Four-Particle Recombination Luminescence of Electron-Hole Liquid and Biexcitons in SiGe Quasi-Ttwo-Dimensional Layers of Silicon Heterostructures in the Visible Spectrum

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In this study, we investigate the energy spectrum and collective effects in the system of excitons in strained SiGe layers in a series of Si/Si$_{1-x}$Ge$_x$/Si heterostructures with $0.05 < x < 0.25$ and the layer thickness $d = 2$–5 nm. We use the low-temperature photoluminescence spectroscopy both in the near-infrared and the visible spectral regions. In the latter case, the luminescence originates from simultaneous recombination of two electrons with two holes.

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1. INTRODUCTION

This work is devoted to the study of the properties of electron-hole excitations in the strained quasi-two-dimensional SiGe layers of Si/Si$_{1-x}$Ge$_x$/Si heterostructure. The layers had a thickness of $d° =°2$-5 nm, less than the exciton Bohr radius in silicon, equal to about 5 nm. Recently [1-3], quasi-two-dimensional electron-hole liquid (EHL) was found in such heterostructures by photoluminescence spectroscopy (PL). In [1,°2] investigations were carried out in structures with a small germanium content $x = 0.05$ in thick layers of 70-5 nm. In [3] EHL was found in a layer thickness of $d = 2$ nm at $x \approx 0.09$. This paper presents the results of the study of structures with $x \approx 0.09$ and a thickness of layer 2 and 4 nm, carried out by low-temperature photoluminescence in the near infrared (NIR) and visible spectral region, arising in the so-called two-electron transitions, i.e., when recombination occurs simultaneously with 2 electrons and 2 holes [4-5]. The energy of the photon emitted in this recombination is approximately 2 times greater than with usual electron-hole recombination, and emission lines are in the yellow-green spectral region. In this spectrum region there is no emission line of free and bound on impurities excitons, as well as other objects that contain less than two electron-hole pairs. Therefore, comparison of the one-electron (observed in the NIR) and two-electron spectra leads to more accurate conclusions about the origin of the various components of the spectrum.

It was found that in the structures with $d = 2$ nm at sufficiently low temperatures and high levels of excitation the electron-hole liquid which consists of quasi-two-dimensional holes and quasi-three-dimensional electrons is formed in SiGe-layer. It is shown that the gas phase includes both the excitons and biexcitons. In the PL spectrum of the heterostructure with $d = 4$ nm only emission of localized excitons and biexcitons was observed, and the radiation EHL was absent. Apparently, this difference arises due to the fact that Si/Si$_{1-}$ $_x$Ge$_x$/Si heterostructures belong to type II, with the Si-Ge layer representing a potential barrier for electrons and a potential well (QW) for holes [6–8]. Generally, nonequilibrium electrons and holes in heterostructures of this type are spatially separated: holes occupy a QW in the SiGe layer and electrons are located predominantly in Si layers in the vicinity of the Si/SiGe hetero-interface; thus, excitons are spatially indirect and the net exciton–exciton interaction is repulsive. At low temperature this excitons are localized in potential wells arising due to the heterogeneity of the composition in the plane of the layer and at the heterointerface. However, if the tunneling transparency of the barrier is high enough, it is possible that the maximum in the electron-density distribution occurs within the SiGe layer [8]. In this case, excitons are spatially direct and an interaction between them may result in the formation of biexcitons and condensation into the EHL. Note that in [1,°2] Si/Si$_{1-x}$Ge$_x$/Si heterostructures with $x = 0.05$ and $d = 25$-70 nm are type-II as well. However, at high excitation levels, band bending that originates from the Coulomb interaction of spatially separated electrons and holes results in the transition of such a structure to type I.

2. EXPERIMENTAL

Strained-layer SiGe structures were grown by molecular-beam epitaxy on Si(001) substrates. Thicknesses of the buffer and the cap Si layers were 100 nm and 100-200 nm, respectively. The PL spectra in the near-infrared (NIR) spectral range were recorded for the sample temperatures $T = 2$–60 K and excitation levels $P = 0.05$–300 W/cm². A He–Cd laser was used as the PL excitation source and the recombination radiation was analyzed by a grating monochromator and detected by a liquid-nitrogen-cooled Ge p–i–n photodiode. Also, spectra of the PL in the visible range were recorded. This PL results from the so-called two-electron transitions, i.e., from simultaneous recombination of two

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electrons and two holes accompanied by the emission of a single photon whose energy equals the total energy of the four recombining particles. These measurements were carried out for $T = 15$ K and $P = 0.3$–150 W/cm^2. A Ti-sapphire laser tuned to $\lambda = 0.75$ μm was used as the PL excitation source. The recombination radiation was analyzed by a grating spectrometer and detected by a liquid-nitrogen-cooled CCD matrix. A comparison of one- and two-electron spectra makes for more reliable identification of different components in the conventional (one-electron) PL spectra, since lines in the two-electron spectra cannot originate from species containing less than two electron–hole pairs.

3. RESULTS AND DISSCUSSION

Fig. 1 shows the NIR PL spectra of the SiGe layers in the structures under study measured at a temperature of 15 K. At a low excitation level the spectra contain a no-phonon free-exciton and localized-exciton lines respectively FE-NP (b) and LE-NP (a) and its transverse-optical phonon replica FE-TO (b) and LE-TO (a). In the spectrum of the structure with a 2-nm SiGe layer, these lines are shifted to higher energies in comparison to the corresponding lines in the spectrum of the structure with a 4-nm SiGe layer, which is related to the larger quantum-confinement energy of holes in the thinner layer. With increasing excitation level on the low-energy side of exciton lines appear narrow emission lines of biexcitons - localized LBiE-NP and LBiE-TO (in the structure with $d = 4$ nm) and free FBiE-NP and FBiE-TO (in the structure with $d = 2$ nm). With a further increase in the excitation level in the spectrum of heterostructure with a layer thickness of 2 nm appears broad no-phonon line EHL (EHL-NP) and its transverse-optical phonon replica EHL-TO. In the heterostructure with a layer thickness of 4 nm EHL does not occur at the maximum used pumping. Apart from lines originating from the emission of the SiGe layer, the PL spectra of the structures under study exhibit well-known lines of bulk Si (originating from the buffer and cap layers), marked respectively in Fig. 1.

The heterostructures under consideration differ from each other in the tunneling transparency of the potential barriers that repel nonequilibrium electrons from the SiGe layer. A 2-nm-thick barrier is rather transparent for electrons and, thus, the maximum in the electron density appears in the middle of the barrier layer. Binding with holes, electrons form spatially-direct excitons and, under increased excitation levels, free biexcitons and the EHL. A 4-nm-thick barrier is much less transparent, which results in spatial separation of electrons and holes; thus, all other factors being the same, the EHL does not form in this case.

The intensity of the FBiE line varies superlinearly (nearly quadratically) with the intensity of the FE line, the latter increasing linearly with the excitation level. This behavior is typical of the free-biexciton PL.

An additional argument in favor of the above interpretation of the photoluminescence lines shown in NIR is provided by the observation in the visible spectral range of the corresponding no-phonon lines associated with the two-electron transitions (Fig. 2).
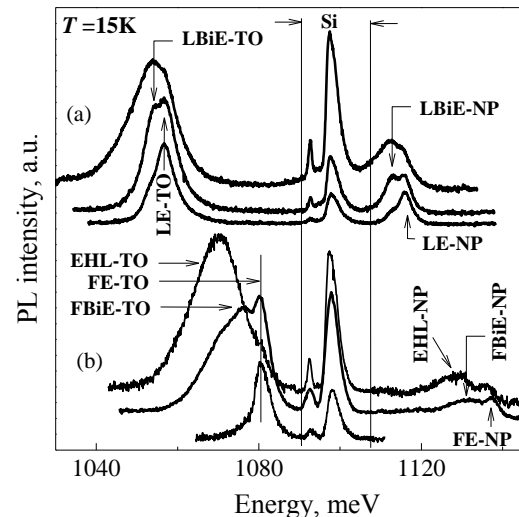
Fig. 1 – The transformation of the PL spectra in the NIR structures with thicknesses of SiGe-layers $d =$ (a) 4 nm and (b) 2 nm, with increasing pump intensity P. (a) – $P = 0.2$, 1.5, 170 W/cm^2, respectively, from the lower to upper range. (b) – $P = 0.35$, 1.9, 170W/cm^2, respectively. $T = 15$ K
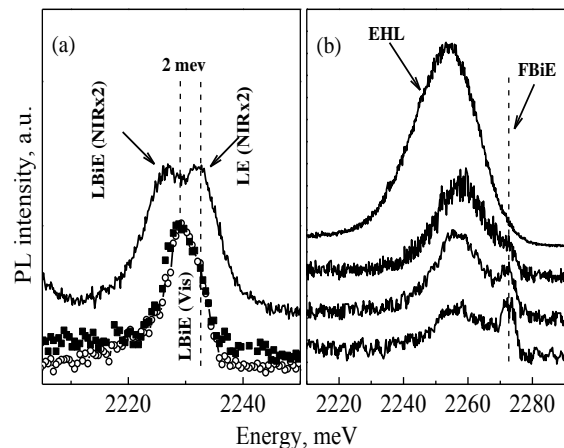
Fig. 2 – Two-electron photoluminescence spectra in the visible region, depending on the density of the pump P. $T = 15$ K. (a) – structure of the SiGe layer with a thickness of 4 nm. $P = 0.3$ (squares), 1.2 (circles) W/cm^2. The upper spectrum was obtained from the NP components of the spectrum measured in the NIR region (Fig. 1a, middle spectrum), by doubling the PL photon energy. (b) - a structure with a layer thickness of 2 nm SiGe. $P = 0.5$, 1, 2.5, 150 W/cm^2 from the lower to the upper range, respectively

In the spectrum of the structure with $d = 4$ nm only a narrow line of localized biexcitons was observed (Fig. 2a). The binding energy of localized biexcitons has been identified (2 meV). It is defined as the difference between the energy PL quanta LBiE in the visible region and double-quantum energy LE-NP in the NIR (see top spectrum in Fig. 2a). In the structure with $d = 2$ nm at relatively low pumping levels the narrow line emission of recombination of biexcitons was observed (Fig. 2b). With increasing excitation broad emission line EHL appeared on the low-energy side of the biexciton lines in the two-electron spectrum. The EHL line is dominant in the spectra at sufficiently high excitation levels, and its shape does not change as P varies from 2 to 150 W/cm^2.

Thus, in this paper we have obtained the following results:

- In Si/Si$_{1-x}$Ge$_x$/Si heterostructures with a tunnel-transparent barrier for electron emission of quasi-two-dimensional electron-hole liquid and free biexcitons was detected in the yellow-green spectral region, arising at two-electron transitions. These results provide new independent evidence of the formation quasi-two-dimensional electron-hole liquid in these heterostructures.

- Requirements for band structure parameters under which the electron-hole liquid and biexcitons of this type forms have been installed.

- In the heterostructures with low transparency tunnel barrier for electrons in the conduction band the photoluminescence of localized spatially indirect biexcitons detected, which was observed up to a temperature of 15 K.

- From the comparison of the spectra in the NIR and visible region the binding energy of localized spatially indirect biexcitons is determined, which is equal to 2.0 meV.

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REFERENCES

1. T.M. Burbaev, E.A. Bobrik, B.A. Kurbatov et al, *JETP Lett.* **85**, 331 (2007).
2. V.S. Bagaev, V.S. Krivobok, S.N. Nikolaev et al., *Phys. Rev. B* **82**, 1153131 (2010).
3. T.M. Burbaev, M.N. Gordeev, D.N. Lobanov et al., *JETP Lett.* **92**, 305 (2010).
4. K. Betzler and R. Conradt, *Phys. Rev.Lett.* **28**, 1562 (1972).
5. T.W. Steiner, L.C. Lenchyshyn, M.L.W. Thewalt et al.,
Solid State Commun. **89**, 429 (1994).
6. M. Rieger, and P. Vogl, *Phys. Rev. B* **48**, 14276 (1993).
7. T. Baier, U. Mantz, K. Thonke et al., *Phys. Rev. B* **50**, 15191 (1994).
8. C. Penn, F. Schaffler, G. Bauer, and S. Glutsch, *Phys. Rev. B* **59**, 13314 (1999).