Luminescent response of GeSi structures grown from ion-molecular beams as a function of laser excitation power

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The effect of laser excitation power on the luminescence properties of Ge/Si quantum dot (QD) structures produced by molecular beam epitaxy irradiated with 2 keV Ge^+ ions was studied. With increasing power, a shift of the GeSi QD emission peak by $\sim$ 10 meV was observed in the photoluminescence spectra of structures grown without ion irradiation. For structures grown under ion irradiation, the peak position remains unchanged, which is due to the appearance of an effective channel for radiative recombination through defects created by ion irradiation in GeSi QDs. It was found that the dependence of the photoluminescence intensity on the laser excitation power for ion-modified structures becomes linear above a threshold power of $\sim 35/cm^2$, which is a characteristic sign of the presence of direct optical transitions.

Keywords: quantum dots, germanium, silicon, epitaxy, ion irradiation, photoluminescence.

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1. Introduction

Today, the major focus in the fundamental research and development of the micro- and nanoelectronics semiconductor materials is made on the nanostructured silicon technology systems [1–4]. Among the promising areas is integrated silicon-based photonics, characterized by a wide range of potential applications, ranging from creation of electronic devices, telecommunications and ending with optical quantum systems [5–10]. The major issue in creating highly efficient radiation silicon-based sources is related to its indirect-gap structure, which leads to a low probability of radiative recombination. Among many proposals for solving this problem, the idea of using a Ge/Si heterosystem with quantum dots (QDs) stands out [11–13]. This system fits naturally into silicon production cycles, and the uncertainty in momentum should increase the probability of radiative recombination. However, the studies conducted have shown that epitaxial structures with GeSi QDs do not provide a noticeable gain in the rate of radiative recombination. Today, the use of ion irradiation in the process of forming quantum dots is considered as one of the ways to solve this problem [5,6,14–21]. It has been proposed to directly expose quantum dots to germanium ions during epitaxial growth. The promise of this approach has been confirmed by recent experiments investigating the luminescent properties of structures with quantum dots modified by ion irradiation during growth [5,6,15–18]. Experiments on fabrication of GeSi nanostructures incorporating clusters of optically active defects [22], using ion implantation of germanium into silicon and subsequent thermal annealing, also support the use of ion irradiation. These structures demonstrated high thermal stability of photoluminescence (PL) compared to epitaxial structures with GeSi QDs obtained without ion irradiation. Similar structures with GeSi QDs were created in Linz University [5,6,16,17] using ion irradiation in the process of molecular beam epitaxy (MBE). The authors of the mentioned studies suggested that the created structures with GeSi QDs contain interstitial point defects ([110]-split interstitials [6]), which provide direct interband optical transitions and, accordingly, high efficiency of radiative recombination. By using GeSi QDs with defects as an optically active medium embedded in a silicon disk microresonator, the authors demonstrated the laser generation effect [5]. Despite a significant amount of work on the creation of light-emitting GeSi nanostructures using ion irradiation [5,6,16–18,22], questions related to understanding the mechanisms of radiative recombination in such structures remain open.

In this study, the dependence of the photoluminescence intensity on the laser excitation power for structures with GeSi QDs grown from ion-molecular beams was studied in detail. The obtained results indicate an increase in the rate of radiative recombination due to direct interband optical transitions involving defects caused by ion irradiation.

2. Methods and experimental details

The structures with GeSi QDs on Si(100) substrates with ion irradiation and without it were grown in the molecular-beam epitaxy setup SIVA-21 by Riber. Si(100) substrates were loaded into the setup chamber after a standard chemical cleaning procedure. The completion of the substrate surface cleaning procedure was removal of SiO_2 protective layer at a temperature of 750 °C in a weak silicon flow. Next, a buffer layer of Si (~ 100 nm) was deposited on the substrate with a gradual increase in the growth temperature from 500 to 600 °C. A multilayer structure consisting of 10 layers of GeSi QDs covered with silicon was grown on the surface of the buffer layer. The QD stack was formed by sequential deposition of germanium layers (each layer with a thickness of ~ 0.8 nm) at a temperature of 500 °C, alternating with silicon layers with a thickness of ~ 15 nm. Silicon layers were grown at a temperature that was gradually increased from 500 to 600 °C. The final, covering layer of silicon was 40 nm. Electron beam evaporators were used as sources of silicon and germanium, which ensured partial ionization of the streams of atoms emitted from the targets. The ionization coefficient was $\sim 3 \cdot 10^{-4}$, which was determined by the parameters of the electron beam evaporators used. Two structures were fabricated, in one of which (structure 1) an accelerating potential was applied to the substrate during germanium deposition stage, providing a germanium ion energy of $E \approx 2$ keV, and in the other (structure 2) germanium deposition was carried out without applying the potential. This approach made it possible to implement irradiation with germanium ions during the formation of quantum dots. The method of growth of structures is described in detail in [18]. The choice of ion energy depended on two factors. When the energy is 2 keV, the projective range of germanium ions is ~ 5 nm, which is just right for generating defects in the surface layer. We also took into account the findings from [6], where the dependence of PL enhancement effect on the energy of germanium ions was investigated. According to these data, the maximum effect was observed in the energy range of 1.5–2 keV.

The surface morphology of the formed structures was studied using atomic force microscopy (AFM). The photoluminescence (PL) studies of Ge/Si nanostructures were carried out on a spectroscopic complex based on a diffraction monochromator MDR-23U, providing a reciprocal linear dispersion of 26 Å/mm. A solid-state laser operating at a wavelength of 405 nm was used as the source of exciting radiation. Optical signals were detected by a Ge $p-i-n$ detector of EO-817H type, cooled to a temperature of ~ 80 K. The measurements were carried out at 78 K using a dedicated optical cryostat model ARS CS202E-DMX. The laser pumping level was varied in the range from 0.5 to 60 W/cm^2.

The defects formation in Ge/Si heterostructure under irradiation with germanium ions was simulated by molecular dynamics (MD) method [19]. The model structure included 30 silicon monolayers located in the (100) plane, covered by 8 semilayers of germanium. The atoms of the surface monolayer were artificially arranged into dimer rows corresponding to the surface superstructure (001)-2×1. The structure had lateral dimensions 17.4×17.4 nm^2. In the plane of the substrate, periodic boundary conditions were imposed on the structure. The interaction between atoms was determined by empirical Tersoff potential [23]. To obtain the initial equilibrium configuration, the system went through a relaxation procedure. In 2 picoseconds, the temperature rose from 10 to 773 K, and a dynamic problem was solved for the heated system, during which the atoms of the heterosystem occupied equilibrium positions. To simulate ion exposure, an atom of Ge was added to the system at a height of 10 nm with an initial velocity directed vertically to the substrate, corresponding to a kinetic energy of 2 keV.

3. Experimental results and discussion

According to AFM data, 3D germanium islands had a shape of *hut*-clusters $h \approx 2-2.5$ nm high and lateral dimensions $L_1 \approx 20-25$ nm and $L_2 \approx 40-50$ nm. The density of islands in structures formed during ion irradiation was ~ 1.2 times higher compared to structures grown without ion irradiation and amounted to $\sim 1.2 \cdot 10^{11}$ cm^{-2}. The obtained results are consistent with the data in [20,21], where pulsed ion irradiation was described to be used in MBE process. In the mentioned studies it was shown that irradiation with Ge^+ ions during the epitaxy of Ge on Si substrates leads to higher density of three-dimensional islands, lower sizes and a narrower size spread. It was hypothesized that interstitial defects formed near the surface by ion irradiation create centers of preferential nucleation of islands.

The effect of ion irradiation is observed not only in changes in surface morphology, but also in the luminescent properties of the studied structures. Figure 1 shows the PL spectra as a function of the exciting laser radiation power (P), measured at a temperature of 78 K for structures with GeSi QDs obtained by MBE method with ion irradiation (Figure 1, *b*, structure 2) and without it (Figure 1, *a*, structure 1). For both cases, broad PL bands are observed in the spectral region of 0.7–0.9 eV, associated with QD radiation. The measurement results showed that PL band intensity for structure 2 is ~ 3 times higher than the band intensity for structure 1 at all laser pump powers. At $P = 0.5$ W/cm^2 the maxima of the spectral position of PL peaks are at 0.8 and 0.775 eV for structure 1 and structure 2, respectively. For structures grown without ion irradiation, the position of the maximum radiation intensity shifts towards higher energies with an increase in the laser pumping level from 0.5 to 60 W/cm^2 (Figure 1, *a*). The magnitude of the shift is 10 meV. Whereas for structures with QDs created under ion irradiation, the position of the maximum radiation

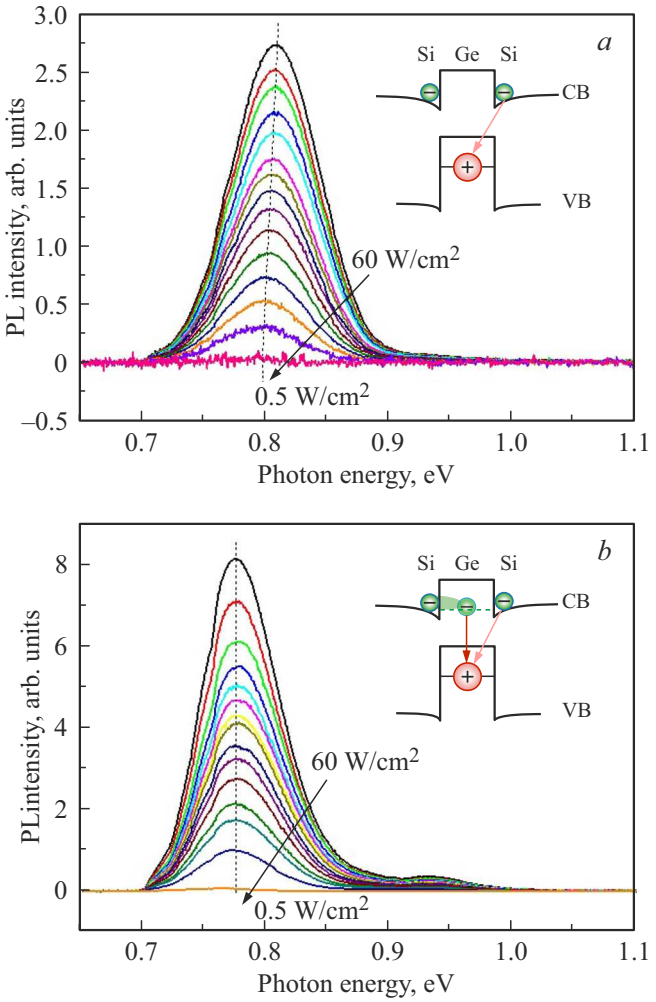

Figure 1. The PL spectra at different powers of laser pumping: *a* — MBE; *b* — MBE with irradiation by Ge^+ ions with energy ~ 2 keV. The insets to the figure show band diagrams with optical transitions in a GeSi quantum dot with a defect (see inset in Figure 1, *b*) and without a defect (see inset in Figure 1, *a*). The spectra were measured at 78 K.

intensity at 0.775 eV remains unchanged (Figure 1, *b*). The absence of a shift in the PL peak with increasing pump power is characteristic of direct-gap quantum dots of type I (with charge carriers localized on one side of the hetero-interface) [24]. This is due to the high rate of radiative recombination of charge carriers through levels in quantum dots. In the non-direct-band quantum dots (and type II direct-band quantum dots with charge carrier localization on different sides of the hetero-interface), as a rule, there is a shift in the peak of PL [24,25], which is due to a lower rate of recombination and, as a result, charge accumulation in quantum dots. Due to the Coulomb interaction, the energy levels shift and the energy of optical transitions rises. In our experiments, GeSi QDs grown under ion irradiation contain defects that provide direct optical transitions in the *k*- space with a high radiative recombination rate. The electron is captured at the defect level and recombines with the hole in the quantum dot. In this case, charge accumulation in QD does not occur and the PL peak does not shift. We also note that for the irradiated structure in PL spectra (Figure 1) an additional band is observed in the energy region ~ 0.932 eV, which, as was noted in the work [22], disappears at temperatures > 125 K. The presence of this band in PL spectra can be associated with the formation of defects of {113} type [22,26–29].

To clarify the mechanisms of radiative recombination in the studied structures, the dependence of the integrated PL intensity (I_{PL}) on the power of the exciting laser radiation P was analyzed. For this purpose, the experimental dependence was approximated by the power function $I_{PL}(P) \sim P^m$. From Figure 2 it is evident that at low pumping levels ($P < 35$ W/cm^2) for both structures there is a sublinear dependence of the integrated PL intensity on the power ($m \approx 0.7$), which is peculiar to GeSi QDs [30–33]. At high pump levels ($P \geq 35$ W/cm^2) for structure 2 the dependence on power becomes almost linear ($m \approx 1.02$), which is typical for I-type quantum dots created on the basis of direct-gap semiconductors [34].

To explain the obtained results, it is necessary to take into account that not all GeSi QDs contain optically active defects created by ion irradiation, but only a relatively small part of them. The coefficient of germanium ionization beam is very small $\sim 3 \cdot 10^{-4}$ [5]. For a period of growth of one layer of quantum dots it provides one irradiation dose $\sim 10^{12}$ cm^{-2}. Simple estimates show that with such a radiation dose and a temperature of $T = 500\,^\circ$C and a germanium ion energy of ~ 2 keV, $\sim 10^8$ cm^{-2} stable defects of the split interstitial type can form in the quantum dot layer. This low density is due to the low probability of forming a stable configuration of this type of defect. According to calculations using MD method, the

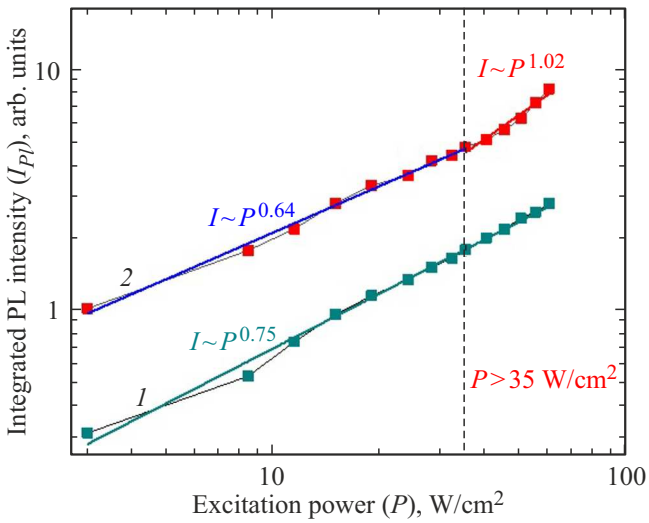

Figure 2. Intergal intensity of GeSi QDs photoluminescence versus laser excitation power for the structures obtained by MBE method: *1* — without irradiation, *2* — with simultaneous irradiation by Ge^+ ions with an energy of ~ 2 keV.

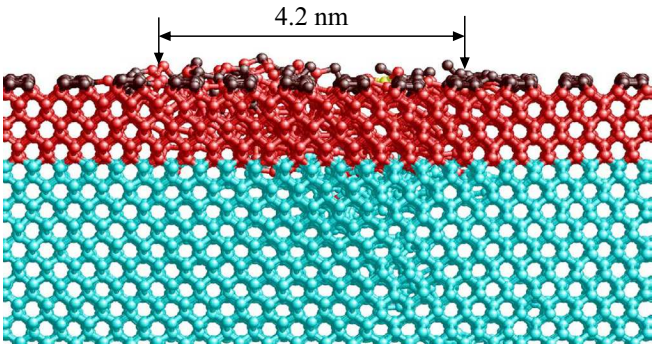

Figure 3. Results of calculations using the MD method of the atomic configuration for a Ge/Si heterostructure 20 ps after exposure to a Ge ion with an energy of 2 keV. Target temperature — 500 °C. Red color corresponds to germanium atoms, blue color to silicon atoms, brown to germanium dimer atoms, yellow to germanium ion. The arrows indicate the characteristic size of the defect cascade induced by the impact of a germanium ion.

characteristic volume of a cascade of defects (Figure 3) created by one germanium ion is $V_c \approx 4\times4\times0.8\,\text{nm}^3$ (here the effective thickness of the germanium layer with quantum dots $h \approx 0.8$ nm is taken into account). At that, according to Kinchin-Pease formula $N = 0.8E/2E_d$ [35,36], each germanium ion with an energy of $E = 2$ keV gives $N \sim 50$ Frenkel pairs (defect formation threshold energy $E_d \approx 15-20$ eV [37]). Since the projective range for ions of such energy is $R_p \approx 5$ nm (calculated using TRIM [36]), the number of interstitials in the germanium layer per ion can be estimated as $n = h/R_p \cdot N \sim 10$. Effectively, one Frenkel pair accounts for a volume $V_i = V_c/n$. For the formation of a split interstitial, two interstitials at a distance of $l \approx 0.25$ nm (the characteristic size of this defect) shall be found [38]. The probability of this event may be estimated as $W_0 \approx l^3/V_i$. It is also necessary to account for the correlations in the interstitials displacement direction, which will result in a decrease in probability by approximately an order of magnitude, which ultimately gives an upper limit on the probability of forming a stable configuration of the split interstitial as $W \sim 0.1 \cdot W_0 \sim 10^{-3}$.

Since the structure is formed at a temperature of 500–600 °C, partial annealing of defects created by ion irradiation occurs. According to MD calculations [39], this leads to a decrease in the defect concentration by $\sim 4-5$ times in the considered temperature range. As a result, the probability of forming a stable configuration of the split interstitial $\sim 10^{-4}$. And precisely at a radiation dose of $\sim 10^{12}\,\text{cm}^{-2}$, one layer of quantum dots contains $\sim 10^{8}\,\text{cm}^{-2}$ split-interstitial type defects. Since the density of the formed quantum dots $N_{\text{QD}} \sim 1.2 \cdot 10^{11}\,\text{cm}^{-2}$, under the condition of a more or less uniform distribution of defects over the layer, it can be assumed that only one in a thousand quantum dots contains an optically active defect. At low laser excitation power, optical transitions occur mainly through defect-free quantum dots, which provides an exponent of $m \approx 0.7$ in the power dependence (Figure 2). However, with increasing pump power, the quantum dots become to be filled more intensely, since the charge carriers in quantum dots without optically active defects do not have time to recombine, and carriers accumulation occurs. At a certain critical pump power, defect-free quantum dots can no longer accept newly arriving photoholes and photoelectrons, and they are forced to tunnel along the quantum dot layer in search of free quantum dots. Once in a quantum dot with an optically active defect, an electron and a hole recombine with high probability, leaving the quantum dot empty again and ready to accept a new pair of carriers. Since the average distance between optically active quantum dots is ~ 30 of the QDs lateral dimensions, and tunnel hops occur through excited states with a large overlap integral [40], it can be assumed that at high pump power, the photocarriers rather quickly find empty quantum dots with optically active defects, and all radiative recombination occurs mainly through this channel, which gives a linear dependence on the power in the experiment (Figure 2). The obtained linear dependence is also consistent with the theoretical findings in [6,14] using DFT (Density Functional Theory) method, which show that interstitial point defects ([110]-split interstitials) located in Ge quantum dot have an electron level in Γ valley and can provide direct optical transitions.

4. Conclusion

A study of the dependence of photoluminescence intensity on the laser excitation power of structures with GeSi QDs formed by molecular beam epitaxy with simultaneous irradiation with Ge^+ ions with an energy of 2 keV demonstrated that at low laser excitation levels ($< 35\,\text{W/cm}^2$) there is a sublinear dependence of the integrated photoluminescence intensity, characteristic of indirect optical transitions of charge carriers in type II quantum dots. At high laser excitation levels ($\geq 35\,\text{W/cm}^2$) the dependence on power becomes linear. The detected linear dependence of the radiation intensity on power, as well as the absence of a shift in the radiation peak at 0.775 eV, indicate the presence of direct optical transitions in the system with GeSi quantum dots created by ion irradiation.

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Conflict of interest

The authors declare that they have no conflict of interest.

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