

Dipolar electron-hole liquid in a double-well SiGe/Si heterosystem

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Abstract. The transition from a dipolar to a spatially direct electron-hole liquid in two-dimensional layers of a type-II (*buffer* Si_{1-y}Ge_y)/*t*Si/*s*Si_{1-x}Ge_x/*t*Si/(*cap* Si_{1-y}Ge_y) heterostructure is investigated by photoluminescence spectroscopy at liquid-helium temperatures at high excitation levels. The transition takes place upon a reduction of the thickness of the *s*Si_{1-x}Ge_x layer, which forms a quantum well for holes in the valence band and a barrier in the conduction band separating the electron quantum wells (*t*Si layers). The main characteristics of both types of electron-hole liquid are determined. The lifetime of dipolar excitons is determined from photoluminescence kinetics measurements.

1. Introduction

Interest in the investigation of multiparticle interactions in low-dimensional electron-hole systems of high density is stimulated by the discovery of new condensed phases with unusual properties in these systems, in particular an electron-hole liquid (EHL) [1] and a Bose-Einstein condensate (BEC) [2–5]. Theoretical analysis indicated that these phases should exhibit superconductivity and superfluidity [6, 7]. Long-range coherence of the BEC was observed experimentally [8, 9]. These studies were carried out in contravariant (type-I) GaAs/AlGaAs heterostructures with double or wide single quantum wells (QWs) for electrons and holes. The application of an external electric field to these heterostructures leads to the appearance of a spatially indirect energy gap. Then, at low temperatures, a system of nonequilibrium spatially indirect (dipolar) excitons forms under photoexcitation. At high excitation levels, various condensed phases emerge in this system. Photoluminescence (PL) spectroscopy is the main tool for the experimental investigation of the properties of these phases.

In this study, we use the PL spectroscopy to study covariant (type-II) Si/SiGe heterostructures, where the electron-hole system should exhibit equally interesting behavior [10–12]. In contrast to GaAs/AlGaAs structures, a spatially indirect electron-hole system can be created in SiGe structures without application of an external electric field. Lifetimes of nonequilibrium charge carriers in silicon, which is an indirect-gap material, are long (more than three orders of magnitude longer than those in GaAs), and this makes it possible to attain high concentrations of charge carriers and investigate their interactions leading to the formation of multiparticle states even at low excitation levels.

Here, we investigate the spectrum of multiparticle excited states in the exciton system in a double-QW SiGe/Si heterostructure in a wide range of excitation levels and temperatures. We study the transformation of the spectrum upon the transition from a dipolar to a spatially direct electron-hole



system. We demonstrate that such a transition takes place as the thickness of the barrier layer separating the two QWs is reduced. We determine the main characteristics of both spatially direct and indirect EHL. Using time-resolved PL spectroscopy, we measure the lifetime of dipolar excitons.

2. Experimental techniques

A system of dipolar excitons was implemented in a $\text{Si}_{1-y}\text{Ge}_y/t\text{Si}/s\text{Si}_{1-x}\text{Ge}_x/t\text{Si}/\text{Si}_{1-y}\text{Ge}_y$ ($x > y$) heterostructure with two QWs for electrons, formed by tensile strained $t\text{Si}$ layers separated by a compressively strained $s\text{Si}_{1-x}\text{Ge}_x$ layer acting as a barrier for electrons and a QW for holes. The heterostructure was grown by molecular-beam epitaxy on a single-crystal $\text{Si}_{0.92}\text{Ge}_{0.08}$ (001) substrate. The thickness of the pseudomorphic buffer layer of the same composition as the substrate was 100 nm. The thickness of the pseudomorphically grown strained $t\text{Si}$ layers was 4 nm. The thickness d of the electron-barrier strained $s\text{SiGe}$ was 4 or 2 nm (structures $(4 \times 4 \times 4)$ and $(4 \times 2 \times 4)$, respectively). A cap layer of the same composition as the substrate completed the design of this structure with two electron QWs.

The energy spectrum of the structures under study was investigated by PL spectroscopy in the near-infrared spectral range. Measurements were carried out at temperatures $T = 1.8\text{--}60\text{ K}$ and excitation levels $P = 0.01\text{--}300\text{ W}\cdot\text{cm}^{-2}$. Quasi-continuous excitation of the structure was performed by a semiconductor laser emitting at a wavelength of $\lambda = 405\text{ nm}$. Recombination radiation was detected by a germanium $p\text{-i-n}$ photodiode.

Analyzing the PL spectra of the structures with different barrier thicknesses ($d = 4$ or 2 nm), we can trace the transition from a dipolar to a spatially direct electron-hole system.

3. Energy-band diagram and the scheme of optical transitions

Figure 1 shows schematically the energy-band diagram of the heterostructures under study. In the first tensile-strained $t\text{Si}$ layer grown on top of the unstrained $\text{Si}_{1-y}\text{Ge}_y$ (001) substrate, the sixfold degeneracy of the conduction-band bottom characteristic of unstrained Si is lifted. The bottom of the conduction band in this layer is formed by two Δ_2 valley located at the $\langle 001 \rangle$ axes of the reciprocal lattice that are perpendicular to the layer plane. The four Δ_4 minima for which the axes of the isoenergy surfaces are perpendicular to the growth direction appear at a higher energy. The compressively strained $s\text{Si}_{1-x}\text{Ge}_x$, grown on top of the first $t\text{Si}$ layer, acts as a barrier for electrons and a well for holes. The degeneracy of the valence-band edge in the $s\text{Si}_{1-x}\text{Ge}_x$ layer is also lifted; the top of the valence band is formed by the quantum-confined states of heavy holes (hh) in the QW. The ground exciton state in such a system is formed by electrons from the Δ_2 valley of the Si conduction band, which occupy the quantum-confinement level in the QW formed by the $t\text{Si}$ layer, and heavy holes occupying the quantum-confinement level in the QW formed in the valence band of the $s\text{Si}_{1-x}\text{Ge}_x$ layer. The second pseudomorphically grown $t\text{Si}$ layer and the cap $\text{Si}_{1-y}\text{Ge}_y$ layer with the same composition as the substrate completes the layout of this structure with two electron QWs.

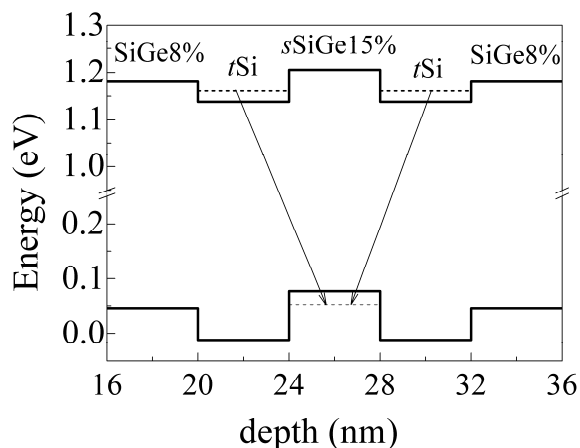


Figure 1. Energy-band diagram and the scheme of optical transitions.

4. PL spectra of the EHL and excitons in dipolar and spatially direct electron-hole systems

Figure 2 shows a series of the PL spectra of the (4×4×4) structure (the thickness of the *s*SiGe barrier layer $d = 4$ nm) measured at $T = 1.8$ K upon an increase in the excitation level P .

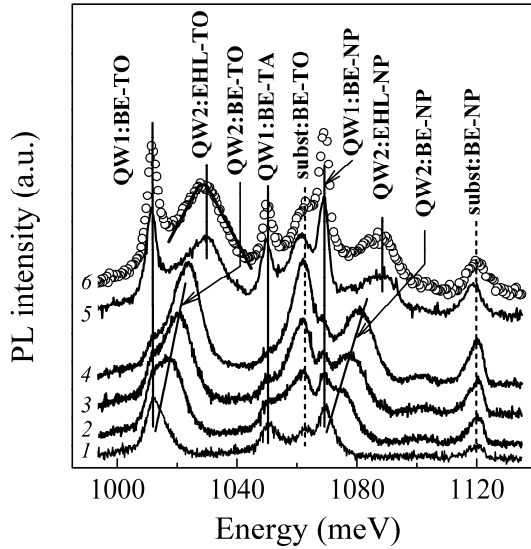


Figure 2. The formation of a dipolar EHL in the (4×4×4) double-QW structure with an increase in the pump level $P = (1)$ 0.7, (2) 1.0, (3) 4.5, (4) 8.0, (5) 15, and (6) 90 $\text{W}\cdot\text{cm}^{-2}$; $T = 1.8$ K. The designations of the PL lines are explained in the text. For spectrum 6, the solid line shows the calculated EHL line shape.

Dashed vertical lines mark the PL lines originating from the recombination of bound excitons (BE) in the substrate and the buffer and cap layers; the corresponding no-phonon line and its transverse-optical (TO) phonon replica are designated as (subst: BE-NP) and (subst: BE-TO), respectively. Solid vertical lines mark the PL lines related to the *t*Si – *s*SiGe transitions (figure 1). As the excitation level increases, the lines from the second QW (QW2:BE-NP and QW2:BE-TO) experience a blue shift as a result of Coulomb band bending caused by the accumulation of spatially indirect (dipolar) excitons collected from the substrate. Apparently, only the lower QW for electrons participates in the collection of excitons produced by photoexcitation from the substrate. This makes it possible to distinguish the radiative transitions from the *lower t*Si to the *s*SiGe layer (lines QW2, which experience a blue shift at high excitation levels) and from the *upper t*Si to the *s*SiGe layer (lines QW1: BE-NP, BE-TA (transverse-acoustic phonon replica), and BE-TO, which do not experience a blue shift). At low temperatures and high excitation levels, we observe in this sample a two-dimensional dipolar EHL with spatially separated electrons and holes, manifested in the emergence of the PL lines QW2: EHL-NP and EHL-TO. The shape of these lines does not vary with increasing excitation level, which is a signature of the EHL. In addition, we see that, for $P > 15 \text{ W}\cdot\text{cm}^{-2}$, the blue shift ceases. This takes place because the particle density in the EHL is constant and only the size and concentration of the EHL drops increase with increasing excitation level. We found that the critical density of the observed EHL is about 2 K.

Figure 3 shows a series of the PL spectra of the (4×2×4) structure (the thickness of the *s*SiGe barrier layer $d = 2$ nm) measured at $T = 1.8$ K upon an increase in the excitation level.

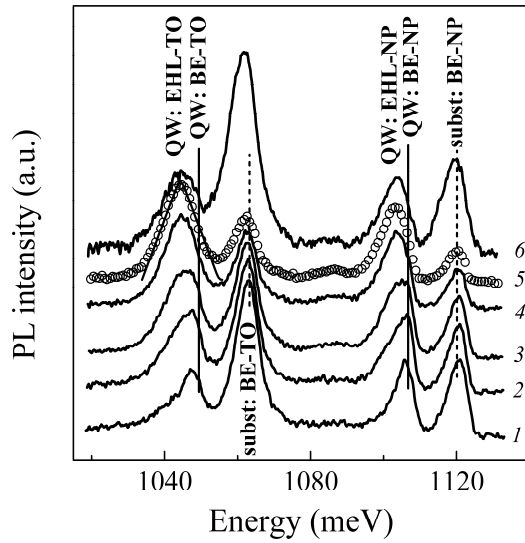


Figure 3. Formation of a spatially direct EHL in the (4×2×4) with an increase in the pump level $P = (1)$ 0.09, (2) 0.4, (3) 0.75, (4) 1.0, (5) 8, and (6) 80 $\text{W}\cdot\text{cm}^{-2}$, $T = 1.8$ K.

The designations of the PL lines are explained in the text. For spectrum 5, the solid line shows the calculated EHL line shape.

Dashed vertical lines mark the PL lines originating from the substrate and the buffer and cap layers. Solid vertical lines mark the PL lines related to the $t\text{Si} - s\text{SiGe}$ transitions. In contrast to the previous case, no blue shift occurs in these spectra with increasing excitation level. The wave function of electrons spreads into the thin $s\text{SiGe}$ barrier, so that the heterostructure becomes effectively type-I. The EHL line appears with increasing excitation level at the red tail of the exciton line. For $P > 1 \text{ W}\cdot\text{cm}^{-2}$, the line shape becomes independent of the excitation level, which gives evidence of the EHL formation. The EHL is formed by Δ_2 electrons and heavy holes. Thus, in this structure we observe PL from a spatially direct EHL. The critical temperature was found to be 6 K.

We note that the critical temperatures of the spatially direct and dipolar EHL in double-well heterostructures with tensile strained $t\text{Si}$ layers is considerably lower than the corresponding critical temperatures of the EHL in $\text{Si}/s\text{SiGe}/\text{Si}$ structures [10, 12]. We believe that the reduction in the critical temperatures of the EHL of both types is caused by a twofold decrease in the degree of degeneracy of the electron spectrum in $t\text{Si}$ layers of $\text{Si}_{1-y}\text{Ge}_y/t\text{Si}/s\text{Si}_{1-x}\text{Ge}_x/t\text{Si}/\text{Si}_{1-y}\text{Ge}_y$ heterostructures as compared to Si layers in $\text{Si}/s\text{SiGe}/\text{Si}(001)$ heterostructures.

In curve 6 (figure 2) and curve 5 (figure 3), circles show the experimental results and solid lines show the results of calculations of the EHL line shape. The calculations were carried out similarly to the analysis given in [13] for bulk semiconductors. In contrast to [13], we used an energy-independent density of states for electrons and holes. The two-dimensional density of holes in the EHL in the QW formed by the $s\text{SiGe}$ layer obtained from the calculations is $1.2 \cdot 10^{12}$ and $2.2 \cdot 10^{12} \text{ cm}^{-2}$ for the (4×2×4) and (4×4×4) structures, respectively.

One can see from figure 2 and figure 3 that the intensity of the no-phonon exciton and EHL recombination lines is comparable to or even higher than the intensity of their phonon replicas. Apparently, the high intensity of the NP components of the PL spectrum in the structures under study is related to the localization of dipolar excitons and electron-hole drops in lateral wells of the random potential profile resulting from the composition inhomogeneities in the SiGe layer and at heterointerfaces [14, 15].

4.1. Temporal characteristics of photoluminescence

Figure 4 shows the time evolution of the PL spectra of the dipolar exciton system in the (4×4×4) heterostructure measured at a temperature of 5 K (higher than the critical temperature of the dipolar EHL). The spectra in figure 4 are measured upon a stepwise increase in the delay time with respect to the excitation pulse. The evolution of the spectra with increasing delay time manifests the phase transition between the dipolar electron-hole plasma and the gas of dipolar excitons. From the PL decay

kinetics, it was found that the lifetime of dipolar excitons in this structure is $6.3 \mu\text{s}$. This is considerably longer than the lifetime of spatially direct excitons ($<1 \mu\text{s}$ in Si/sSiGe/Si structures [16]).

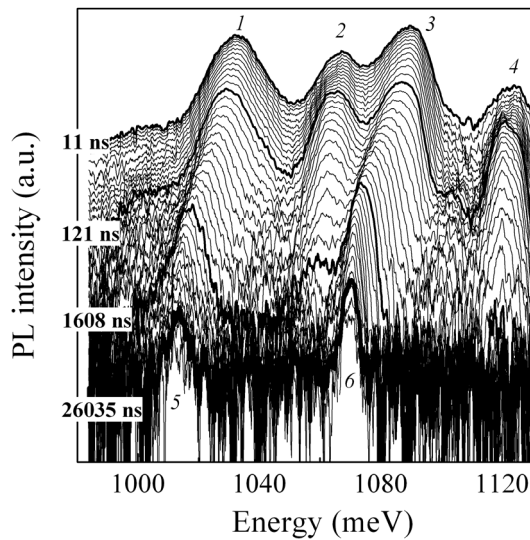


Figure 4. PL spectra of the (4×4×4) structure at $T = 5 \text{ K}$ at different delay times after the excitation pulse. Numbers indicate the PL lines (1) QW2:EHP-TO, (2) subst:BE-TO, (3) QW2:EHP-NP, (4) subst:BE-NP, (5) QW2:BE-TO, and (6) – QW2:BE-NP (see figure 2 and explanations in the text).

5. Conclusions

To summarize, the following results have been obtained.

$\text{Si}_{1-y}\text{Ge}_y/\text{tSi}/\text{sSi}_{1-x}\text{Ge}_x/\text{tSi}/\text{Si}_{1-y}\text{Ge}_y$ heterostructures with two QWs for electrons formed by tensile strained tSi separated by $\text{Si}_{1-x}\text{Ge}_x$ (acting as a barrier for electrons and a well for holes) have been designed and grown.

Using PL spectroscopy, we have observed the formation of two-dimensional EHL in these structures at liquid-helium temperatures and high excitation levels. Either a spatially direct or a dipolar EHL forms depending on the design of the structure. The two-dimensional density of holes is $2.2 \cdot 10^{12}$ and $1.2 \cdot 10^{12} \text{ cm}^{-2}$ for dipolar and spatially direct EHL, respectively. The critical temperatures of spatially direct and dipolar EHL have been determined to be 6 and 2 K, respectively.

The transformation of the spectrum of the excited states upon the transition from a dipolar to a spatially direct electron-hole system has been investigated. This transition takes place as the thickness of the barrier layer separating the two QWs is reduced from 4 to 2 nm.

The lifetime of dipolar excitons has been determined to be $6.3 \mu\text{s}$.

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