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Dipolar biexcitons in lateral traps in Si/SiGe/Si heterostructures

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$Si/Si_{1-x}Ge_x/Si$ heterostructures with long-range lateral potential fluctuations that appear in the capping Si layer near the SiGe/Si heterointerface due to the presence of relaxed areas in the SiGe layer have been studied. Analysis of the low-temperature photoluminescence spectra indicates that, upon the photoexcitation of the structure, the accumulation of nonequilibrium charge carriers, formation of dipolar excitons, and their recombination take place in long-range lateral traps formed by these fluctuations. It is found that, at temperatures $T < 10$ K, a new narrow line appears with an increase in the excitation level at the blue tail of the broad photoluminescence band of dipolar excitons localized by short-range potential fluctuations. At temperatures $T \approx 2$ K, this line is dominant in the spectra even at the lowest excitation levels. It is shown that, at moderate excitation levels, this line is caused by the recombination of free dipolar biexcitons in long-range traps. At high excitation levels, the width of the new line increases by more than a factor of 2 compared to that at lower excitation levels, and under these conditions this line is associated with the recombination of dipolar electron-hole plasma in long-range traps.

Keywords: Two-dimensional systems, electron-hole bilayers, type-II heterostructures, low-temperature photoluminescence.

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1. Introduction

The study of electron-hole bilayers, i.e., systems in which nonequilibrium electrons and holes are located in adjacent spatially separated layers, in quasi-two-dimensional semiconductor heterostructures is a very important task. Back in the 1970s in a number of theoretical studies [1–4] it was shown that, under appropriate conditions, excitons in bilayers (dipolar excitons (DE)) can form both a metallic electron-hole liquid (EHL) and a dielectric exciton liquid, as well as a Bose–Einstein condensate (BEC) of dipolar excitons. The condensed matter phases listed above may have unusual properties, such as large-scale coherence, superconductivity, and superfluidity. Bilayers were experimentally implemented in type-I GaAs/AlGaAs heterosystems upon photoexcitation of structures with double (or wide single) quantum wells (QW) subjected to an electric field perpendicular to the plane of the structure. Bose–Einstein condensation of DEs was observed in these structures [5–9]. It is known that exciton BEC in two-dimensional systems can occur only in the presence of lateral traps confining the motion of excitons to a region of finite dimensions. Traps (of micrometer size) can occur due to the presence of a lateral long-range fluctuation potential [11,12].

In this paper, we study the behavior of a system of nonequilibrium charge carriers in type-II Si/SiGe-based heterostructures, whose band diagram intrinsically ensures that photoexcited electrons and holes separate and accumulate in the neighboring layers of the structure. Experimental studies of such systems are interesting from the point of view of obtaining new data on collective effects in a DE ensemble [13–15].

2. Samples and experimental techniques

In the studied structures, a single layer of $Si_{1-x}Ge_x$ was grown between two layers of pure silicon, and the thickness and composition of the alloy layer corresponded to the metastable region of its pseudomorphic growth [16]. The $Si_{1-x}Ge_x$ layer in such structures (for a low germanium content) forms a QW for holes in the valence band and a low potential barrier for electrons in the conduction band [17,18]. $Si/Si_{1-x}Ge_x/Si$ heterostructures with strained (laterally compressed) c-$Si_{1-x}Ge_x$ layers were grown by molecular beam epitaxy on Si(001) substrates and consisted of the following layers: the lower (buffer) Si layer with a thickness of 100 nm, the $Si_{1-x}Ge_x$ layer with different concentrations of germanium x and thicknesses d, and the upper (cap) silicon layer with a thickness of 100 nm. Previously, we have shown by X-ray diffraction analysis that, with an increase in the growth temperature of the upper Si layer in these structures, there appear micrometer-sized areas where the SiGe layer is partially relaxed and the Si layer grown on top of it turns out to be strained (laterally stretched) [19]. In this situation, the areas of the upper Si layer subject to tensile strain (let's call them t-Si), covering the areas with a relaxed SiGe layer, form QWs for electrons in the conduction band at the heterointerface.

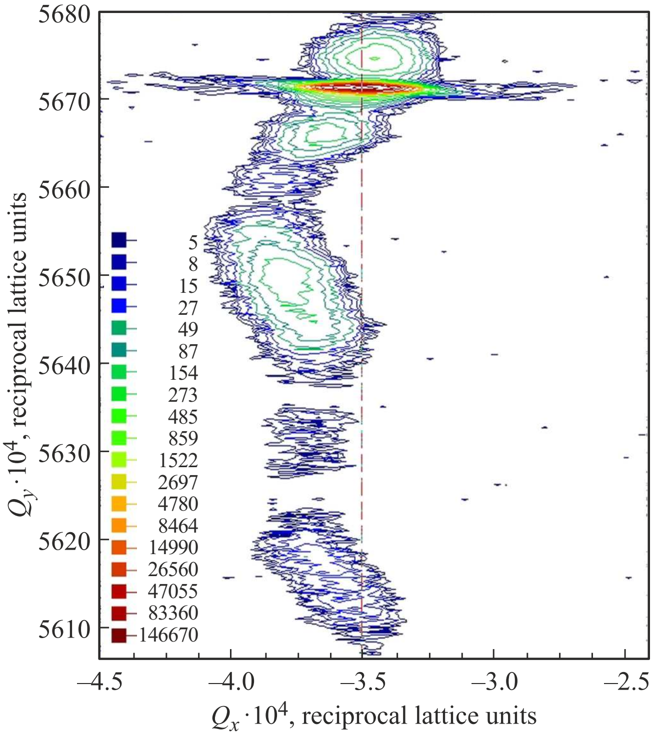

Figure 1. Fragment of a two-dimensional region of the reciprocal space [20] near the (004)Si reflection of the structure under study. Continuous contours indicate lines of equal X-ray intensity, and the dashed line corresponds to the center of the substrate peak along the axis Q_x.

Below, we consider in detail the properties of the structure with the parameters of the $Si_{1-x}Ge_x$ layer $x = 0.1$ and $d = 45$ nm (in such a thick layer, quantum confinement of holes can be neglected) grown at a temperature of 550°C. These parameters correspond to the metastability region [16]. The cap Si layer was grown at a temperature of 650°C.

X-ray diffraction analysis of this structure was carried out on a PANalytical X'PERT PRO MRD Extended X-ray diffractometer with a Ge(220)×4 primary four-crystal monochromator in combination with an X-ray mirror and a third Ge(220)×3 crystal analyzer. A fragment of the X-ray diffractogram for a two-dimensional region of the reciprocal space [20] near the (004) Si reflection is shown in Fig. 1.

The strongest peak from the substrate has coordinates $Q_{y\mathrm{Si}} \times 10\,000 = 5671$, $Q_{x\mathrm{Si}} \times 10\,000 = -3.5$, while the center of the SiGe peak with $Q_{y\mathrm{SiGe}} \times 10\,000 = 5650$ is shifted along the horizontal axis by $\Delta(Q_x \times 10\,000) = -0.25$ from the center of the substrate peak indicated in the figure by a vertical dashed line. This shift along the Q_x axis means that there is misorientation between the (004) planes of the substrate and the SiGe epitaxial layer, which indicates the existence of 60-degree misfit dislocations at the Si/SiGe heterointerface. Misorientation occurs because, due to the inevitable deviation of the original substrate surface from the exact plane (001), dislocations formed during plastic relaxation will have the component of their Burgers vector perpendicular to the substrate predominantly oriented in one direction.

The value $\Delta(Q_x)$ is related to the values of angles in direct space by the expression [20]

$$\Delta(Q_x) = \sin\theta_\mathrm{B}\cos(\theta_\mathrm{B} - \omega)\Delta\omega,$$

where θ_B is the value of the Bragg angle for the Si(004) reflection, ω is the angle of incidence of the X-ray beam on the sample, and $\Delta\omega$ is the deviation of the beam from the angular position of the maximum of the substrate peak. For the symmetric reflection (004) $\omega \approx \theta_\mathrm{B}$, so that $\Delta\omega = 4.4 \cdot 10^{-5}$ rad. This value of misorientation at the small-angle interface between the substrate and the epitaxial SiGe layer corresponds to the distance of $6.2\,\mu$m between the misfit dislocations with the Burgers vector $1/2\langle 110\rangle$ (for estimation, we assume that the Burgers vectors of all arising dislocations are oriented in the same way).

Thus, the spacing between dislocations at the Si/SiGe heterointerface is large enough (macroscopic) that a strained pseudomorphic SiGe layer compressed along the layer plane can grow away from dislocations, whereas a SiGe layer with a partially relaxed structure grows near dislocations. Atomic force microscopy shows that a modified layer with a width of about $1\,\mu$m is formed around each misfit dislocation. Considering that in silicon the atomic spacing along the [110] direction is approximately 0.2 nm, one missing half-plane of the misfit dislocation is distributed between 5000 planes.

As a result, the SiGe layer consists of alternating sections of macroscopic dimensions with strained (compressed) structure and sections with partially relaxed structure. After the SiGe layer is overgrown with the Si cap layer, the plane of the SiGe/(Si-cap) heterointerface is divided into sections with a pseudomorphic strained (laterally compressed) layer c-SiGe, overgrown with an unstrained Si layer, and sections with a partially relaxed layer c-SiGe, overgrown with a strained (laterally stretched) layer t-Si [19]. These sections of t-Si form long-range (micrometer-sized) lateral electron traps. A potential well for electrons is also formed in the vertical direction, since the strain of the Si-cap layer gradually decreases towards the surface of the structure (see Fig. 2, b).

The PL spectra were studied at temperatures T=1.8–15 K and excitation intensities P=7–300 mW/cm^2. The spectra were obtained using a diffraction spectrometer with two output ports, one with a liquid nitrogen cooled germanium p–i–n-photodiode for recording spectra in the near infrared (NIR) range, and the other with a liquid nitrogen cooled CCD matrix detector for recording spectra in the visible range. The spectral resolution of the setup was 1 meV. We remind that the luminescence of Si/SiGe structures in the visible spectral range ($2E_g$ luminescence) can occur due to two-electron transitions, i.e. simultaneous recombination of two electrons and two holes, as a result

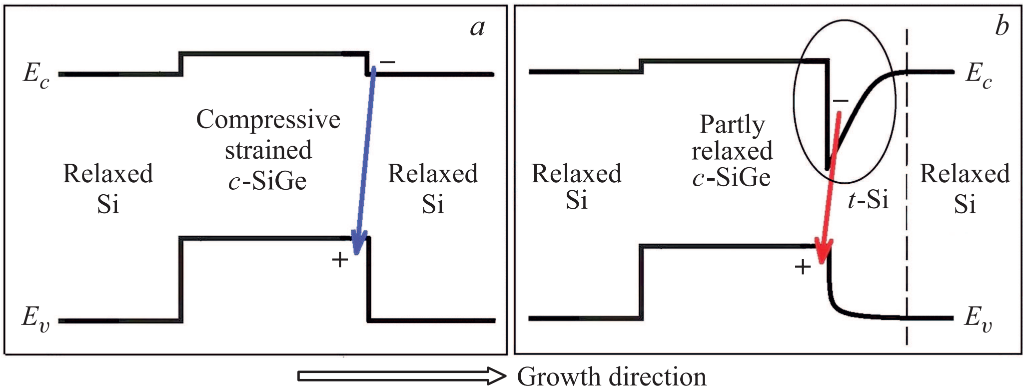

Figure 2. Schematic representation of the Si/SiGe/Si heterostructure band diagram in the direction of its growth. *a*) A pseudomorphic section of the structure. *b*) A section with a partially relaxed layer *c*-SiGe and an interfacial QW for electrons (circled by a thin line) in the tensile strained layer *t*-Si on top of it.

of which a photon is emitted with an energy equal to the total energy of recombining particles [21,22]. Luminescence in the NIR region of the spectrum occurs upon conventional single-electron transitions, when one electron-hole pair recombines in each emission act. Comparison of one- and two-electron spectra contributes to more reliable interpretation of the origin of various components in the PL spectrum, since there are no emission lines of single excitons (both free and bound) in the two-electron spectra. In our measurements, the PL spectra in the NIR and visible ranges could be recorded under strictly the same experimental conditions. In this case, radiation from a titanium-sapphire laser ($\lambda_{\text{exc}} = 790$ nm) was used to excite the PL of the samples. The PL spectra in the NIR range were also measured under excitation by a helium-cadmium laser ($\lambda_{\text{exc}} = 440$ nm). However, in this case, the $2E_g$ luminescence of the structures is not observable against the background of a wide band of „ordinary" hot luminescence. The exciting radiation was focused on the surface of the sample into a spot with a diameter of about 2 mm. PL was collected using a lens normally from the illuminated surface of the structure and focused onto the entrance slit of the spectrometer.

3. Results and discussion

Figure 2 schematically shows the real-space band diagram of the investigated heterostructure in the direction of its growth. The arrows show radiative transitions between electrons in the upper Si layer and holes in the SiGe layer. Fig. 2, *a* shows the band diagram of a pseudomorphic section of the structure, i.e., a section with a fully strained (laterally compressed) layer *c*-SiGe, covered with an unstrained upper layer Si-cap. In this case, dipolar excitons are formed from heavy holes in the fully strained *c*-SiGe layer and electrons from Δ_4-valleys [23] in the unstrained upper Si-cap layer (excitons formed by Δ_2-valley electrons have a slightly higher energy).

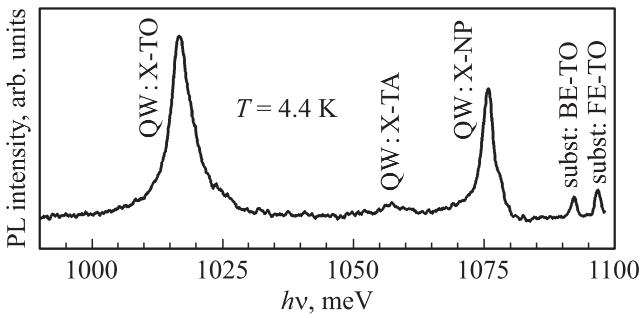

Figure 3. Survey spectrum of the PL of the studied structure in the NIR region ($\lambda_{\text{exc}} = 440$ nm, $P = 170\,\text{mW/cm}^2$, $T \approx 4.4$ K).

Fig. 2, *b* shows the band diagram of a section with a partially relaxed layer *c*-SiGe. In this case, an interfacial QW for electrons (IQW) is formed in the conduction band near the SiGe/Si-cap heterointerface in the laterally stretched *t*-Si layer. The IQW is circled by a thin line in Fig. 2, *b*. Now, dipolar excitons and biexcitons are formed from heavy holes in the partially relaxed *c*-SiGe layer and electrons of the Δ_2-valley [18] in the *t*-Si layer, where IQWs represent lateral traps for electrons. As a result, long-range lateral traps for DE are obtained in such areas with a partially relaxed SiGe layer.

Figure 3 shows a survey NIR spectrum of the PL of the heterostructure under study, obtained at an average excitation level of $P = 170\,\text{mW/cm}^2$ ($T \approx 4.4$ K).

Similarly to the spectra of homogeneous Si/SiGe/Si structures with a completely pseudomorphic strained layer of SiGe [24], this spectrum features emission lines arising from the recombination of nonequilibrium dipolar electron-hole pairs (in this case, electrons in IQWs in the *t*-Si layer and holes in the SiGe layer): no-phonon line QW : X-NP and lines of its phonon replicas QW : X-TA and QW : X-TO with emission, respectively, of transverse acoustic (TA) and optical (TO) phonons. The subst : FE-TO and subst : BE-TO lines are caused by recombination of free and bound bulk excitons with TO-phonon emission in the silicon substrate, buffer and cap layers [25].

The intensity of the PL of dipolar excitons in Si/SiGe/Si structures significantly depends on the efficiency of collecting photoexcited charge carriers and excitons from Si layers to the SiGe/Si interface. In structures with IQW traps, the ratio of the integrated intensities of the PL of dipolar excitons to the PL from the substrate, buffer and protective Si layers turned out to be significantly higher than the corresponding ratio in homogeneous Si/SiGe/Si structures without traps [24]. Note also that the QW lines in the spectra of structures with traps turned out to be rather narrow. Thus, the half-width of the QW:X-NP line in Fig. 3 is approximately 2 meV, whereas the half-width of a similar line in homogeneous structures without traps is about 4 meV [14].

Figure 4 shows the behavior of no-phonon PL lines of the studied structure at different excitation levels and temperatures. At $T = 4.4$ K and low excitation intensities (Fig. 4, *a*, $P \leq 16\,\mathrm{mW/cm^2}$), the spectrum is dominated by a low-energy emission band, indicated in Fig. 4, *a* as SiGe/*t*-Si:LE-NP.

With an increase in the excitation intensity, the maximum of this band shifts towards high energies. On the high-energy side of the band SiGe/*t*-Si:LE-NP, a weak emission line, designated QW:Ex-NP, is observed in the spectra, the energy position of which does not change with increasing pump level. We believe that this band and this line arise due to the recombination of DE inhabiting long-range lateral traps, i.e., formed by holes in the relaxed sections of the SiGe layer and electrons at the quantum-confinement levels in IQWs in the corresponding strained regions of the upper Si layer. At low temperatures and low excitation levels, these DE are localized in potential wells of short-range fluctuation profile, which leads to the formation of the band SiGe/*t*-Si:LE-NP. Its half-width, therefore, is determined mainly by the spread of short-range potential levels associated with statistical fluctuations of the Ge distribution in the SiGe layer [26]. The emission of the QW:Ex-NP line occurs upon the recombination of „almost free" DEs in strain-induced long-scale lateral traps, whose dimensions are large enough for excitons inside them to move freely.

With an increase in the pump level (Fig. 4, *a*, $P > 16\,\mathrm{mW/cm^2}$), on the high-energy tail of the band SiGe/*t*-Si:LE-NP, about 2 meV below the QW:X-NP line, there appears a narrow line QW:X-NP whose width remains unchanged up to the pump level $P \approx 170\,\mathrm{mW/cm^2}$. The ratio of the intensities of lines QW:X-NP and QW:Ex-NP increases linearly with increasing pump level. This behavior indicates that the QW:X-NP line is caused by recombination radiation of dipolar biexcitons with binding energy $\Delta \approx 2$ meV relative to dipolar excitons QW:Ex. Note also that the maximum of the band SiGe/*t*-Si:LE-NP continues to shift towards higher energies with an increase in the excitation level, and its intensity relative to the other lines in the spectrum decreases. This is explained by the saturation of more localized states (and deeper states are saturated faster due to their greater occupancy). This band is almost unobservable at $P = 170\,\mathrm{mW/cm^2}$.

The density of excitons generated by pump radiation is equal to (taking into account the Fresnel reflection of pump radiation)

$$n_{\mathrm{ex}} = \eta \left(\frac{n-1}{n+1} \right)^2 \frac{\tau P}{h\nu},$$

where n is the refractive index of the material, $h\nu$ is the pump photon energy, τ is the lifetime of excitons, and η is the efficiency of their generation. Assuming $\tau = 1\,\mu$s and $\eta = 0.5$, we get as an estimate $n_{\mathrm{ex}} \approx 10^{11}\,\mathrm{cm^{-2}}$ for $P = 170\,\mathrm{mW/cm^2}$. Thus, the narrow QW:X-NP line appears under conditions of weak excitation $n_{\mathrm{ex}} a_{\mathrm{ex}}^2 \ll 1$. At $28\,\mathrm{mW/cm^2} < P < 170\,\mathrm{mW/cm^2}$, the half-width of the SiGe:X-NP line is significantly smaller than the half-width of the SiGe/*t*-Si:LE-NP band, so its nature cannot be related to localized states in the short-range fluctuation potential.

Fig. 4, *b* shows how the NIR spectra of no-phonon PL change with increasing temperature at $P = 50\,\mathrm{mW/cm^2}$. It can be seen that the half-widths of the lines QW:X-NP and QW:Ex-NP are comparable at all temperatures. At $T = 1.8$ K, only the QW:X-NP line with a half-width of about 2 meV is seen in the spectrum of the structure. As the temperature increases, the QW:X-NP line goes out, while the QW:Ex-NP line lights up. At $T = 15$ K, only the line QW:Ex-NP is observed, its half-width being 3−4 meV. Note that the energy position of the maximum of this line shifts with an increase in temperature towards high energies (by no more than 1 meV when the temperature changes from 6 to 15 K).

Fig. 4, *c* shows how the PL spectra change with increasing temperature at a low excitation level of $P = 7\,\mathrm{mW/cm^2}$. Only the line QW:X-NP is visible at $T = 1.8$ K in the spectrum of the structure, similarly to the case of higher excitation (Fig. 4, *b*). This line rapidly goes out with an increase in temperature, and only the PL band SiGe/*t*-Si:LE-NP and the line QW:Ex-NP are observed in the spectra at $T = 4.4$ K at a low excitation level. The intensity of the PL band SiGe/*t*-Si:LE-NP drops significantly with a further increase in temperature, and its maximum shifts towards low energies. The intensity of the QW:Ex-NP line decreases relatively weakly, and the energy position of its maximum, as with a higher level of excitation (Fig. 4, *b*), slightly shifts to high energies with increasing temperature.

The observed behavior of the PL spectra agrees well with the proposed interpretation of the lines in these spectra. The decrease in intensity and disappearance of the QW:X-NP line and the appearance of the QW:Ex-NP line with an increase in temperature is explained by thermal dissociation of biexcitons. The concentration of biexcitons is quadratically related to the concentration of excitons; at equilibrium we have $n_{\mathrm{biex}} \propto n_{\mathrm{ex}}^2 \exp(\Delta/kT)$. Since the relative concentration of biexcitons decreases with a decrease in the excitation level, at a low level ($P = 7\,\mathrm{mW/cm^2}$) the QW:X-NP line is seen only at the lowest temperatures, while at a higher

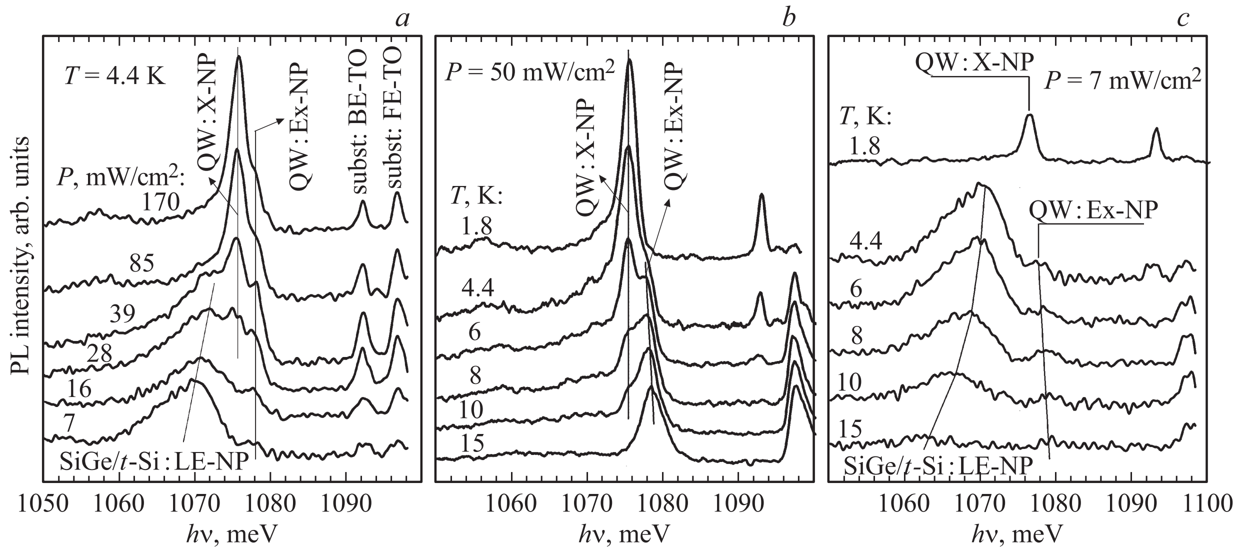

Figure 4. Spectra of single-electron no-phonon PL of the studied structure in the NIR region when excited by radiation with a wavelength of $\lambda_{\text{exc}} = 440$ nm, obtained *a*) at $T = 4.4$ K and various excitation levels, and (*b, c*) at different temperatures and excitation levels: *b*) $P = 50$ mW/cm^2, *c*) $P = 7$ mW/cm^2. The spectra are normalized for the convenience of perception.

level ($P = 50$ mW/cm^2) this line disappears only when $T > 10$ K. It should also be noted that no signs of the formation of an electron-hole liquid are detected in the PL spectra: with a further increase in the excitation level, the formation of an electron-hole plasma is observed (see below), i.e., the condensed phase in the structures under study turns out to be unstable.

The decrease in the PL intensity and the shift of the maximum of the band SiGe/*t*-Si : LE-NP to low energies with an increase in temperature can be explained by the thermal emptying of traps with low binding energy in the short-range fluctuation potential.

Figure 5 compares the normalized single-electron no-phonon PL spectra in the NIR region with the normalized spectra of $2E_g$ luminescence in the visible range obtained at a temperature of $T = 1.8$ K at low and high excitation levels in the region of the QW : X-NP line. The upper and lower photon-energy axes correspond to the spectra in the NIR and visible spectral ranges, respectively. For convenient comparison, the scale of the lower energy axis is compressed twofold relative to the upper one, so that the positions of the lines in both spectra can be compared visually, without any recalculations.

Fig. 5, *a* shows the PL spectra recorded at a low excitation level ($P = 10$ mW/cm^2). In this case, the NIR region of the spectrum, as noted above, is dominated by the line QW : X-NP. Its half-width is about 2 meV. It can be seen (referring to the scale of the visible range) that the doubled value of the energy of its maximum is approximately 2 meV lower than the energy of the maximum of the $2E_g$ luminescence line in the visible range. As expected, this shift is approximately equal to the binding energy of the biexciton determined from the NIR PL spectra, which confirms the biexciton nature of the PL line under discussion. The data shown in Fig. 5, *a* prove that at low excitation levels $P < 170$ mW/cm^2 the QW : X-NP line is indeed the emission line of dipolar biexcitons QW : BiEx-NP with binding energy $\Delta \approx 2$ meV relative to excitons QW : Ex. The efficient formation of biexcitons in the studied structure also indicates the existence of long-range traps in which excitons accumulate upon photoexcitation. A weak tail at low energies from the QW : X-NP line, which, as can be seen from Fig. 5, *a*, is observed in the PL spectrum both in the NIR and visible ranges, possibly occurs upon recombination of biexcitons localized in potential wells of the short-range profile caused by fluctuations in the composition of the $Si_{1-x}Ge_x$ layer. With an increase in the excitation level, two excitons can be localized in these wells with the formation of localized biexcitons (LBiEx). In this case, the half-width of the LBiEx PL band reflects the depth and lateral size distributions of the wells of the random short-range potential. According to Ref. [24], in Si/SiGe/Si structures without long-range traps, the half-width of the band of no-phonon emission from localized excitons and localized biexcitons in the spectra of the NIR range is approximately 4 meV, and the half-width of the line of localized biexcitons in the spectrum of $2E_g$ luminescence in the visible range is approximately 8 meV.

Fig. 5, b shows the spectra of the structure recorded at a high level of excitation $P = 275$ mW/cm^2, almost twice as high as the maximum level in Fig. 4, *a*. It can be seen that, at this level, the maximum of the QW : X-NP line in the single-electron PL spectrum, is shifted towards higher energies from its position at a low level, and its half-width has increased from 2 to 4 meV. At the same time, the contour of this line, recalculated to the scale of the visible range, nearly coincides in position, shape and width with the contour of the $2E_g$ luminescence line. It should be

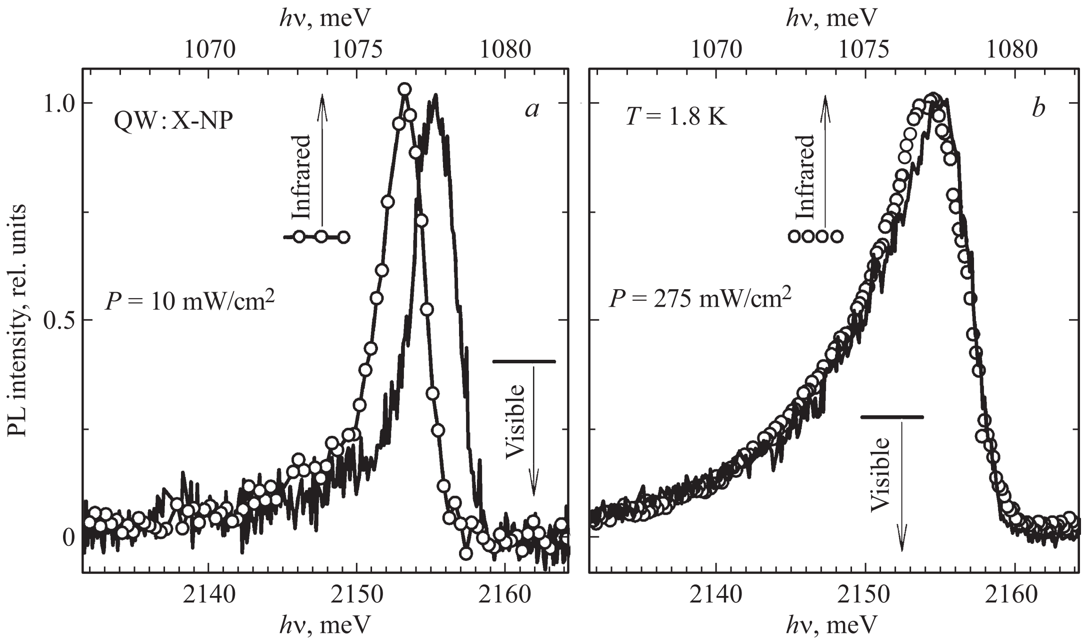

Figure 5. Comparison of single-electron no-phonon PL spectra of the studied structure in the NIR range (lines with empty points, energy scale at the top) with the spectra of $2E_g$ luminescence in the visible range (continuous lines, energy scale at the bottom) at $T = 1.8$ K. The spectra are recorded under excitation by radiation with a wavelength of $\lambda_{\text{exc}} = 790$ nm at *a*) low ($P = 10$ mW/cm^2) and *b*) high levels of excitation ($P = 275$ mW/cm^2).

noted that at a high level of excitation, the half-width of the $2E_g$ luminescence line increased from 4 to 8 meV, mainly due to the broadening of the low-energy tail, while its high-energy slope remains nearly unchanged. The position of the maximum intensity shifted towards lower energies by no more than 1 meV. The broadening of the PL line and the absence of a shift corresponding to the binding energy indicates [13] that, at a high level of excitation, the emission is caused by recombination of electron-hole pairs in (dipolar) electron-hole plasma in long-range traps.

4. Conclusion

The structural properties and the spectra of low-temperature PL ($T = 1.8-15$ K) of Si/SiGe/Si heterostructures with a partially relaxed SiGe layer are investigated. The existence of long-range fluctuations in the potential profile at the SiGe/Si interfaces in the studied structure is shown. At low excitation levels, a wide band of recombination radiation from DEs localized in the wells of short-range fluctuation potential profile is observed in the low-temperature PL spectra of these heterostructures. At sufficiently low temperatures ($T < 10$ K), with an increase in the excitation level on the high-energy tail of this band, there appears a new line, which is also observed in the spectra of $2E_g$ luminescence in the visible range. The width of the new line at low excitation levels is significantly smaller than the width of the DE band. Comparison of the no-phonon spectrum in the NIR range and the $2E_g$ spectrum in the visible range shows that, at relatively small excitation levels, this line is due to recombination of free dipolar biexcitons confined in long-range lateral traps at the SiGe/Si interface. The absence of real-space condensation of dipolar excitons and biexcitons with the formation of an electron-hole liquid suggests the possibility of observation of the PL of Bose–Einstein condensate of dipolar biexcitons at lower temperatures.

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Conflict of interest

The authors declare that they have no conflict of interest.

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