Supplemental Material to the article
“Visible luminescence of a dense biexciton gas in SiGe/Si quantum wells under anisotropic deformation”

1. The nature of 2Eg-luminescence for quantum wells with a germanium content of $\sim 10\,\%$ at low and moderate excitation densities.

Below, we present the main arguments in favor of the biexciton mechanism of radiative recombination in SiGe/Si quantum wells (QWs) at a temperature of 5 K. Trends are fully preserved for a concentration range of 9–13 %. Therefore, further results will be given for one of the structures with a germanium content of $\sim 9.5\,\%$, for which the experimental data are the most complete.

At a temperature of 5 K under conditions of pulsed excitation (355 nm, 5 ns), for quantum wells with a germanium content of 9–13 %, one main emission line is detected. As carriers recombine, this line replaces the radiation of a degenerate electron-hole plasma. This line is characterized by a fixed lifetime ($\sim 1\mu$s) and a weak dependence of the shape on the concentration of electron-hole pairs. The corresponding electronic state is a ground state of the electron-hole system in a wide range of its densities.

A joint analysis of the IR- and 2Eg-spectra confirms the many-particle nature of the discussed line. The structure of the 2Eg-spectrum (a narrow symmetrical line) observed at moderate excitation densities is characteristic of interactions in which only two electron-hole pairs participate [JETP **121**(6), 1052 (2015)]. In other words, at moderate excitation densities the line under discussion corresponds to biexciton luminescence.

With increasing excitation power density, the decay time corresponding to this line remains fixed, but some changes in the shape of the line in the 2Eg-spectrum are observed (appearance of the long-wave tail). Estimation of the electron-hole pairs concentration on the basis of measurements of the PL decay time in two-color experiments indicate that the appearance of the tail corresponds to the conditions under which scattering of the biexcitons must be significant [JETP **121**(6), 1052 (2015)]. It seems that the appearance of a weak interaction between the biexcitons leads to the indicated change in the shape of the line. Features of the multiparticle states photoluminescence for the structure with a quantum well $Si_{0.91}Ge_{0.09}/Si$ are considered in detail in the article [JETP **121**(6), 1052 (2015)].

Depending on the composition of SiGe QW, various multiparticle states determine the electronic spectrum of a two-dimensional system. The analysis of stationary and time-resolved photoluminescence at various temperatures and excitation densities indicates that in the quantum wells with germanium content (~ 3–$6\,\%$) the first state is an electron-hole droplets (EHD). The EHD, which is a degenerate electron-hole plasma occupying a bounded region of the quantum well, is characterized by a large line width (~ 0 meV), a shorter lifetime (150–400 ns), and fixed shape of the emission line with pumping. The second state corresponds to the “gas” phase radiating from the regions not occupied by the EHD. With a gradual increase of germanium content, the EHD becomes unstable, while the second multiparticle state is preserved. This state corresponds to the biexciton line discussed above for QWs with a germanium content of 9–13 %. More detailed experimental and theoretical analysis of the EHD radiation is presented in [Vasilchenko, JETP, 2018, accepted for publication]

Thus,

(i) the line observed in the emission spectrum of quantum wells with a germanium content of 9–13 % at a temperature 5K corresponds to a multiparticle state that includes two electron-hole pairs or more.

(ii) this state is characterized by a fixed lifetime, which does not depend, in particular, on the power density of the stationary excitation. The invariance of the lifetime excludes the connection between the state under discussion and the degenerate plasma, for which the lifetime of the electron-hole pairs depends on the concentration due to Auger processes.

(iii) at moderate excitation densities, the discussed multiparticle state in the 2Eg-spectrum is characterized by a relatively narrow, symmetrical line. That excludes the “recoil” effect when two electron-hole pairs are simultaneously recombined. The absence of a recoil effect indicates that the state is formed by only two electron-hole pairs.

(iv) An increase in the excitation power density leads to the appearance of an interaction between the biexcitons, which affects the shape of the emission line. Nevertheless, the role of biexciton states remains dominant.

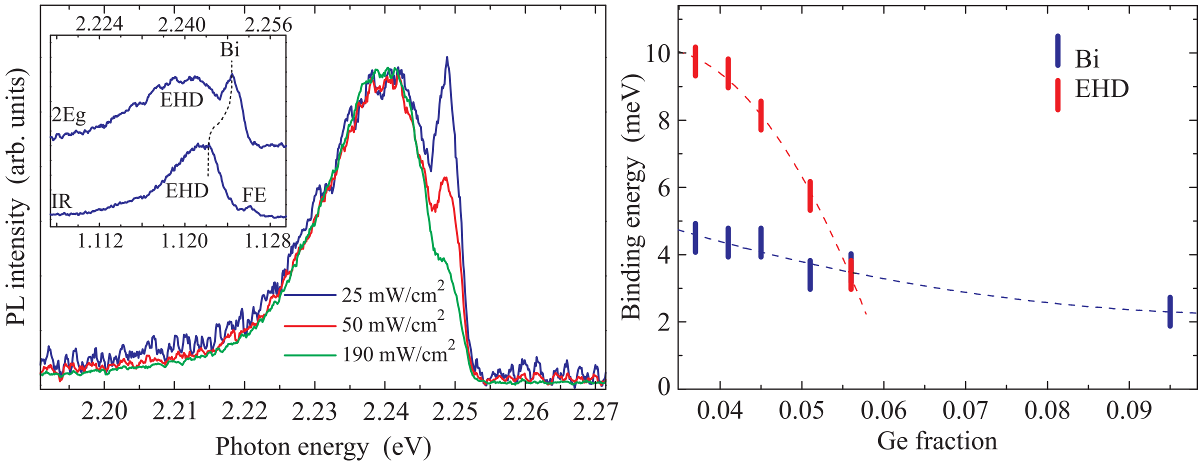

Figure 1: 2Eg-photoluminescence spectra at a temperature of 5 K, which demonstrate the coexistence of a dense electron-hole liquid and biexcitons in a shallow quantum well with a germanium content of $\sim 5\,\%$ (left). The binding energy for two electron-hole pairs for EHD and biexcitons (right)

2. The role of localization

2.1. Influence of localization and scattering of biexcitons by short-range potential on mixing of bright and dark states for 2Eg-luminescence. The random substitution of silicon atoms by germanium atoms in the layer of a quantum well leads to the appearance of a short-range potential, which leads to a violation of translational symmetry and makes the quasimomentum of one-electron, exciton, and biexciton states less well defined. From this point of view, localization is one of many mechanisms of influence (random) short-range potential on different types of electronic states. The appearance of a zero-phonon (NP) line in the IR luminescence spectra is determined by a short-range potential scattering, there is no direct relationship between the intensity of the NP line and exciton localization.

As a rule, the role of localization just turns out to be of secondary importance for SiGe/Si QWs. In particular, the intensity ratio NP/TO practically does not change with increasing temperature during transition from localized exciton states (the thermal energy kT is much smaller than the potential fluctuations σ) to delocalized ($kT \gg \sigma$). At the same time, the NP/TO ratio usually increases monotonically with increasing Ge content of the quantum well, regardless of the degree of localization of the exciton states.

Separately, we should note a weak electron-phonon coupling for exciton states in silicon-germanium heterostructures (the Frohlich mechanism is practically turned off due to the nonpolar nature of the bonds). A consequence of this is, in particular, the extremely low intensity of phonon replicas in 2Eg-luminescence spectra. Therefore, the comparable intensities of the zero-phonon (NP) line and TO-phonon replica in IR-spectra indicate that the uncertainty of the quasimomentum is much less than $2\pi/a$, where a is the lattice constant. Otherwise, a noticeable mixing of the states corresponding to the direct and indirect gap would lead to a abrupt increase in the intensity of the NP line with respect to TO phonon replica. This means that the "bright" and "dark" states of the biexcitons in the 2Eg-luminescence are well defined. This conclusion is independently confirmed by the fact that obtained 2Eg Pl amplification factor ($\sim 7/3$ at 5 K) is conserved for quantum wells with a large germanium content.

At the same time, scattering by a short-range potential and localization can lead to an noticeable mixing of states in the region of small wave vectors, thus weakening the quasimomentum conservation law. This means that the observation of 2Eg luminescence is possible for a certain range of quasimomenta (energies) of biexcitons.

2.2. Influence of localization of biexcitons on low-temperature luminescence spectra in IR and visible spectral ranges. As a rule, the effects of localization are manifested in the photoluminescence spectra in the form of shifts in the emission line with increasing temperature. Fig. 2 illustrates the spectra of 2Eg- (left) and IR- (right) photoluminescence for a quantum well with a germanium content of 12 % at bent deformation of $\sim 2 \times 10^{-4}$ along the [100] direction. In this case, the ground state of biexcitons in the quantum well turns out to be "bright". The 2Eg-spectra in Fig. 2 are recorded with an increased resolution, so the spectral width of the slit (~ 1.2 meV) does not noticeably affect the shape of the luminescence line.

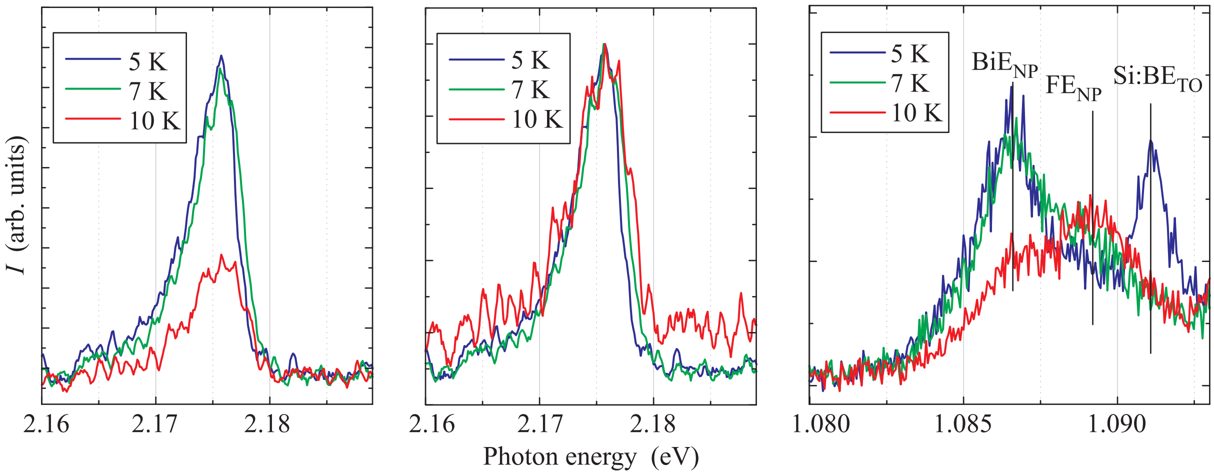

Figure 2: Temperature dependence of 2Eg-photoluminescence spectra (on the left), the same spectra normalized to the maximum intensity (at the center) and the temperature dependence of the IR photoluminescence spectra (on the right) for the sample with a germanium content of 12 % deformed $\sim 2 \times 10^{-4}$ along the direction [100]. The excitation power density is 50 mW/cm^2, the excitation wavelength is 780 nm. For the IR- and 2Eg-spectra luminescence spectra, the emission lines of biexcitons and free excitons in the quantum well are denoted by $\mathrm{BiE_{NP}}$ and $\mathrm{FE_{NP}}$, respectively. The spectral width of the slit is ∼1.2 meV for 2Eg-spectra and 0.3 meV for IR spectra

Using PL data in Fig. 2 the potential fluctuations for biexcitons can be estimated as $\sigma_{\mathrm{Bi}} \sim 1.7$ meV, which corresponds to the energy fluctuations of the exciton ground state energy $\sigma_{Ex} \sim 0.85$ meV. This value of σ_{Ex} is typical for $\mathrm{Si/Si_{1-x}Ge_x/Si}$ QWs with germanium content $x \sim 10$ %. A weak temperature shift of ~ 0.7 meV in the 2Eg-spectrum excludes its unambiguous connection only with localized states.

As one can see from Fig. 2 IR- and 2Eg-photoluminescence of biexcitons decay synchronously with increasing temperature(see Fig. 2). This means that the possible effects of localization do not significantly affect the radiative recombination probability of biexcitons with respect to 2Eg luminescence. The most probable reason for the weak dependence of the radiative recombination probability on the degree of localization is the mixing of the biexciton wave function with small wave vectors.