be ($N_{Be} = 5 \times 10^{17}$ cm^{-3}) (a), $GaAs_{0.89}Bi_{0.11}$/GaAs:Be ($N_{Be} = 1 \times 10^{18}$ cm^{-3}) (b) and $GaAs_{0.898}Bi_{0.102}$/GaAs:Be ($N_{Be} = 3 \times 10^{18}$ cm^{-3}) SQWs simulated with the *nextnano*^{3} software [11].

It is important to note that the buffer, barrier and cap layers of the studied SQW samples were *p*-type GaAs with Be doping concentration of 5×10^{17} cm^{-3}, 1×10^{18} cm^{-3} and 3×10^{18} cm^{-3} for the samples B367, B369 and B372, respectively. The *p*-type doping of barrier layers was used in order to improve PL intensity as shown earlier [13, 14]. The influence of Be concentration on the electronic structure and PL was investigated by numerical calculations presented in Figs. 3(d–g). All three SQWs were simulated using the single-band effective-mass approximation scheme to calculate band-gaps of GaAsBi/GaAs:Be, band edges of light and heavy holes in the valence band (VB) and the conduction band (CB) edge at Γ-point, the lowest energy eigenstates and squared moduli of their eigenfunctions (Ψ^2). It can be seen in Figs. 3(d, f) that for the structures B367 and B369, with a lower Be doping concentration in barrier layers, $N_{Be} = 5 \times 10^{17}$ cm^{-3} and $N_{Be} = 1 \times 10^{18}$ cm^{-3}, respectively, the Ψ^2 of the lowest eigenstates in VB is asymmetric. It leads to a lower strength of the overlap integral of electron and heavy hole wave functions, together with a lower PL intensity. Meanwhile, for the structure B372 with the highest Be doping concentration in the barrier layer, $N_{Be} = 3 \times 10^{18}$ cm^{-3}, the overlap of electron and heavy hole wave functions increases (Fig. 3 (g)) and determines a higher PL intensity.

The PL peak position variation with temperature is shown in Fig. 4. It is seen that there is no characteristic S-shaped PL peak position dependence often observed for GaAsBi QW structures for all the three investigated samples. Therefore, the fundamental band-gap shrinkage of the semiconductors can be well-described by the empirical Varshni equation [15]

$$E_g(T) = E_0 - \frac{\alpha T^2}{\beta + T}, \quad (1)$$

where E_0 is the band-gap energy at 0 K temperature, and α and β are the Varshni fitting parameters. The best fitting parameters of PL peak position versus temperature data using the Varshni equation are listed in Table 2. The Varshni parameters of bulk GaAs are also shown in Table 2 for comparison. Note that the Varshni fitting parameters, α and β, of GaAsBi SQWs are slightly smaller than those reported for GaAs. This shows that the band-gap of GaAsBi SQW with a high content of bismuth is less sensitive to temperature than the conventional III–V compounds.

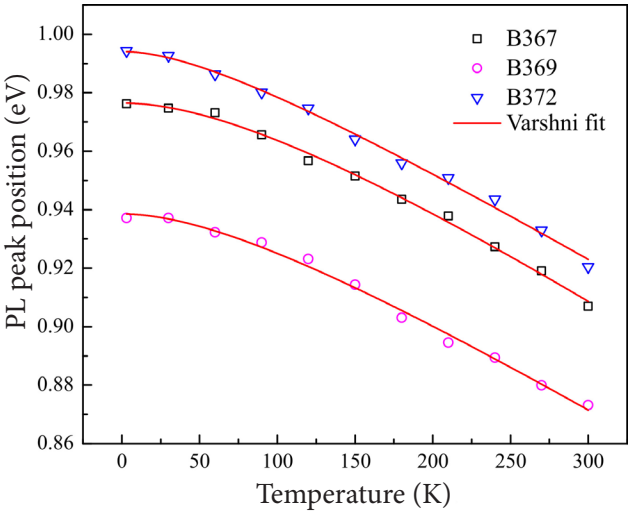

Fig. 4. Temperature dependences of the spectral position of PL bands for all the investigated structures. Symbols correspond to the experimental data, whereas the solid lines are fitted using the Varshni equation (1).

Table 2. Best fitting parameters of the PL peak position versus the temperature curve using the Varshni equation (1).

Sample No.	E_0, eV	α, meVK^{-1}	β, K
GaAs [18]	1.519	0.540	204
B367	0.976±0.001	0.367±0.045	187±59
B369	0.939±0.001	0.328±0.043	147±57
B372	0.994±0.001	0.319±0.030	104±38

The impact of temperature on the integrated PL intensity of sample B369 with the highest Bi concentration is depicted in Fig. 5. The thermal quenching of PL signal for all the samples was analysed by fitting experimental data with the Arrhenius-type expression

$$I = \frac{I_0}{1 + \alpha_1 \exp(-E_{\alpha 1} / k_B T) + \alpha_2 \exp(-E_{\alpha 2} / k_B T)}, \quad (2)$$

where I_0 is the integrated intensity at 3 K, and a_1 and a_2 are fitting parameters relating to activation energies E_{a1} and E_{a2}, respectively. Equation (2) takes into account two thermal quenching processes, which describe thermal activation of carriers in two temperature regimes. Lower activation energy $E_{a1} = 6.2 \pm 0.4$ meV (B367), $E_{a1} = 5.5 \pm 0.9$ meV (B369) and $E_{a1} = 7.3 \pm 1.0$ meV (B372) can be assigned to Bi

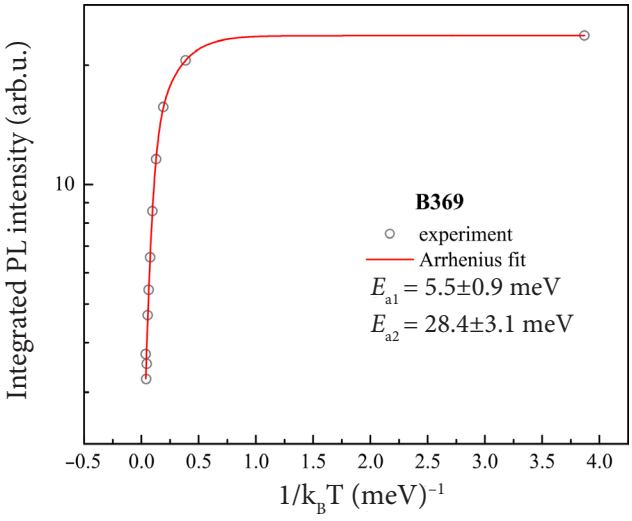

Fig. 5. Arrhenius plot of the integrated PL intensity of GaAsBi SQW with the highest Bi concentration (sample B369). The least squares method fitting was performed using the Arrhenius-type function (2). Symbols correspond to the experimental data and straight lines to the simulation.

pairs and clusters. And the higher temperature range activation energies, E_{a2} = 34.9±2.4 meV (B367), E_{a2} = 28.4±3.1 meV (B369) and E_{a2} = 34.5±4.3 meV (B372), can be ascribed to the thermal delocalization of carriers from the localized states to the mobility edge [16].

Finally, the room temperature PL spectra dependence on photoexcitation power allows one to get a deeper insight into the carrier recombination mechanisms behind PL. The integrated PL intensity dependences on excitation power in the double logarithm scale are shown in Fig. 6. The dependence is linear, and the slope was estimated by fitting the experimental data with the power law $I_{PL} \propto P^{m}_{exc}$. The best fit showed that m = 1.27±0.04, m = 1.21±0.06 and m = 1.15±0.06 for the samples B367, B369 and B372, respectively. The index m value around 1.2 indicates that radiative recombination is dominant even at room temperature [17]. That shows a high quality of QW structures. Even though, the Bi fraction in QWs is high and a low PL intensity with a broad PL linewidth (FWHM, full width at half maximum) (>100 meV) is expected for such structures. However, the investigated samples showed a high PL intensity even at room temperature and FWHM was around 80 meV due to a low interface roughness.

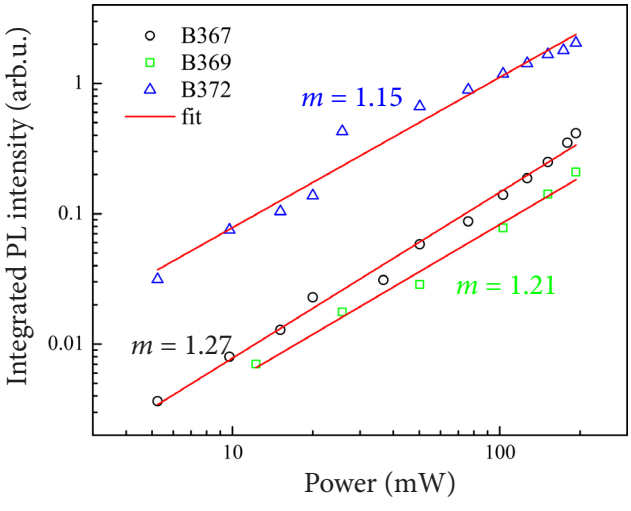

Fig. 6. Room temperature integrated PL intensity dependence on the excitation power in a double logarithm scale for GaAsBi SQW (symbols) and fitting with the power law function (lines).

4. Summary and conclusions

In conclusion, this study has confirmed that the GaAs$_{1-x}$Bi$_x$/GaAs:Be SQW structures (x ~ 0.1) grown on the GaAs substrates with the targeted NIR wavelength region are characterized by a high crystalline quality, a smooth surface and sharp interfaces between the layers and exhibited enhanced PL and lower FWHM of QW structures. The high resolution TEM measurements have evidenced that the optimized growth by carefully selecting the key parameters – growth rate, Bi flux and As to Ga ratio – even at such low temperatures allows one to achieve the epitaxial structure, which is the main criterion for a strong photoluminescence signal in the NIR spectral region. These structures can be used as a gain region for semiconductor lasers for biomedicine, environmental sciences, defense and other applications. These achievements motivate further investigations dedicated to reveal physical mechanisms for development of GaAsBi-based infrared lasers.

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GaAsBi KVANTINIŲ DUOBIŲ SU 10 % BISMUTO FOTOLIUMINESCENCINĖS SAVYBĖS

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Santrauka

Darbe pristatomas žemose temperatūrose molekulių pluoštelio epitaksijos metodu užaugintų $GaAs_{1-x}Bi_x$, kur $x \sim 0{,}1$, kvantinių duobių su *p* tipo GaAs barjerais fotoliuminescencijos (PL) tyrimas. PL matavimai buvo atlikti plačiame 3–300 K temperatūrų intervale, naudojant skirtingas sužadinimo galias. Tirtos GaAsBi kvantinės duobės pasižymi aukšta kristaline kokybe, glotniais paviršiais, aštriais perėjimais tarp sluoksnių, lėmusiais didesnį PL intensyvumą ir mažesnį PL juostų pusplotį. Temperatūriniai PL matavimai parodė, kad temperatūrinė optinių šuolių energijos priklausomybė atitinka fundamentinį Varshni dėsnį su parametrais, mažesniais nei GaAs. Krūvininkų rekombinacijos analizė atskleidė, kad spindulinės rekombinacijos kanalas dominuoja net kambario temperatūroje. Atlikti skaitmeniniai GaAsBi/GaAs:Be kvantinių duobių skaičiavimai rodo, kad esant didžiausiai Be koncentracijai barjeruose elektronų ir sunkių skylių banginių funkcijų persiklojimas didesnis, o tai lemia išaugusį PL intensyvumą.