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NEAR-INFRARED PHOTOLUMINESCENCE IN *n*-GaAs/AlGaAs QUANTUM WELLS WITH DIFFERENT LOCATIONS OF COMPENSATING ACCEPTOR IMPURITY

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Abstract. In the paper, comparative studies of near-IR photoluminescence (PL) in structures with GaAs/AlGaAs quantum wells possessing different selective doping profiles have been performed. The PL spectra recorded at 5 K for different intensities of interband optical pumping were analyzed and main channels of radiative recombination were determined. The dependences of the main PL line intensities on the pump level were obtained. The results of the studies performed suggest that *n*-GaAs/AlGaAs nanostructures with the compensating acceptor impurity located not in the *n*-GaAs quantum well, but in its barriers, are preferable for terahertz radiation generation.

Keywords: quantum well, GaAs, AlGaAs, photoluminescence, optical pumping, IR radiation, terahertz radiation

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ФОТОЛЮМИНЕСЦЕНЦИЯ БЛИЖНЕГО ИК-ДИАПАЗОНА В КВАНТОВЫХ ЯМАХ *n*-GaAs/AlGaAs С РАЗЛИЧНЫМ ПОЛОЖЕНИЕМ КОМПЕНСИРУЮЩЕЙ АКЦЕПТОРНОЙ ПРИМЕСИ

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Аннотация. В работе проведены сравнительные исследования фотолюминесценции (ФЛ) ближнего ИК-диапазона в структурах с квантовыми ямами *n*-GaAs/AlGaAs с различными профилями селективного легирования. Проанализированы спектры ФЛ, зарегистрированные при температуре 5 К для различной интенсивности межзонной оптической накачки, определены основные каналы излучательной рекомбинации. Получены зависимости интенсивности основных линий ФЛ от уровня накачки. Результаты проведенных исследования позволяют утверждать, что с точки зрения эффективности использования наноструктур GaAs/AlGaAs для генерации терагерцового излучения на примесных переходах, наиболее эффективны структуры с особым профилем легирования, когда компенсирующая акцепторная примесь располагается не в квантовой яме *n*-GaAs, а в формирующих ее барьерах.

Ключевые слова: квантовая яма, GaAs, AlGaAs, фотолюминесценция, оптическая накачка, терагерцовое и инфракрасное излучение

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Introduction

Optical transitions associated with impurity states in quantum wells (QWs) have been the subject of much attention in recent years. This subject has gained prominence as sources of terahertz (THz) radiation have been developed, operating with optical or electrical pumping of nonequilibrium charge carriers, based on electron and hole transitions involving impurity states. The use of selectively doped quantum wells instead of uniformly doped bulk single crystals provides a convenient opportunity to control the energy spectrum of impurity states by changing the QW parameters, which makes it possible to change the operating frequency of a THz radiation source based on impurity transitions of charge carriers. Computations for a given semiconductor pair (QW/barrier) indicate that the frequency of THz radiation can be controlled by varying the width of the QW, as well as its doping profile [1, 2].

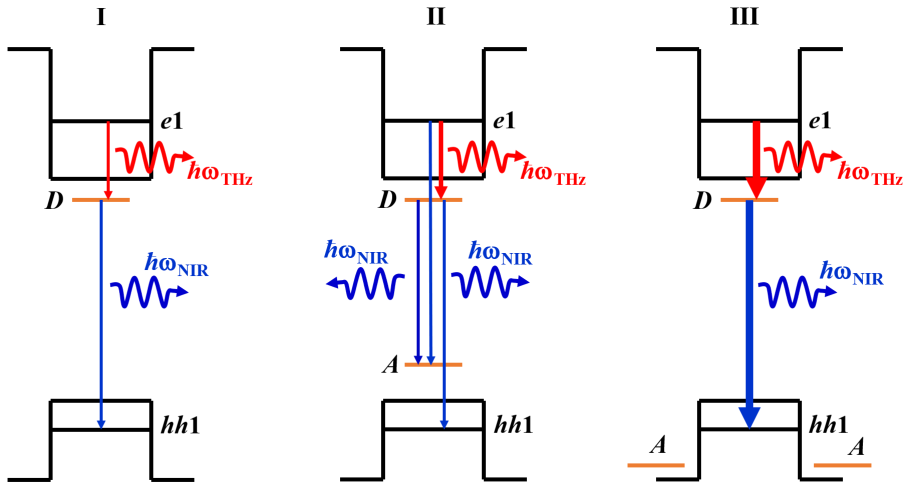

Fig. 1. Diagrams of radiative optical transitions for structures of different types: I, obtained by selective doping by donors (*D*) in the QW; II, donors compensated by acceptors (*A*) directly in the QW; III, donors in the QW compensated by acceptor doping of barriers forming the QW

The figure only shows the ground impurity states *D* and *A*, as well as the first levels of size quantization in the conduction band (*e*1) and in the heavy-hole subband (*hh*1). THz, NIR correspond to radiation in THz and near-IR ranges; photoluminescence lines for free and bound excitons are not shown

The simplest nanostructures where photoluminescence (PL) in the near-IR and THz ranges is simultaneously observed are QWs selectively doped with donors (structure I in Fig. 1). Nonequilibrium electrons are generated in the conduction band of such a structure under interband optical pumping, while holes are generated in the valence band; the electrons and the holes are then trapped in the QWs and gradually descend to the bottom of the size quantization subbands *e*1 and *hh*1, respectively, during thermalization. A nonequilibrium hole can recombine with an electron localized at the donor center at low temperatures (this corresponds to the $D \to hh1$ electron transition), with the donor consequently becoming ionized. The ionized donor can then trap a nonequilibrium electron from the conduction band (the $e1 \to D$ transition), which is accompanied by the emission of THz radiation.

Similar studies were carried out in GaAs/AlGaAs QWs using silicon as a donor impurity [3, 4]. It was established in later studies [5] that if donors in such a structure (structure I in Fig. 1) are compensated by acceptors (beryllium) directly in the QWs (structure II in Fig. 1), then an increase by about an order of magnitude can be achieved at low temperatures in the integral intensity of THz radiation due to impurity electron transitions. The possible reasons for this may lie in a decrease in the equilibrium occupancy of the donor states involved in the $e1 \to D$ transition, as well as in an additional increase in the depletion rate of the ground state of donor *D*, which is the final state for this transition, due to the presence of donor-acceptor recombination ($D \to A$ transitions) in a type II structure.

We have developed a new class of structures with selectively doped QWs, aimed at additionally improving the generation efficiency of THz radiation under interband optical pumping (structure III in Fig. 1). The compensating acceptor impurity in this structure is located in the barriers forming the QWs rather than in the QWs themselves. Unlike the type II structure, the type III structure does not include an undesirable channel of radiative recombination due to $e1 \to A$ electron transitions reducing the concentration of nonequilibrium electrons in the *e*1 electron subband, which is the ground state for radiative transitions of the THz range ($e1 \to D$). This should provide a gain in the generation efficiency of THz radiation under interband optical pumping.

An additional gain is also possible because the negative charge of acceptors occupied by electrons in the type III structure is localized outside the QWs, and therefore their electric field does not prevent the trapping of nonequilibrium electrons to donor levels in the QWs, in particular, during $e1 \to D$ radiative transitions.

The goal of the work was to compare near-IR photoluminescence in type III and type II structures, analyzing the PL spectra measured at liquid helium temperature for different intensities of interband optical pumping, to subsequently determine the main channels of radiative recombination.

Characteristics of the samples

Heterostructures with multiple selectively doped GaAs/AlGaAs QWs were grown on epi-ready semi-insulating GaAs (001) substrates with an undoped GaAs buffer layer about 200 nm thick at substrate temperature $T_s \approx 580$ °C by molecular beam epitaxy (MBE) using a two-chamber setup from SemiTEq (Russia). Standard Ga and Al effusion cells and an As valve cracking cell were used as sources of molecular beams. Si and Be were used as *n*- and *p*- type doping impurities, respectively.

Two heterostructures were grown by MBE: they consisted of two (top and bottom) $Al_{0.9}Ga_{0.1}As$ barrier layers each 30 nm thick, 50 GaAs quantum wells 7.6 nm thick separated by $Al_{0.3}Ga_{0.7}As$ barriers (7.0 nm), and a GaAs top layer 5 nm thick. The deposition rate of all layers in the heterostructure was approximately 10 nm/min.

The differences between the two grown structures consisted in different doping schemes for GaAs QWs and $Al_{0.3}Ga_{0.7}As$ barriers. Selective doping of the central region (about 2.6 nm) in the GaAs QWs was carried out in the type II structure simultaneously by both Si and Be at a concentration of $1.2 \cdot 10^{17}$ cm^{-3}. The $Al_{0.3}Ga_{0.7}As$ barriers remained undoped.

In the other case (type III structure), the central region of the QW (about 2.6 nm) was also doped with Si (the surface density of donors was $3 \cdot 10^{10}$ cm^{-2}, which corresponds to a volume concentration of $1.2 \cdot 10^{17}$ cm^{-3}), while *p*-$Al_{0.3}Ga_{0.7}As$ regions, 5 nm thick, with the same surface density of acceptors ($3 \cdot 10^{10}$ cm^{-2}), were formed in the central part of the $Al_{0.3}Ga_{0.7}As$ barriers by doping with Be.

Experimental technique

The samples were placed in a Montana Instruments Cryostation s50 closed-cycle optical cryostat, which has a low vibration level and allows the samples to be cooled to a temperature of about 5 K. The cryostat was equipped with two sequentially installed warm and cold windows made of fused silica, used for optical measurements. A continuous-wave solid-state laser on a neodymium-doped yttrium-lithium fluoride crystal (Nd:YLF) at 527 nm was used for optical excitation of the structures. The laser's power output was tuned with a variable attenuator, which is a rotary optical filter with an optical density gradient. Pumping radiation was directed to the sample via a series of mirrors and focused on the sample surface with a 10× micro lens (Mitutoyo M Plan NIR). The diameter of the laser spot on the surface of the samples was about 20 µm.

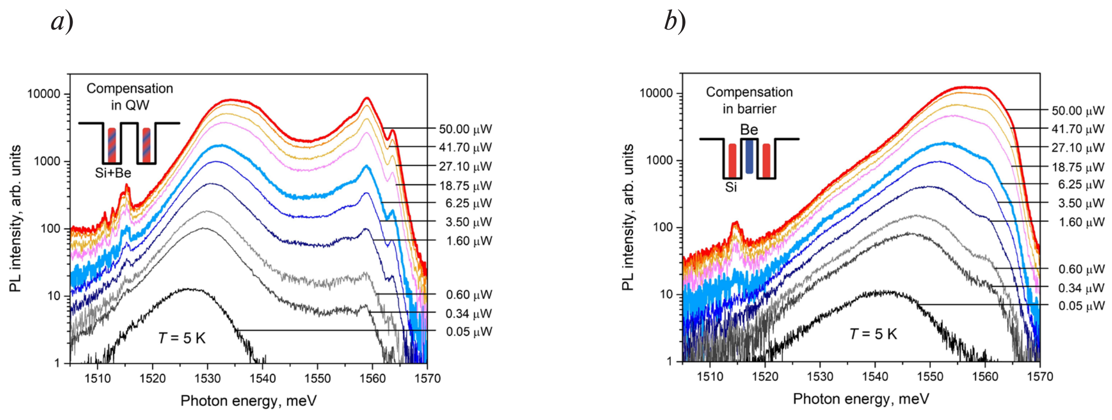

Fig. 2. Experimental PL spectra under interband pumping for a structure with *n*-GaAs/AlGaAs QWs, where donors were compensated by acceptors directly in the QWs (*a*), as well as for the case of compensation in the region of the barriers forming the QWs (*b*)
The numbers of the curves correspond to the pumping intensity

Near-IR PL radiation emitted from the samples was focused to a collimated beam by the same micro lens. The near-IR PL spectra were measured with an Andor Shamrock 500i monochromator with a 1200 l/mm triple-grating turret. Near-IR radiation was detected by a silicon CCD matrix with thermoelectric cooling. The spectral resolution of the obtained PL spectra is characterized by a value of about 0.06 nm (about 0.11 meV).

Results and discussion

PL spectra were obtained for both grown structures at helium temperature in the spectral range from 1505 to 1570 meV, which covers the entire near-IR photoluminescence band (Fig. 2). Evidently, the PL spectra are transformed considerably with an increase in the power of interband optical pumping from 0.05 to 50 μW.

Each spectrum was decomposed into several spectral lines with different profiles (Lorentzian or Gaussian) to analyze the nature of this transformation. The spectral positions of these lines were selected heuristically and assumed to be independent of the pumping level. The resulting spectral curve for a given pumping power was found as the sum of individual spectral lines, whose amplitudes and widths were determined by fitting the resulting curve to the experimental PL spectrum.

The Lorentz profile is characteristic for homogeneously broadened lines, and the Gaussian profile for inhomogeneously broadened lines. The recombination lines of free excitons are assumed to be homogeneously broadened, since their spectral position depends mainly on the width of the quantum wells, which was the same throughout the structure (thanks to precise molecular beam epitaxy). All other lines in the PL spectra are associated with the presence of impurity states in quantum wells. The actual energy spectrum of impurity states essentially depends on the position of the impurity relative to the center of the quantum well, producing inhomogeneous broadening of PL lines due to different positions of the impurities in the quantum well.

It is more convenient to start the analysis of PL mechanisms with a type III structure, where compensation was carried out by placing a layer doped with acceptors in the barrier. Fig. 3 shows the decomposition of the experimental PL spectrum into five lines at two pumping levels. We associate the line with the highest frequency (1563.8 meV) with recombination of free excitons formed from electrons of the $e1$ subband and holes of the $hh1$ subband. These excitons are denoted as $X_{e1\text{-}hh1}$ from now on. The spectral position of the radiative recombination line $X_{e1\text{-}hh1}$ detected experimentally differs from the one expected theoretically by −2.6 meV, likely indicating that the real width of the quantum well is slightly higher (by about 4%) than the nominal one. Computations suggest that the energy of the interband optical transitions $e1 \rightarrow hh1$ at a quantum well width of 7.9 nm is 1572.5 meV, the binding energy of the exciton $X_{e1\text{-}hh1}$ is 8.6 meV [6], and the recombination of this exciton should be accompanied by emission of photons with an energy of 1563.9 meV, which coincides with the experiment up to the spectral resolution. This line has a Lorentzian shape.

The next line (1561 meV) is clearly due to recombination of the exciton bound to the donor (this line is denoted as XD). In the experiment, this line is shifted relative to the $X_{e1\text{-}hh1}$ line by −2.8 meV, which is close to the theoretically calculated value of the binding energy of an exciton bound on a neutral donor equal to −2.5 meV for a quantum well with a width of 7.9 nm [7]. This line has a Gaussian profile.

The next PL line is the most intense; we associate it with the $D \rightarrow hh1$ radiative transitions playing a positive role in the $e1 \rightarrow D$ radiative transitions of the THz range (as discussed above). This line has an asymmetric shape. This is because the doped layer in the quantum well has a finite thickness (approximately 30% of the well width) and the binding energy is the highest for the donors localized in the center of the quantum well, decreasing as the donors move away from the center (see [8]). The energies of $D \rightarrow hh1$ transitions also change accordingly. For simplicity, we described this asymmetric line by a sum of two Gaussian profiles (with spectral positions of the peaks at 1551.0 and 1556.1 meV). Notably, recombination from the deepest donor levels that are the first to be occupied by nonequilibrium electrons is predominant at low pumping levels. These transitions are gradually saturated with increased pumping, and the transitions from lower levels become more and more intense. This is accompanied by a high-frequency shift of the peak on the resulting curve of the two Gaussian profiles. With an increase in pumping from 6.25 to 50 μW, the peak at 1556.1 meV becomes significantly more intense than

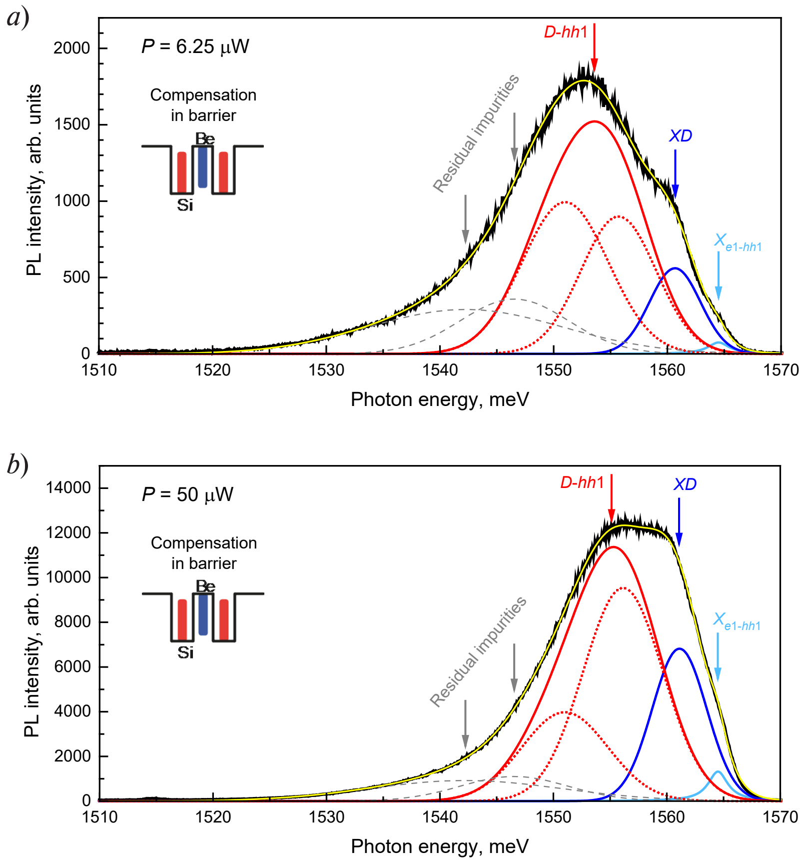

Fig. 3. PL spectra for a structure with *n*-GaAs/AlGaAs QWs, with compensation by acceptor doping of barriers, at different pumping powers, μW: 6.25 (*a*) and 50 (*b*)

Bold solid curves correspond to the experiment, light curves over these lines show the sum of spectral profiles for individual PL lines. Thin solid curves correspond to the profiles of the main PL lines ($X_{e1\text{-}hh1}$, *XD*, *D-hh*1), dashed curves demonstrate weak lines of radiative recombination associated with residual impurities. The spectral positions of PL peaks are marked by arrows. The PL line induced by $D \to hh1$ transitions was described by a sum of two Gaussian profiles (dotted curves)

the peak at 1551.0 meV. As a result, the maximum of the asymmetric line $D \to hh1$ shifts from 1553.6 meV by 1.7 meV towards high frequencies (see Fig. 3). We should note that the energy of the photons corresponding to this peak is lower than the energy of interband transitions $e1 \to hh1$ (1572.5 meV) by 17–19 meV, which is in good agreement with the experimentally observed peak of THz photoluminescence due to electron transitions to the ground state of the donor (in a quantum well of the same width) [5].

There are two more PL lines, which are observed at photon energies less than 1550 meV and have significantly lower intensity compared to the main line $D \to hh1$. We associate these lines with uncontrolled residual acceptor impurities arising during the growth of structures by the MBE method [9] and describe them with a Gaussian profile.

The PL spectra for the type II structure, where donor compensation by acceptors was performed directly in the quantum well, are more complex (Fig. 4). Firstly, they contain three of the above radiative recombination lines: the free exciton line ($X_{e1\text{-}hh1}$), the line of the exciton bound to the donor (*XD*), and the line corresponding to the transitions $D \to hh1$. Another relatively narrow line appears between the last two lines in this structure, clearly corresponding to recombination of the exciton bound to the acceptor (*XA*). It is observed in the experiment at a photon energy of 1559 meV, i.e., shifted by −5 meV relative to the free exciton line $X_{e1\text{-}hh1}$. This shift coincides with the result obtained earlier in the study of beryllium acceptor centers in $GaAs/Al_{0.3}Ga_{0.7}As$ quantum wells of a close width [10] with an accuracy up to 20%.

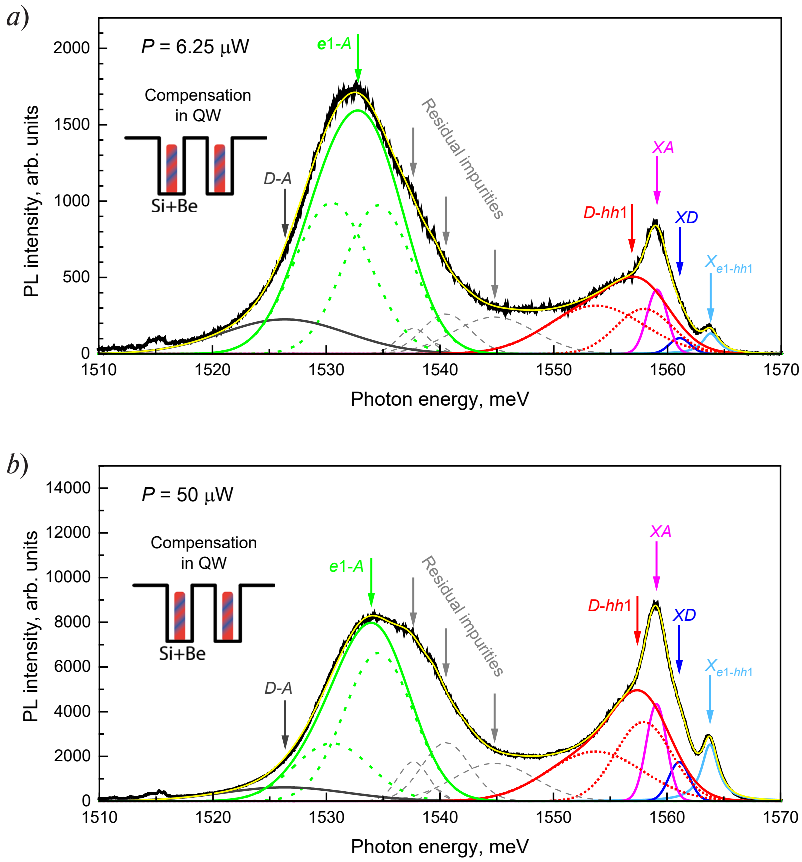

Fig. 4. PL spectra for structure with n-GaAs/AlGaAs QWs, where donor compensation with acceptors was performed directly in the QWs, at different pumping intensities, μW: 6.25 (*a*) and 50 (*b*)

Bold solid curves correspond to the experiment, light curves over these lines show the sum of spectral profiles for individual PL lines. Thin solid curves show the profiles of the main PL lines, dashed curves correspond to weak lines of recombination radiation associated with residual impurities. The spectral positions of PL peaks are marked by arrows. The PL lines induced by $D \rightarrow hh1$ and $e1 \rightarrow A$ transitions were described by a sum of two Gaussian profiles (dotted curves)

Several lines which we associate with various transitions involving acceptors are observed at photon energies lower than 1550 meV. They can be described with Gaussian profiles. The most intense among them is the asymmetric line, which we associate with $e1 \rightarrow A$ transitions. Similar to the $D \rightarrow hh1$ line, this line was also described by the sum of two Gaussian profiles, since the binding energy of the beryllium acceptor in the center of the quantum well is 35 meV and decreases with distance from the center of the quantum well [11]. The spectral positions of the Gaussian profiles, 1531 and 1535 meV, were found by fitting to the experimental PL spectra. With an increase in pumping from 6.25 to 50 μW, the peak of the asymmetric line $e1 \rightarrow A$ shifts from 1533 meV by 1.1 meV towards high frequencies (see Fig. 4). A similar transformation of the PL line due to $e1 \rightarrow A$ transitions was previously observed in structures with GaAs/AlGaAs quantum wells that were not subjected to special doping [12]. Three relatively weak PL lines are observed in the spectral band between the lines $e1 \rightarrow A$ and $D \rightarrow hh1$, which may be associated with uncontrolled residual acceptors. Finally, a weak broad PL line is observed near the energy of 1526 meV, which is rapidly saturated with an increase in pumping (see Figs. 4 and 5,*b*). The spectral position of this line corresponds to the transitions $D \rightarrow A$ (the estimated energy of these transitions is 1525 meV if donors and acceptors are located in the center of the well and 1531 meV at the edge of the doped region).

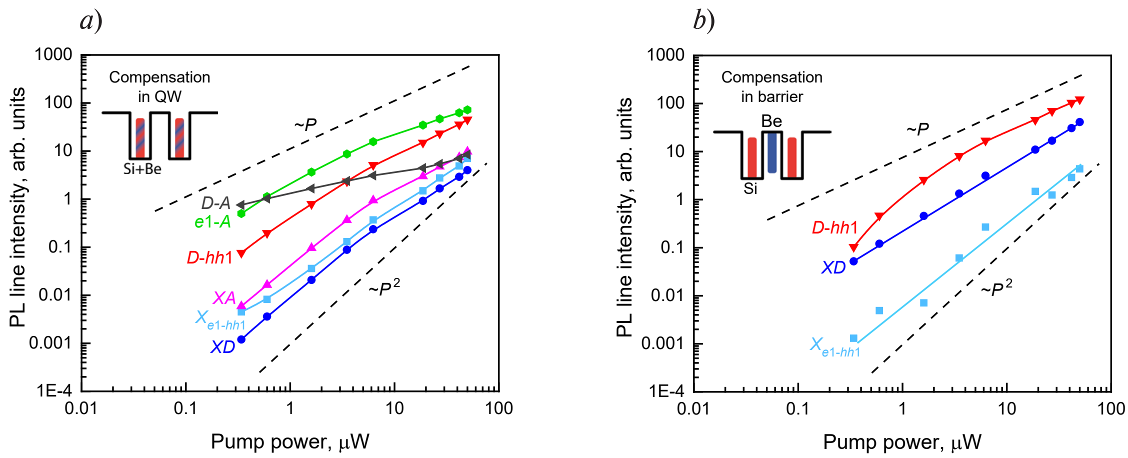

Fig. 5. Experimental dependences (symbols) of the intensity of the main PL lines on the pumping level for different structures with *n*-GaAs/AlGaAs QWs: with donor compensation by acceptors directly in QWs (*a*) and with acceptor doping of barriers (*b*). Solid lines are a guide for the eye

If the pumping levels are sufficiently high, relatively weak narrow lines are detected in the PL spectra of both structures (types II and III) near the photon energy of 1515 meV, not associated with quantum wells but caused by exciton recombination in GaAs bulk layers (substrate and buffer layer) [12].

The intensities of the main radiative recombination lines as functions of optical pumping are shown in Fig. 5. The area under the spectral profile of each PL line is plotted along the vertical axis.

Considering these dependences, let us compare the efficiency of both structures in generating THz radiation at $e1 \to D$ transitions. First of all, the intensity of the line $D \to hh1$ in the type III structure, where compensation was carried out by placing an acceptor-doped layer in the barrier, is 2.3 times higher (at maximum pumping) than the total intensity of the lines $D \to hh1$ and $D \to A$ in the type II structure (where compensation was carried out in the quantum well). Thus, the depopulation rate of the ground donor level is considerably increased in the type III structure compared with the type II structure, which means we can expect an approximately twofold increase in the intensity of THz radiation at $e1 \to D$ transitions. It is also important to note that the line $e1 \to A$ playing a negative role in the type II structure is absent in the type III structure. Even though similar recombination channels associated with uncontrolled residual acceptors are detected in the type III structure, the role of this undesirable recombination mechanism is reduced by 2.2 times.

Thus, studies of the near-infrared PL spectra have confirmed that the generation efficiency of THz radiation associated with impurity transitions under interband optical pumping of GaAs/AlGaAs nanostructures can be improved using a special profile of selective doping: donors in quantum wells and acceptors in barriers.

Conclusion

This paper reports on comparative studies of near-IR photoluminescence in structures with GaAs/AlGaAs quantum wells with different selective doping profiles. Analyzing the photoluminescence spectra at liquid helium temperature for different intensities of interband optical pumping, we established the main mechanisms of radiative recombination. We obtained the dependences for the intensity of the main photoluminescence lines versus the pumping level. We can conclude from our findings that the most efficient GaAs/AlGaAs nanostructures for generating THz radiation at impurity transitions are structures with a special doping profile where the compensating acceptor impurity is located in the barriers forming the *n*-GaAs quantum well rather than in the well itself.

Therefore, the intensity of terahertz photo- and electroluminescence associated with donor states can be significantly increased.

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