Comparative Analysis of the Optical and Physical Properties of InAs and In$_{0.8}$Ga$_{0.2}$As Quantum Dots and Solar Cells Based on them

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Abstract—InAs and In$_{0.8}$Ga$_{0.2}$As quantum dots in a GaAs matrix as well as GaAs solar cells with quantum dots of both types in the $i$-region are obtained by metalorganic vapor-phase epitaxy. As a result of investigations by photoluminescence and transmission electron microscopy, it is found that the In$_{0.8}$Ga$_{0.2}$As quantum-dot array is highly uniform, contains a smaller number of large imperfect quantum dots, and also provides a decrease in mechanical stresses in the structure. An analysis of the spectral dependences of the internal quantum yield shows that the quality of a solar-cell matrix after embedding up to 20 rows of In$_{0.8}$Ga$_{0.2}$As quantum dots remains at a level close to the reference GaAs solar cells. In this case, a linear increase in the additional photocurrent generated due to the absorption of sub-bandgap photons in In$_{0.8}$Ga$_{0.2}$As quantum dots is provided with an increase in the number of rows of quantum dots, since the value of the photocurrent gain per row is preserved.

Keywords: photocurrent, quantum dots, solar cells
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1. INTRODUCTION

Heterostructures with space-charge limitation in all three dimensions such as quantum dots (QDs) have become one of the most important objects of investigation in the physics of low-dimensional semiconductor structures [1]. Currently, it is InAs/GaAs that remains the most studied material system for the formation of QDs. A sufficiently large difference in the crystal-lattice parameters between these materials (~6.6%) makes it possible to form QDs in the Stranski–Krastanov mode [2]. In this mode, the appearance of QDs is preceded by the layer-by-layer growth and formation of a thin wetting layer. In this case, the selection and control of the nucleation parameters of QDs make it possible to obtain dislocation-free coherently strained islands of a high density with a narrow size distribution [3]. Due to their interesting optical properties, the QDs have found application in several fields of semiconductor industry. For example, InAs/GaAs QDs made it possible to create lasers with a record low threshold current density [4], high temperature stability, and higher gain [5]. Telecommunication lasers emitting at a wavelength of 1.3–1.55 µm were also produced on the basis of such QDs [6, 7]. Over the past two decades, the approaches were actively developed, which allow one to use systems with QDs for improving the photoelectric characteristics of modern semiconductor solar cells (SCs). The idea of such SCs consists in forming an intermediate band inside the device matrix for absorbing low-energy photons in order to increase the short-circuit current [8–11]. Despite the large number of theoretical studies in this direction, it was impossible to fully implement such an approach in practice. However, a number of studies showed an increase in the spectral sensitivity of single-junction GaAs SCs with embedded InAs QD arrays [12–14].

The carrier-generation effect in QD arrays due to the absorption of subgap photons can be used to increase the photogenerated current of the middle Ga(In)As subcell in the structure of multi-junction SCs, in particular, based on InGaP/Ga(In)As/Ge materials. In the future, it can solve the existing problem of the mismatch of the currents of subcells in a multi-junction SC [15, 16], which enables one to raise the overall photocurrent of such a structure and increase the efficiency. However, in SCs with embedded arrays of InAs QDs, there are a number of problems, which prohibits achieving a high efficiency. The main problem is the reduction in the operating voltage. In publications, it is customary to consider a decrease in the open-circuit voltage ($V_{oc}$) of the SCs [17]. The drop in $V_{oc}$ is caused by the introduction of additional recombination levels into the band gap of
decrease and, correspondingly, the decrease in recombi-
nation current through such levels should drop. In this study, In$_{1-x}$Ga$_x$As QDs increases as compared with the InAs QDs in the sublattice of group-III atoms), this difference can be partially solved by using the InGaAs solid solution. The difference in the crystal-lattice parameters between InAs and GaAs is 6.6%. However, when a certain amount of Ga is introduced into the QD composition (up to 20% in the sublattice of group-III atoms), this difference decreases by ~1%. Using multiple stacking, such a mismatch leads to a less stressed QD array. In addition, the electron-transition energy in the InGaAs QDs increases as compared with the InAs QDs in GaAs, which should lead to a decrease in the recombina-
tion current of saturation and, consequently, to a
reduction in the losses associated with the voltage drop. In this study, In$_{1-x}$Ga$_x$In$_y$As/GaAs QDs with $x \approx 0.8$ are proposed as an alternative to classical InAs/GaAs QDs and the growth technologies of SCs with QDs, which implies the incorporation of layers that compensate for stresses in the structure. The results of investigation and comparative analysis of the optical and physical properties of InAs and In$_{1.8}$Ga$_{0.2}$As QDs as well as the photoelectric characteristics of SCs based on them are presented.

2. EXPERIMENTAL

The experimental structures were grown by the method of metal-organic vapor-phase epitaxy (MOVPE) in an installation with a horizontal-type reactor. Organometallic compounds were used as sources of group-III elements: trimethylgallium (TMGa), trimethylaluminum (TMAI), and trimethylindium (TMIn). Arsine (AsH$_3$) was used as the source of the group-V element arsenic. All structures were grown on vicinal GaAs(100) substrates misorient-
ed by 6° to the [111] direction, which made it possible to grow the SC structures at high rates. For investigat-
ing single QD rows by photoluminescence (PL), heterostructures were grown, the schematic representa-
tion of which is shown in Fig. 1a. The structure was grown in the following sequence: at a temperature of 700°C, a wide-gap Al$_{0.3}$Ga$_{0.7}$As barrier 250 nm thick and a part of a GaAs waveguide 250 nm thick were grown; then, the reactor was cooled to a temperature of 525°C after which the QD material was deposited, and coherent islands were deposited with the formation of a wetting layer in the Stranski–Krastanov mode; Further, growth was interrupted in order to provide the final formation of the QDs [24] after which the QDs were overgrown with a 5-nm-thick GaAs coating layer at the same temperature to protect the QDs from decomposition upon further heating of the reactor; then, the installation reactor was heated to 700°C, and a GaAs waveguide with a thickness of 250 nm, and a 50-nm-wide Al$_{0.3}$Ga$_{0.7}$As barrier was grown.

For both types of QDs in the heterostructures described, the amount of deposited material varied from 1 to 4 mono layers (MLs). The deposition rate of the QD material was 0.062 ML/s for InAs [12] and 0.167 ML/s for In$_{0.8}$Ga$_{0.2}$As [25]. The growth rate of the GaAs waveguide and the Al$_{0.3}$Ga$_{0.7}$As barriers as well as all high-temperature layers amounted to 1.5 μm/h. In this case, the growth rate of the GaAs coating layer was reduced to 0.75 μm/h in order to improve its quality at a low growth temperature and, thereby, preserve the optical properties of the QDs [12].

We also grew SC structures including GaAs SCs without QDs (Fig. 1b) and two series of GaAs SC structures with InAs and In$_{0.8}$Ga$_{0.2}$As QD arrays embedded into the $i$-GaAs region (Fig. 1c). Several QD rows were grown through 35-nm-thick intermediate GaAs layers. Both series of SCs with QDs included...
structures with 10, 15, and 20 layers of QDs in the array (in Fig. 1c, \( N \) is the number of QD rows). All the basic layers of the SC structure were grown at 700°C, the QDs and the GaAs cap layer at 520°C, and the intermediate layers at 600°C.

To obtain the PL spectra from the experimental heterostructures containing QDs, we used an Nd:YAG laser with a radiation wavelength of \( \lambda = 532 \) nm and a power of up to 350 mW as a radiation source. The radiation from the sample was focused on the entrance slit of an MDR-23 monochromator using a collecting lens. All measurements were carried out by a cooled Ge-detector of optical radiation according to the standard method of synchronous detection.

For measuring the quantum yield of the SC with embedded QD arrays, we used an installation, which included an ultraviolet halogen lamp, a grating monochromator with a dispersion of 2 nm/mm in the scanned wavelength range from 300 to 1200 nm, an optical chopper with a frequency of 90 Hz, and a highly sensitive synchronous-detection system. In the framework of this experiment, the synchronous-detection technique made it possible to detect the weak output signal reasonably accurately in the QD absorption region and to significantly suppress noise.

The images of QD rows in the cross section and in the plan-view were obtained by transmission electron microscopy (TEM) using a JEOL JEM 2100F microscope at an accelerating voltage of 200 kV. The samples were prepared by preliminary mechanical grinding followed by final treatment with an \( \text{Ar}^+ \)-ion beam with an energy of \( \sim 3.5 \) keV.

3. RESULTS AND DISCUSSION

3.1. Critical Thickness of InAs and InGaAs for Forming Quantum Dots

In the Stranski–Krastanov mode, the onset of QD formation occurs when a certain critical thickness \( \theta_c \) of the nucleation material is achieved at which two-dimensional layer-by-layer growth passes into three-dimensional island growth. The value of \( \theta_c \) depends on the difference in the lattice parameters of the QD and matrix material. When QDs are grown by molecular-beam epitaxy, the \( \theta_c \) value can be determined using in-situ measurements of fast-electron diffraction [26, 27]. For MOVPE, this technique is inaccessible, and it is the ex-situ analysis of the PL spectra from heterostructures with embedded QD arrays that is usually used for establishing the optimal technological conditions for the growth of QDs.

The PL spectra of the series of InAs/GaAs heterostructures were measured with various amounts of InAs material. For the PL spectrum of each heterostructure with QDs, we calculated the relative integrated quantum efficiency (\( \eta_{\text{eff}} \)), which was normalized to the value of the PL intensity highest among the entire series of the samples. The value of \( \theta_c \) for the formation of QDs corresponded to the peak of the dependence \( \eta_{\text{eff}} \) on the material amount \( H \), which was determined for InAs QDs at \( \sim 1.7 \) ML (Fig. 2, curve 1). It correlates well with the values obtained from direct measurements of fast-electron diffraction in molecular-beam epitaxy, where it was shown that the \( \theta_c \) values for the InAs/GaAs quantum dots are in the range from 1.5 to 1.7 ML depending on the technological parameters used [26–29]. Indeed, for the amount <\( \theta_c \) of deposited material, narrow PL spectra with a low
expected, the In0.8Ga0.2As QDs have a thicker wetting
layer. This effect was really observed with an increase to
1270

nt 

f 

for In0.8Ga0.2As QDs (Fig. 3d). In this case, as
shown near individual islands, which indicates their
 structural perfection. We estimated the average QD
height, which was 4 nm for InAs QDs (Fig. 3c), and
observed in the parameters of the crystal lattices of GaAs
and In 0.8Ga0.2As (Fig. 3b), no dislocations are
formed, and the parameter C

C

is achieved, the broadened PL spectrum characteristic of QDs is observed. With an increase in the material amount >\(\theta_C\), the intensity of the spectra begins decreasing due to the coalescence of islands.

In the same way, we determined the parameter \(\theta_C\) for the In0.8Ga0.2As QDs, whose value amounted to ~2.0 ML (Fig. 2, circles). Due to the smaller difference in the parameters of the crystal lattices of GaAs and In0.8Ga0.2As and, as a result, a lower mechanical stress, more material is required to begin the formation of islands. As a result, the thickness range increases at which QDs with a high integrated intensity are formed, and the \(\eta(H)\) dependence for In0.8Ga0.2As QDs has a large half-width.

### 3.2. Physical Parameters of InAs and InGaAs Quantum Dots

To investigate the physical parameters of the QDs, TEM measurements were performed on the grown heterostructures. Both in the case of InAs (Fig. 3a) and In0.8Ga0.2As (Fig. 3b), no dislocations are observed near individual islands, which indicates their structural perfection. We estimated the average QD height, which was 4 nm for InAs QDs (Fig. 3c), and 5 nm for In0.8Ga0.2As QDs (Fig. 3d). In this case, as expected, the In0.8Ga0.2As QDs have a thicker wetting layer.

The TEM images of the In0.8Ga0.2As-QD layer in the frontal configuration (Fig. 3e) made it possible to carry out a statistical estimate of the QD concentration, which was \(\sim 9.7 \times 10^{10} \text{ cm}^{-2}\). In comparison with studies in which high-density layers of InAs QDs were obtained [30], an increase in the average lateral size of the InGaAs QDs is observed due to the appearance of relatively large QDs. In the TEM images in the plan-view, large QDs were mainly detected in the form of crater-shaped objects (indicated by arrows in Fig. 3e), the formation of which can also be associated with the presence of a small concentration of defect clusters. The height of such objects can significantly exceed the thickness of the GaAs cap layer, which leads to their incomplete overgrowth. During heating of the reactor before deposition of the next GaAs layer, such objects dissolve [31]. The edges of the formed hollows after GaAs overgrowth generate stress fields, which can be clearly seen in the TEM image.

Using the TEM images, the lateral sizes of the In0.8Ga0.2As QDs were estimated, the statistics for which are shown in Fig. 3f. It should be noted that the percentage of QDs with dimensions exceeding 24 nm, which were statistically taken into account, includes mostly the crater-shaped objects described above. The lateral sizes of defect-free In0.8Ga0.2As QDs are in the range from 14 to 24 nm, which correlates with the data published in publications for objects of this type [32].

### 3.3. Optical Properties of InAs and InGaAs Quantum Dots

To determine the position of the PL peaks, their asymmetric shape was approximated by two Gaussian curves (curves in Figs. 4a and 4b) reflecting the contributions of QD families denoted as QD1 and QD2. As a result of approximation, the experimental temperature dependences of the position of the PL peaks were plotted for both families of InAs and In0.8Ga0.2As QDs (Figs. 4c and 4d, points). From the obtained dependences, a significant long-wavelength shift of the PL peaks with increasing temperature is seen, which was previously noted in publications for both InAs/GaAs QDs [33, 34] and In0.8Ga0.2As QDs [35]. Using the Varshni formula [36, 37], the data of the dependences were approximated (Figs. 4c and 4d, lines).

At low temperatures (\(T < 150°C\)), both families for both types of quantum dots under investigation participate in radiative recombination, but the carrier transport between them is largely suppressed due to the low temperature. Thus, the recombination channels in which the density of states is higher dominate in intensity. In the case of InAs QDs, the PL peak from the QD1 family (QDs with small sizes) dominates in intensity over QD2 (the QD family with relatively large sizes). It can be concluded that the population of relatively small QDs is dominant. In the case of In0.8Ga0.2As QDs, the intensity parity between the QD1 and QD2 peaks is maintained, which indicates a more uniform size distribution of such QDs. In this case, the intense peak from the wetting layer of the In0.8Ga0.2As QDs indicates the fact that it acts as an independent effective radiative-recombination channel. This effect was really observed with an increase to

\[\text{Fig. 2. Dependence of the relative integrated quantum PL yield (\(\eta_{\text{int}}\)) on the amount of material (H) for (1) InAs and (2) In0.8Ga0.2As QDs.}\]
50% in the indium content in the QDs [38]. Since most of the defects are frozen out at low temperatures, the experimental temperature dependences of the peak positions in this region correlate well with the calculation according to Varshni law for bulk material for both types of QDs (Figs. 4c and 4d). In the temperature range from 150 to 250°C, parity in the intensity of the PL peaks of QD1 and QD2 is achieved for both InAs QDs and In0.8Ga0.2As QDs. In both cases, the mechanisms of thermal ejection from small QDs to large ones are activated, which entails a redistribution of the centers of radiative recombination between the QD1 and QD2 families. The radiative recombination through large QDs (QD2 family) becomes energetically more advantageous. We described such a model of carrier transport in QDs depending on the temperature in sufficient detail previously in [39]. In the case of InAs QDs, there is a general deviation of the dependence of the positions of the QD1 and QD2 peaks from Varshni law for a bulk material (Fig. 4c). For QD1, red-shift acceleration is observed, the nature of which has the following explanation. If at low temperatures radiative recombination occurs only through the dominant population of small QDs, then, the carriers are thermally ejected from them and captured by QDs with relatively large sizes with increasing temperature. The slowdown of the red shift for QD2 indicates the thermal activation of defect QDs of a very large size (defect clusters), the defrosting of defects, and the triggering of nonradiative-recombination processes. The PL peaks behave in a similar way in the case of In0.8Ga0.2As QDs (Fig. 4d). However, in the case of QD2, it is possible to observe complete adherence to Varshni law, which indicates the high homogeneity of such QDs in size and the relatively low concentration of large defect QDs, which
were observed in the TEM image (Fig. 3f). This fact indicates the higher quality of the formed In$_{0.8}$Ga$_{0.2}$As islands. At temperatures of $>$250°C for InAs QDs, the same tendency for the PL peaks to deviate from Varshni law remains. For the In$_{0.8}$Ga$_{0.2}$As QDs, the position of the QD1 peak returns to the approximation curve according to Varshni law and completely corresponds to this law in the case of QD2. Since the population of relatively large islands is predominant (Fig. 3f), we observe the dominance of the QD2 peak, which is consistent with the Fermi–Dirac distribution of the density of states. On the whole, the results of analysis of the PL spectra enable us to conclude that the use of InGaAs QDs with a low Ga content instead of classical InAs QDs is promising for their stacking in SCs with QDs.

3.4. Solar Cells Based on InAs and InGaAs Quantum Dots

Solar cells based on the grown structures (Fig. 1c) were fabricated by forming the back and front metal contacts with subsequent etching out of the $p^+$-GaAs contact layer from the photoactive surface of the sample. For these experimental SC structures, the spectral characteristics of the internal quantum yield were measured (Fig. 5).

In SCs with 10 embedded rows of InAs QDs, the appearance of the spectral sensitivity beyond the GaAs absorption edge (Fig. 5a) and the total increase in the photocurrent due to the absorption of sub-band-gap photons are observed. However, the stacking of 15 and 20 rows of QDs in the array leads to a noticeable decrease in the internal quantum yield ($\eta_{\text{int}}$) in the GaAs absorption region, which is explained by the increasing effect of mechanical stresses and a decrease in the diffusion length of minority charge carriers.

For SCs with In$_{0.8}$Ga$_{0.2}$As QDs, on the contrary, the spectral sensitivity in the range up to 880 nm remains at the sensitivity level of the reference sample even when 15 rows of QDs are stacked (Fig. 5b). A small decrease in $\eta_{\text{int}}$ comparable in magnitude to that for SCs with 10 and 15 rows of InAs QDs is observed.
when stacking up to 20 rows of In$_{0.8}$Ga$_{0.2}$As QDs, but it cannot be called dramatic. This is consistent with the PL-measurement data, which showed a higher quality of In$_{0.8}$Ga$_{0.2}$As QDs.

Using the spectral dependences $\eta_{\text{int}}$, we calculated the photocurrent (spectrum AM0) generated by the absorption of photons in GaAs and in QD arrays (see Table 1). In this case, the total photocurrent of the reference GaAs SC was 35.23 mA/cm$^2$. Despite the fact that InAs QDs make a greater contribution to the photogenerated current beyond the GaAs absorption edge than In$_{0.8}$Ga$_{0.2}$As QDs when embedding up to 15 rows of QDs, SCs with In$_{0.8}$Ga$_{0.2}$As QDs have a larger quantum yield with an increase in the number of QD rows to 20 due to preservation of the quality of the $p$--$n$ junction in GaAs at the level of the reference GaAs SC. As a result, with an increase in the number of InAs QD rows to 20, the photogenerated current of the $p$--$n$ junction in GaAs as well as the total photocurrent of the structure (Fig. 6a, curve 1) decrease even as compared to the reference SC (the general drop in $J_g$ in the GaAs absorption region upon the embedding of 20 rows of InAs QDs amounted to 1.73 mA/cm$^2$). This fact is explained by an increase in the number of defects introduced by the more stressed InAs/GaAs system and by the effect of the tunnel–trap mechanism of carrier transport at a low current density at which the spectral characteristics of the SC are measured. At the same time, the value of the total photocurrent of the SC with the In$_{0.8}$Ga$_{0.2}$As quantum dot is always higher than that of the reference SC (the largest increase in the photocurrent was 1.27 mA/cm$^2$). This value remains almost unchanged with increasing number of QD rows (Fig. 6a, curve 2), since a small

![Fig. 5. Spectral characteristics of the internal quantum yield of SCs with (a, 2–4) InAs QDs and (b, 2'–4') In$_{0.8}$Ga$_{0.2}$As QDs, as well as the spectral characteristics in the absorption region of (c, 2–4 QDs) InAs QDs and (d, 2'–4') In$_{0.8}$Ga$_{0.2}$As QDs: SCs with 10 (2, 2'), 15 (3, 3'), and 20 (4, 4') QD rows. I is the reference SC.](image-url)
decrease in the photosensitivity in the GaAs absorption region is compensated by an increase in the photocurrent generated in In$_{0.8}$Ga$_{0.2}$As QDs. Indeed, although the contribution of QDs to the SC photocurrent increases with the number of QD rows in both cases; for In$_{0.8}$Ga$_{0.2}$As QDs, the growth rate in $J_g$ is much higher (Fig. 6a, curves $1'$ and $2'$). From calculation of the photocurrent generated by one row of QDs (Fig. 6b), it follows that if the photocurrent generated by one row of InAs QDs sharply decreases with increasing number of rows to 15, the contribution of one row of In$_{0.8}$Ga$_{0.2}$As QDs remains almost unchanged (the values amount to $0.045-0.048$ mA/cm$^2$).

4. CONCLUSIONS

The physical and optical properties of the InAs and In$_{0.8}$Ga$_{0.2}$As QDs obtained by the MOVPE technique were studied. On the basis of measurements of the PL spectra and TEM investigations, it was confirmed that both types of QDs have a bimodal size distribution, but the In$_{0.8}$Ga$_{0.2}$As QDs are somewhat more homogeneous and less imperfect than the InAs QDs.

GaAs-SC structures with embedded InAs and In$_{0.8}$Ga$_{0.2}$As QDs were grown and their photoelectric characteristics were investigated. It was shown that the use of In$_{0.8}$Ga$_{0.2}$As QDs instead of classical InAs QDs can also provide a linear increase in the photocurrent generated in the QD array with an increase in the number of QD rows up to 20 instead of only maintaining the quality of the SC matrix at the level of the reference sample. The constant values of the photocurrent generated by one row of In$_{0.8}$Ga$_{0.2}$As QDs (0.05 mA/cm$^2$) obtained in this case enable us to state that this QD system is promising for embedding in QD SC without using the growth technology of compensating layers.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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