St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 2026. Vol. 19. No. 1.1
Научно-технические ведомости СПбГПУ. Физико-математические науки. 19 (1.1) 2026

Conference paper
UDC 538.958
DOI: https://doi.org/10.18721/JPM.191.107

Temperature-dependent infrared photoluminescence study of GeSn/Si and GeSiSn/Si multiple quantum wells

I.V. Chumanov [1] ✉, D.D. Firsov [1], D.V. Kolyada [1], O.S. Komkov [1], I.V. Skvortsov [2], V.I. Mashanov [2], I.D. Loshkarev [2], V.A. Timofeev [2]

[1] St. Petersburg Electrotechnical University "LETI", St. Petersburg, Russia;
[2] Rzhanov Institute of Semiconductor Physics Siberian Branch of RAS, Novosibirsk, Russia
✉ chumanov2000@yandex.ru

Abstract. The study of the luminescence properties of multiple $Ge_{0.53}Sn_{0.47}/Si$ and $Ge_{0.3}Si_{0.56}Sn_{0.14}/Si$ quantum wells allowed the energies of interband transitions between the electron Δ subband in silicon and the hole level in the quantum well to be estimated. Temperature studies revealed an initial increase in photoluminescence intensity between 8 and 30 K, followed by a decrease at higher temperatures. Approximation of the obtained results allowed the activation energies of the processes responsible for the temperature quenching of luminescence to be determined.

Keywords: photoluminescence, multiple quantum wells, germanium, silicon, tin, semiconductors, solid solutions, FTIR spectroscopy

Funding: The study was funded by the Russian Science Foundation grant № 25-19-00742.

Citation: Chumanov I.V., Firsov D.D., Kolyada D.V., Komkov O.S., Skvortsov I.V., Mashanov V.I., Loshkarev I.D., Timofeev V.A., Temperature-dependent infrared photoluminescence study of GeSn/Si and GeSiSn/Si multiple quantum wells, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 19 (1.1) (2026) 45–50. DOI: https://doi.org/10.18721/JPM.191.107

Конференционная статья
УДК 538.958
DOI: https://doi.org/10.18721/JPM.191.107

Исследование температурной зависимости инфракрасной фотолюминесценции множественных квантовых ям GeSn/Si и GeSiSn/Si

И.В. Чуманов [1] ✉, Д.Д. Фирсов [1], Д.В. Коляда [1], О.С. Комков [1], И.В. Скворцов [2], В.И. Машанов [2], И.Д. Лошкарев [2], В.А. Тимофеев [2]

[1] Санкт-Петербургский государственный электротехнический университет «ЛЭТИ» им. В.И. Ульянова (Ленина), Санкт-Петербург, Россия;
[2] Институт физики полупроводников им. А.В. Ржанова Сибирского отделения РАН, г. Новосибирск, Россия
✉ chumanov2000@yandex.ru

Аннотация. Изучение люминесцентных свойств множественных квантовых ям $Ge_{0.53}Sn_{0.47}/Si$ и $Ge_{0.3}Si_{0.56}Sn_{0.14}/Si$ позволило оценить энергии межзонных переходов между электронной Δ-подзоной в кремнии и дырочным уровнем в квантовой яме. Температурные исследования выявили изначальный рост интенсивности фотолюминесценции от 8 до 30 K, а затем – её снижение при дальнейшем нагреве. Аппроксимация полученных результатов позволила определить энергии активации процессов, ответственных за температурное гашение люминесценции.

Ключевые слова: фотолюминесценция, множественные квантовые ямы, германий, кремний, олово, полупроводники, твердые растворы, Фурье-спектроскопия

Финансирование: Исследование выполнено за счет гранта Российского научного фонда № 25-19-00742.

Ссылка при цитировании: Чуманов И.В., Фирсов Д.Д., Коляда Д.В., Комков О.С., Скворцов И.В., Машанов В.И., Лошкарев И.Д., Тимофеев В.А. Исследование температурной зависимости инфракрасной фотолюминесценции множественных квантовых ям GeSn/Si и GeSiSn/Si // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2026. Т. 19. № 1.1. С. 45–50. DOI: https://doi.org/10.18721/JPM.191.107

Introduction

Semiconductor materials based on Group IV elements of the Periodic Table are actively studied in modern micro- and nanoelectronics. Solid solutions of silicon, germanium, and tin are of particular interest. Their key advantage is in compatibility with modern CMOS technology, paving the way for the creation of new optoelectronic devices. Such devices can operate effectively in a wide spectral range, including near (1.5–3 μm), mid (3–8 μm), and long-wavelength (8–14 μm) infrared. This makes them promising for use in optical communication lines, gas analysis, environmental monitoring systems and other applications [1].

One such material is the $Ge_{1-x}Sn_x$ solid solution, which, at a certain Sn content over 6–10 at. % for unstrained layers, exhibits a direct band gap structure [2, 3]. The introduction of additional strains can lead to a shift in the boundary of the direct band gap for tin, either upward or downward [4, 5]. Optoelectronic devices operating in the near and mid-infrared ranges have already been demonstrated using this solid solution [6, 7].

The growth technology for this material faces a number of challenges that affect the achievement of high-quality layers. The significant difference in the lattice parameters of Sn and Ge (Si) leads to low tin solubility (< 1% in Ge and < 0.1% in Si) and other undesirable processes, such as tin segregation during structure growth, defect formation, and elastic mechanical stresses in the layers [8].

A new stage in the development of this area is the study of ternary $Ge_{1-x-y}Si_xSn_y$ solid solutions, in which independent control of the band gap and lattice parameter becomes possible by adjusting the composition. The possibility of forming thin layers of these materials, forming multiple quantum wells (MQWs), has previously been demonstrated [9].

This paper presents the results of a photoluminescence (PL) study of multiple GeSn and GeSiSn quantum wells with an increased tin content and silicon barriers.

Materials and Methods

The studied samples were grown using molecular beam epitaxy at the Institute of Semiconductor Physics. Initially, a 150 nm-thick silicon buffer layer was formed on silicon (100) wafers. Ten periods of multiple quantum wells were then formed on top of this layer: $Ge_{0.3}Si_{0.56}Sn_{0.14}$/Si in the first sample and $Ge_{0.53}Sn_{0.47}$/Si in the second sample. To minimize undesirable tin segregation, silicon barriers were grown in two stages: at 150 °C, the well layer was coated with 2 nm of silicon, and at 500 °C, the layer thickness was further increased by 5 nm. Upon completion of the structure growth, a 10 nm-thick silicon capping layer was formed.

In addition to differences in the composition of the QW layer, the samples differ in the nominal well thickness and the growth temperature. For the first sample, the $Ge_{0.3}Si_{0.56}Sn_{0.14}$ layer thickness was 1 nm, and the growth temperature was 150 °C. The well layer of the second sample has a thickness of 0.17 nm (~1 monolayer), but it was grown at room temperature. The samples were also subjected to high-temperature annealing in a quartz furnace under an argon atmosphere. Annealing of each structure was performed for several samples at different temperatures

(600, 625, 650, 675, and 700 °C) for 10 minutes. The samples that demonstrated the most intense photoluminescence signal were selected for the studies: $Ge_{0.3}Si_{0.56}Sn_{0.14}$/Si after annealing at 625 °C and $Ge_{0.53}Sn_{0.47}$/Si at a temperature of 650 °C.

Photoluminescence spectra were recorded on a Vertex 80 FTIR spectrometer equipped with a liquid-nitrogen-cooled InSb photovoltaic detector. Luminescence was excited by a 405 nm diode laser with a power of up to 140 mW. Samples were cooled in a Janis CCS-150 closed-cycle helium cryostat, capable of achieving temperatures as low as 8 K. Luminescence signal recording was performed using gated integration with short laser pulses [10]. This approach minimized sample surface heating during the study.

Results and Discussion

At low temperatures, the $Ge_{0.53}Sn_{0.47}$/Si and $Ge_{0.3}Si_{0.56}Sn_{0.14}$/Si MQW samples exhibit an intense photoluminescence signal in the energy range of ~0.87 eV and ~0.76 eV, respectively. Fig. 1 shows the PL spectra for each sample, measured at 8 K. The shape of the spectra indicates the presence of several radiative recombination processes. The nature of the most intense high-energy peak is explained by interband optical transitions between the electron Δ subband in silicon and the hole level in the QW layer of the solid solution. This was further confirmed by calculating the band diagram for quantum wells of similar composition [11]. In the lower energy region, a broad shoulder is observed, which is explained by the luminescence signal from defects in the crystal structure of the sample [12].

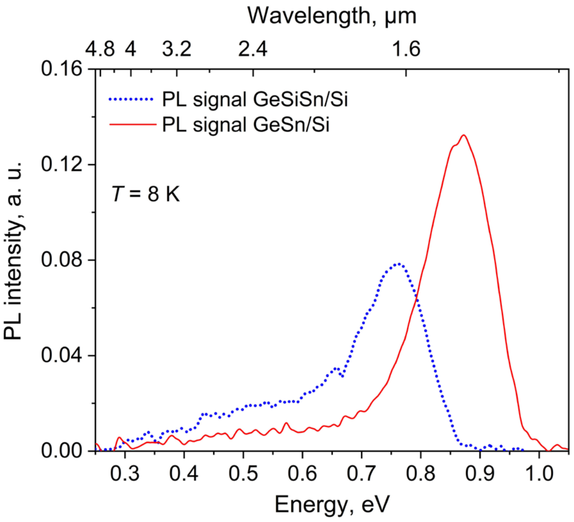

Fig. 1. Comparison of PL spectra of $Ge_{0.53}Sn_{0.47}$/Si and $Ge_{0.3}Si_{0.56}Sn_{0.14}$/Si MQW samples at 8 K

A comparison of the spectra of the samples shown in Fig. 1 shows that the sample with the $Ge_{0.53}Sn_{0.47}$/Si MQW has a higher ratio of the PL signal from interband transitions to the signal in the defect region. At the same time, in the sample with the $Ge_{0.3}Si_{0.56}Sn_{0.14}$/Si MQW, defects produce more intense luminescence, while the signal from optical transitions involving QWs is noticeably weaker.

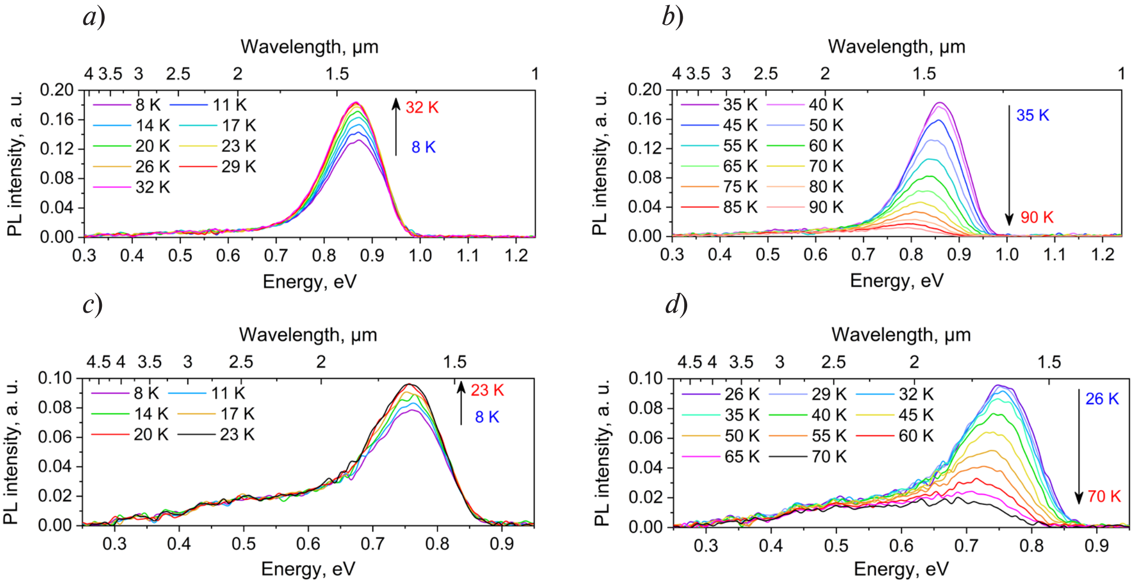
a)
b)
c)
d)

Fig. 2. PL spectra of $Ge_{0.53}Sn_{0.47}$/Si (*a*, *b*) and $Ge_{0.3}Si_{0.56}Sn_{0.14}$/Si (*c*, *d*) MQW samples at different temperatures

Arrows show the shift of the PL peak with increasing temperature for clarity

A comparison of the PL spectra at different temperatures for the sample with the $Ge_{0.53}Sn_{0.47}$/Si MQW (Fig. 2, *a*, *b*) reveals that the PL signal intensity increases with increasing temperature up to 29–32 K and then decreases, starting from 35 K. The second sample, containing the $Ge_{0.3}Si_{0.56}Sn_{0.14}$/Si MQW, exhibits similar behavior with increasing temperature (see Fig. 2, *c*, *d*). This behavior can presumably be explained by a combination of several factors. As the sample heats, the number of phonons required for the observed (indirect) radiative transitions increases. Another process may be the thermal activation of photogenerated charge carriers trapped by defect levels in the band gap. Thus, up to a certain critical temperature, an enhancement of the interband transition signal is observed due to the release of trapped charge carriers [13].

With increasing temperature, both samples exhibit a ‘red’ shift in peak position, which is typical for materials of this class. Fig. 3, *a* shows the temperature dependence of the transition energy for each sample.

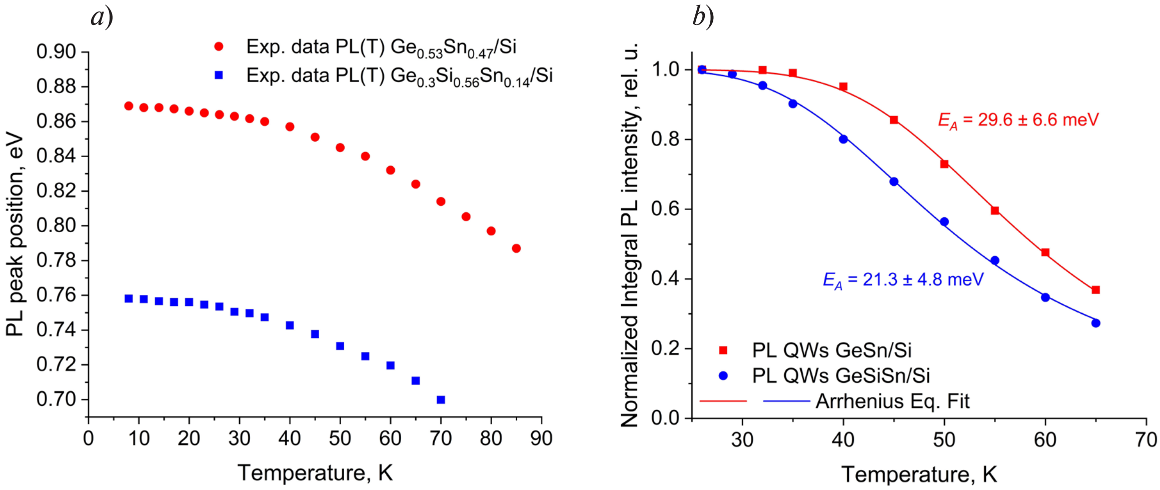

Fig. 3. Temperature dependences of the peak position (*a*) and the integral of the PL intensity (*b*)

It should be noted that the ‘red’ shift for the studied samples reaches ~60 meV and ~80 meV with temperature increases of 62 K and 72 K, respectively (see Fig. 3, *a*). We suggest that the strong temperature dependence in the low-temperature region may be due to charge carriers localized in the minima of the spatial inhomogeneity of multiple quantum wells [9].

Fig. 3, *b* shows the temperature dependences of the integrated photoluminescence intensity of the interband transition. An approximation was performed for each sample using the Arrhenius equation:

$$I = \frac{I_0}{1 + C\exp\left(-\frac{E_A}{k_B T}\right)}, \qquad (1)$$

where I_0 is the intensity at a temperature of 0 K, C is the ratio of radiative to nonradiative carrier lifetimes, and E_A is the activation energy of the process that leads to quenching of photoluminescence.

The approximation used allowed us to determine the activation energies of the nonradiative process leading to luminescence decay in the structures with increasing temperature, and obtain the C coefficients corresponding to the relation between the nonradiative and radiative recombination lifetimes. The value of the C coefficient is approximately three times higher for the sample with GeSn layers. With increasing temperature, this is manifested in a steeper slope of the dependence and a rapid quenching of luminescence (see Fig. 3, *b*). The values of the obtained energies are significantly (an order of magnitude) lower than the energy of thermal emission of holes from the quantum well. Excluding Auger recombination mechanisms, since for thin QWs they are of a thresholdless and quasi-thresholdless nature [14], the most probable scenario for the temperature quenching of PL is presumably the Shockley–Read–Hall (SRH) mechanism.

Conclusion

This paper presents the results of a study of the temperature dependence of photoluminescence in $Ge_{0.53}Sn_{0.47}/Si$ and $Ge_{0.3}Si_{0.56}Sn_{0.14}/Si$ samples with MQWs, measured by FTIR spectroscopy using gated integration. The spectra contained a signal from interband transitions involving hole levels in the QWs and an electron Δ subband in the silicon barriers. Luminescence from defects in the sample structure was also observed. The interband PL intensity initially increases with sample heating, which is explained by the increase in the phonon number and the thermal activation of charge carriers localized at defect levels in the band gap. Further heating of the sample leads to a decrease in the PL intensity, which we associate with the process of nonradiative recombination by the SRH mechanism; the activation energy of the process was ~21 and ~30 meV for samples with MQW $Ge_{0.3}Si_{0.56}Sn_{0.14}/Si$ and $Ge_{0.53}Sn_{0.47}/Si$, respectively. In conclusion, we note that the sample with GeSn layers demonstrates better temperature stability of luminescence due to the higher activation energy.

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THE AUTHORS

CHUMANOV Ivan V.
chumanov2000@yandex.ru
ORCID: 0009-0009-2564-6100

FIRSOV Dmitrii D.
d.d.firsov@gmail.com
ORCID: 0000-0001-7608-9580

KOLYADA Dmitry V.
kolyada.dima94@mail.ru

KOMKOV Oleg S.
oleg_sergeevich@mail.ru
ORCID: 0000-0002-8999-1175

SKVORTSOV Ilya V
i.skvortsov@isp.nsc.ru
ORCID: 0000-0002-2153-1615

MASHANOV Vladimir I.
mash@isp.nsc.ru
ORCID: 0000-0003-4420-6695

LOSHKAREV Ivan D.
idl@isp.nsc.ru
ORCID: 0000-0003-4771-3705

TIMOFEEV Vyacheslav A.
Vyacheslav.t@isp.nsc.ru
ORCID: 0000-0003-4093-7802

Received 15.12.2025. Approved after reviewing 18.02.2026. Accepted 27.02.2026.