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Novel self-assembled quantum dots in the GaSb/AlAs heterosystem

T. S. Shamirzaev[1)], *D. S. Abramkin, A. K. Gutakovskii, M. A. Putyato*

Rzhanov Institute of Semiconductor Physics, Siberian Branch of RAS, 630090 Novosibirsk, Russia

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Novel self-assembled quantum dots (QDs) in the GaSb/AlAs heterosystem were obtained and studied by means of transmission electron microscopy, steady-state and transient photoluminescence. A strong intermixing of both III and V group materials results in the fabrication of quaternary alloy QDs in the AlAs matrix. The QDs have atypical energy structure: band alignment of type I with the lowest electronic state at the indirect X minimum of the conduction band.

Self-assembled semiconductor quantum dots (QDs) have currently attracted much attention for their potential applications and a variety of new interesting physical properties [1]. Among manifold of different QDs systems, the GaSb/AlAs heterosystem is considered as a promising one for the development of memory units. Marent et al. predict that the unit based on GaSb/AlAs QDs should have a storage time of more than 10^6 years [2]. On the other hand, our recent calculation predicted that the QDs have very interesting energy structure: band alignment of type I (both electron an hole are located inside QDs) with the lowest electron states at the indirect minimum of the conduction band [3]. Such energy structure provides long exciton lifetime [4], which make the QDs promising for experimental investigation long-time exciton spin relaxation processes that impossible in direct band gap QDs (InAs/GaAs) where exciton spin is frozen during the short radiative lifetime of the exciton [5]. However, to the best of our knowledge, fabrication and experimental study of QDs self-assembled in the GaSb/AlAs system has not been performed yet.

In this letter, we for the first time demonstrate formation of self-assembled GaSb QDs in AlAs matrix. Investigation of the structure and energy spectrum of the QDs by means of transmission electron microscopy (TEM) and photoluminescence (PL) together with computational work reveal that due to strong intermixing of the Al, Ga and As, Sb sublattices such QDs consist of $Ga_xAl_{1-x}Sb_yAs_{1-y}$ alloy. It is demonstrated that depending on the alloy composition the QDs can have band alignment of type II or type I with lower electronic state belonging to the indirect X_{XY} minimum of the conduction band. The QDs fabricated in this study have band alignment of type I.

The studied self-assembled QDs were grown by molecular-beam epitaxy on semi-insulating (001)-oriented GaAs substrates. The structures have one QDs sheet sandwiched between 50-nm-thick AlAs layers grown on top of a 200-nm-thick GaAs buffer layer. Lower AlAs layer was grown at the substrate temperature of 600 °C. Then the temperature was reduced down to 450 °C and the QDs were formed. The nominal amount of deposited GaSb was equal to 1.15 monolayer. In order to reduce Sb segregation the upper AlAs layer was grown at the same temperature as the GaSb QDs. A 12-nm-thick GaAs cap layer protects AlAs against oxidation. The atomic structure of the QDs was studied by means of transmission electron microscopy employing a JEM-4000EX operated at 400 keV. Steady-state PL was excited by a He-Cd laser ($h\nu = 3.81$ eV) with power densities within the range of 0.5–25 W/cm^2. Transient PL was excited by a pulsed N_2 laser ($h\nu = 3.68$ eV) with a pulse duration of 7 ns and a peak energy density of 5 μJ/cm^2. The PL was detected with a double diffraction grating spectrometer equipped with a cooled photomultiplier operated in the photon counting mode.

Fig. 1a demonstrates a TEM plane view image of a GaSb/AlAs structure with the QDs. One can see coherently strained QDs in the image. The number and diameters of the QDs were estimated within an area of 4 mm^2. The density of the QDs is about 1.5×10^9 cm^{-2}, which is much less than typical values obtained at similar growth condition for QDs in heterosystems InAs/AlAs ($1.2 \cdot 10^{11}$ cm^{-2}) and GaSb/GaAs ($1.4 \cdot 10^{10}$ cm^{-2}) with similar lattice mismatch (7%) [6–9]. The size dispersion of the QDs is shown in Fig. 1b as a histogram. The QDs have a typical diameter of about 36 nm, which is also larger than that for QDs in the similar lattice mismatch heterosystems InAs/AlAs (5.5 nm) and GaSb/GaAs (22–28 nm) [6–8].

The low density and relatively large size of the GaSb/AlAs QDs suggest a high value of adatoms diffusion length. As a rule, these growth conditions favour materials intermixing, therefore, the QDs can contain an

[1)] e-mail: tim@isp.nsc.ru

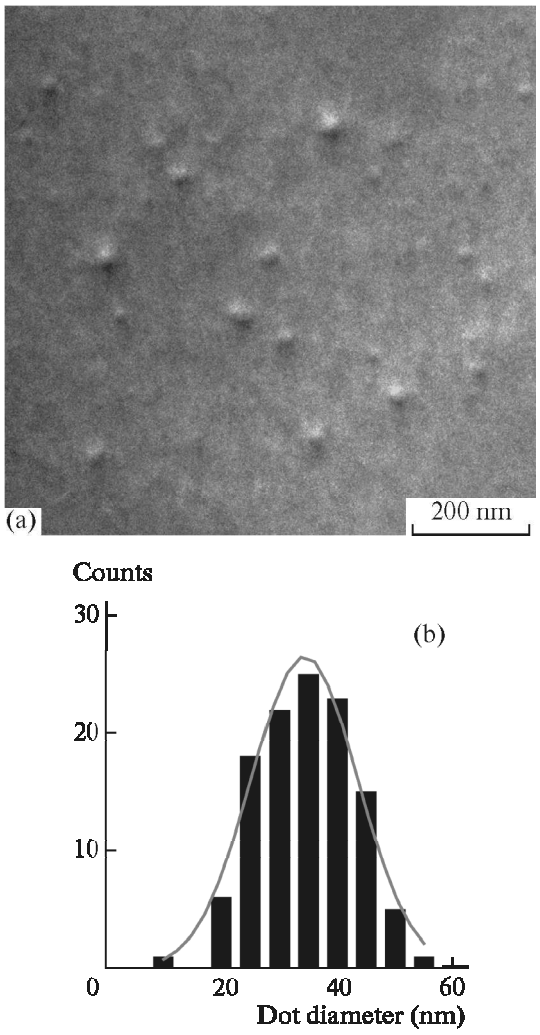

Fig. 1. (a) – TEM plane view image of the QDs self-assembled in GaSb/AlAs heterosystem. (b) – The size distribution of the QDs

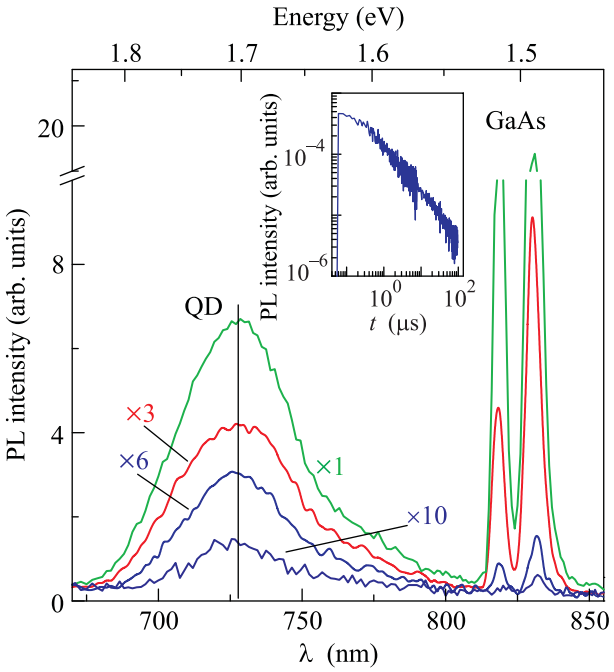

Fig. 2. Low temperature (5 K) PL spectra of the structure with QDs self-assembled in GaSb/AlAs heterosystem as a function of excitation power density, from top to bottom, W/cm^2: 25, 9, 3, 0.5. The insert shows transient PL measured at the maximum of the QD band

alloy instead of pure GaSb. PL data confirm intermixing. The PL spectra of the structure with the QDs are shown in Fig. 2. The spectra contain a band connected with recombination in GaAs buffer, marked in the figure as GaAs, and additional wide band related to exciton recombination in the QDs, marked in the figure as QD. The QD band has the energy of about 1.65–1.7 eV, while calculation predicts 0.7 eV for electron-hole optical transition energy in QDs consisting of pure GaSb in AlAs matrix [3].

Band structure parameters of quaternary alloy $Ga_xAl_{1-x}Sb_yAs_{1-y}$ are not defined. Using the band structure parameters of GaSb, AlSb, GaAs, and AlAs taken from Ref. [10] we use a linear approximation between the parameters GaSb, GaAs (AlSb, AlAs) to estimate parameters of ternary alloy $GaAs_ySb_{1-y}$ ($AlAs_ySb_{1-y}$). The parameters of quaternary alloy $Ga_xAl_{1-x}Sb_yAs_{1-y}$ were constructed via a linear approximation between the ternary alloy parameters. Then energy structure of $Ga_xAl_{1-x}Sb_yAs_{1-y}$/AlAs heterostructures was calculated as a function of compositions x and y.

Comparison of the calculated energy of electron – heavy hole optical transition in the heterostructures with experimental data (energy position of the QD band) allows us to estimate the composition of the alloy inside the QDs. The best agreement between calculation and experimental data was obtained for alloy composition (x, y) values shown in Fig. 3. One can see that in contrast to GaSb/GaAs QDs, which definitely have band alignment of type II [7], QDs self-assembled in the heterosystem of GaSb/AlSb can have band alignment of type II or type I with the lowest conduction band states belonging to the indirect X minimum of the conduction band (see the insert to Fig. 3) depending on composition. The boundary between the composition related to band alignment of type II and type I is shown in the figure by a dash-dot line.

A method for the determination of the band lineup of heterostructures based on the PL technique has been proposed by Ledentsov et al. [11]. They theoretically

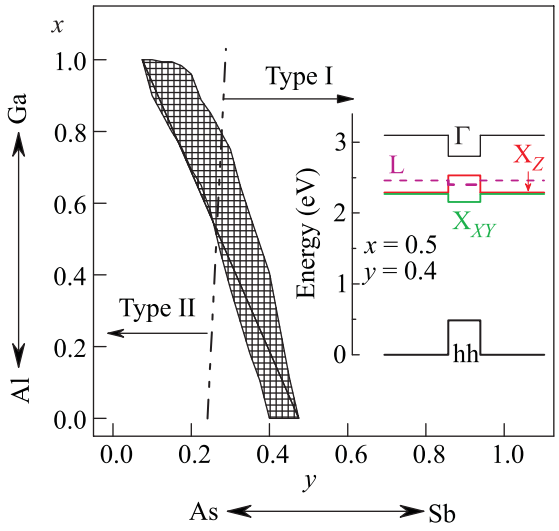

Fig. 3. Filled area marks compositions (x, y) corresponding to electron-hole optical transition in $Ga_xAl_{1-x}Sb_yAs_{1-y}$/AlAs QDs lying in the range of 1.65–1.70 eV. The insert presents energy spectrum calculated for $Ga_{0.5}Al_{0.5}Sb_{0.4}As_{0.6}$/AlAs QDs

calculated and experimentally demonstrated that the energy position of the PL band should blue-shift proportionally with the cube root of the excitation power density (P) for any structures with type II alignment. The blue shift is characteristic to all type-II heterostructures (both quantum wells (QWs) and QDs) and reflects the dipole layer formation caused by a spatial separation of non-equilibrium holes confined in the QW or QD and electrons confined in the nearby matrix region. This PL technique was successfully used for many type II systems such as GaSb/GaAs QWs and QDs [7, 11], and GaAs/AlAs QWs [12].

In order to check the energy structure of the QDs, we measured transient PL and steady-state PL spectra as a function of excitation power density. The indirect structure of the QD conduction band gap is evidenced by long PL decay time [13], which lies in microsecond range as one can see in insert to Fig. 2. The QD PL band does not demonstrate any blue shift with an increase in the excitation power density within the range of 0.5–25 W/cm^2 (see. Fig. 2), thus we can conclude that the QDs have band alignment of type I.

Note that calculation does not describe the experimental data (the composition of the $Ga_xAl_{1-x}Sb_yAs_{1-y}$ alloy) at $y = 1$ with any x value. Therefore the QDs in any case contain arsenic. One can also see in Fig. 3 that the band alignment of type I can be realized only for $x \leq 0.8$. Thus segregation of Sb during the growth process stimulates intermixing not only for V group As and Sb materials, which is typical for GaSb/GaAs structures [9, 14], but also for the III group materials Ga and Al, similar to the recently observed intermixing of Ga and In at interface of InAs/GaSb superlattice [15].

In conclusion, self-assembled QDs have been formed for the first time in the GaSb/AlAs heterosystem. It was demonstrated that strong intermixing of both group III and V elements occurs during the QDs formation. As a result, the QDs contained $Ga_xAl_{1-x}Sb_yAs_{1-y}$ quaternary alloy. Type I band alignment with electronic state belonging to the indirect X minimum of the conduction band was shown for the QDs. The structure is promising object to investigation of spin relaxation of exciton located in QD.

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