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Photoinduced light absorption in Ge/Si quantum dots

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Abstract. Photoinduced absorption in nanostructures with doped Ge/Si quantum dots as well as in undoped structures, was studied. Spectra of photoinduced absorption at cryogenic temperatures under direct and indirect in real space resonant interband pumping, as well as time-resolved spectra were obtained. Two high-energy peaks detected in the absorption spectra may be associated with intraband hole transitions from the ground and excited states of quantum dots to the continuous spectrum. The low-energy peak corresponds to interlevel transitions of holes between the ground and excited states. The dynamics of the decay of high-energy peaks can be described in terms of fast and slow components associated with the capture of photoexcited carriers on the levels in a quantum dot. Structures with Ge/Si quantum dots can be used to develop mid-infrared detectors.

Keywords: quantum dots, GeSi, absorption, time-resolved spectra

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Материалы конференции
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Фотоиндуцированное поглощение света в квантовых точках Ge/Si

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Аннотация. Исследовано фотоиндуцированное поглощение света в наноструктурах с легированными квантовыми точками Ge/Si, а также в нелегированных структурах. Получены спектры фотоиндуцированного поглощения при криогенных температурах при прямой и непрямой в реальном пространстве резонансной межзонной накачке, а также их временная динамика. Два высокоэнергетичных пика в спектрах можно связать с межуровневыми дырочными переходами из основного и возбужденного состояний квантовых точек в непрерывный спектр. Низкоэнергетичный пик соответствует межуровневым переходам дырок между основным и возбужденным состояниями точек. Временная динамика затухания высокоэнергетичных пиков может быть описана через быструю и медленную компоненты, связанные с особенностями захвата фотовозбужденных носителей на уровни в квантовой точке. Возможным применением структур с квантовыми точками Ge/Si могут быть детекторы среднего инфракрасного диапазона.

Ключевые слова: квантовые точки, GeSi, поглощение, спектры с временным разрешением

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Introduction

The study of the properties of quantum dots (QDs) is one of the promising directions in the development of optoelectronic devices [1, 2] for applications in various fields from medicine and energetics to electronics and astrophysics. QDs have certain advantages over quantum wells and bulk materials. Three-dimensional confinement of electrons and holes leads to a discrete spectrum of quantized states inside the QD. By controlling the size and composition of QDs, it is possible to obtain the necessary energies of interlevel, intraband and interband transitions of charge carriers, i.e., control the spectral range in which the structure can operate. Various devices can be created based on QDs, in particular photodetectors [3] or lasers with optical and electric pumping at room temperature [4, 5].

Photodetectors based on Ge/Si QDs operating in the mid and far (terahertz) infrared spectral ranges offer several advantages and features compared to quantum well detectors. Among them, for example, the ability to use normal radiation incidence, facilitating the creation of matrix photodetectors [6], high surface density of dots, long lifetime of nonequilibrium charge carriers, compatibility with silicon electronics, enabling the implementation of silicon-based integrated electronic devices [7]. Silicon is widely used in electronics due to its ability to form transistor structures and thin insulator layers. However, unlike QD nanostructures, it is not well-suited for creating light-emitting structures due to its indirect gap of the bulk material. The interband photoluminescence (PL) spectrum of Ge/Si QDs lies in the region of photon energies lower than the band gap of silicon. In CMOS technology [8], this is fundamentally important, both for the development of new devices such as integrated photosensitive arrays in the near and mid-IR spectral ranges, and for the development of a fundamentally new architecture of integrated optical micro- and nanoelectronics based on silicon. Therefore, the urgent task is to find ways to integrate QDs with silicon technology to fully exploit the advantages of optical information transmission in transistor chips. However, on the way to achieving this goal, the problem arises of the insufficiency of the fundamental basis describing the levels and states of charge carriers in Ge/Si QDs.

In addition, QDs can be used to confirm the fundamental principles of quantum theory of particle interactions. The ability to change the number of charge carriers localized in QDs by adjusting the doping level or through the interband optical pumping makes it possible to study many-particle interactions, including the applicability of the generalized Kohn theorem [9, 10].

The objective of this work is to study the dynamic characteristics of photoinduced light absorption in Ge/Si QDs. This study aims to provide information on the distribution of equilibrium and nonequilibrium charge carriers across states inside the QD.

Samples and experimental setup

Nanostructures with QDs were grown using molecular beam epitaxy on a Si(100) substrate. These nanostructures consisted of 10 layers of Ge/Si QDs, which were separated by 15 nm thick Si layers. The QDs were obtained by deposition of 7 monolayers of Ge. In order to prevent mixing of the wetting layer and the resulting islands, Ge/Si QDs were formed using the method of deposition of a surfactant (Sb) before deposition of Ge [11, 12]. This method leads to a decrease in the size of Ge islands, sharpening of their boundaries [13] and an increase in the density of the QD array. As a result, QDs were formed by a Ge_xSi_{1-x} solid solution with an average germanium content of $x \sim 60$–65%. The surface density of the QD islands was about $2{\cdot}10^{11}$ cm^{-2}. Doping with an acceptor boron impurity was carried out by introducing a δ-layer in each silicon layer at a distance of 5 nm from each Ge layer. In this work, samples with an impurity concentration of $8{\cdot}10^{11}$ cm^{-2}, as well as undoped structures, were studied. The dimensions of the QDs were determined using transmission electron microscopy. It was found that the QD islands had an average base size of 14 nm and an average height of 2.7 nm.

The minimum of the conduction band of silicon lies in the <100> direction of the Brillouin zone between Γ and *X* points at point Δ. In Ge the conduction band minimum is located at point *L* at the band boundary in the <111> direction [14]. Thus, the position of the minimum of the conduction band changes from Δ to *L* depending on the concentration of germanium in the GeSi solid solution. It is known that at a Ge concentration of less than 80%, the band structure of the GeSi solid solution is similar to Si. The conduction band profile of a structure with Ge/Si QDs, formed by sixfold degenerate Δ-valleys, depends on the direction due to the splitting of the valleys by compressive and tensile stresses arising during the growth of the structure. In the <001> growth direction, this leads to the formation of a type-II heterointerface with a double Δ-valley (see Fig. 1, solid lines). In the plane of the QD layers in the <100> and <010> directions, four Δ-valleys form a type-I heterointerface (see Fig. 1, dashed lines) [15, 16].

After growing the samples were manufactured in multi-pass geometry. The input and output edges of the sample were grinded at an angle of 45° in order to increase the optical path for probing radiation in the sample due to total internal reflection. This sample geometry also made it possible to measure absorption for *s*- and *p*-polarized light.

Previously, we estimated the energy of hole states in a quantum dot using a simple "quantum box" model [17]. The solution was obtained for a constant potential inside a dot, which was represented in the form of a rectangular parallelepiped with dimensions corresponding to the real sizes of quantum dots. This model took into account the finite height of the confining potential and the difference in effective masses. The schematic band diagram of Ge/Si QDs in Fig. 1 illustrates some energy levels of holes and electrons. Additionally, the figure also includes scheme of charge carrier transitions during interband optical pumping (arrows).

Photoinduced intraband absorption spectra for stationary and time-resolved conditions were obtained using a Bruker Vertex 80v Fourier transform spectrometer with a KBr beamsplitter and a HgCdTe photovoltaic detector cooled to liquid nitrogen temperature with a Ge entrance window. Optical pumping was carried out using continuous-wave Nd:YAG solid-state laser (modulation 600 Hz) with radiation wavelengths of 532 nm or 1064 nm, and pulsed Nd:YAG solid-state laser (pulse duration 10 ns, repetition frequency 20 Hz) with radiation wavelength of 532 nm. The sample was placed in a Janis PTCM-4-7 closed-cycle cryostat with an operating temperature of

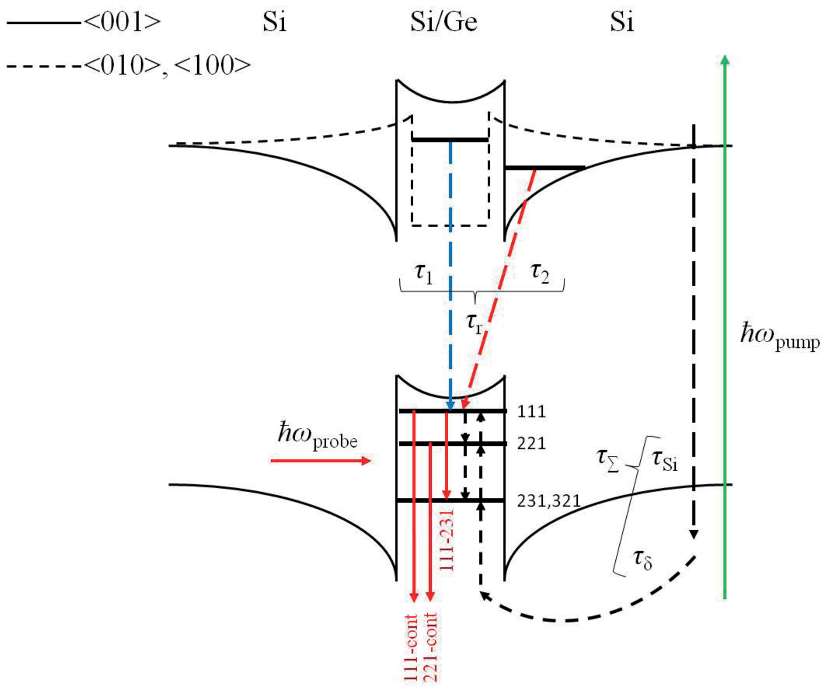

Fig. 1. Schematic representation of the band diagram of Ge/Si QDs in a silicon matrix. Optical transitions within QD states, processes of interband pumping and recombination of photoexcited charge carriers are shown by arrows

4.2–300 K. The globar radiation was directed to the sample through a ZnSe window, optical pumping was carried out through a fused silica window.

Experimental results

The spectra of changes in the intensity of light $|\Delta I|$ transmitted through an undoped sample were obtained using continuous-wave laser (see Fig. 2) at different pump powers and different wavelengths. Excitation of nonequilibrium charge carriers was carried out in two different ways. In the first case, direct in real-space interband optical pumping with a wavelength of 532 nm was used (see green arrow in Fig. 1). In the second case, indirect real-space resonant (1064 nm) pumping was realized from the hole levels in a potential well in a QD to an electron level in a potential well at the heterointerface between the type-II potential barrier of a QD and silicon, arising in the growth direction in the silicon matrix. The obtained absorption spectra of *p*-polarized radiation revealed peaks corresponding to hole transitions from the ground level 111 to the continuous spectrum with a photon energy of 280 meV (arrow 111 → cont), from the excited level 221 to the continuum above the QD with a photon energy of 230 meV (arrow 221 → cont) and peak with a photon energy of 130 meV (arrow 111 → 231), associated with absorption of hole from the ground level 111 to the degenerate excited level 321, 231. These peaks were discovered earlier when measuring equilibrium absorption in QDs [18]. Peak 111 → 321, 231 also manifests itself in the photoinduced absorption spectra for *s*-polarized radiation. This is due to the fact that absorption at these interlevel transitions, according to the polarization selection rules, should occur only for light polarized perpendicular to the growth axis of the structure, and such a polarization component in the geometry of our experiment present in both *s*- and *p*-polarized radiation. A comparison of non-resonant and resonant in real-space pumping showed that the capture time of charge carriers from the barrier to the QD does not affect the photoinduced absorption spectra.

Fig. 3 shows the spectra of transmission changes observed when the undoped sample was pumped with 532 nm continuous-wave radiation at different temperatures. These spectra were obtained by normalizing spectra plotted in Fig. 2 to the equilibrium spectra $\Delta T = |\Delta I| / I_{\text{equilibrium}}$. The spectra contain the previously described peaks for p-polarized light. As the temperature increases, the intensity of 111 → cont peak decreases at slower rate compared to the 221 → cont peak.

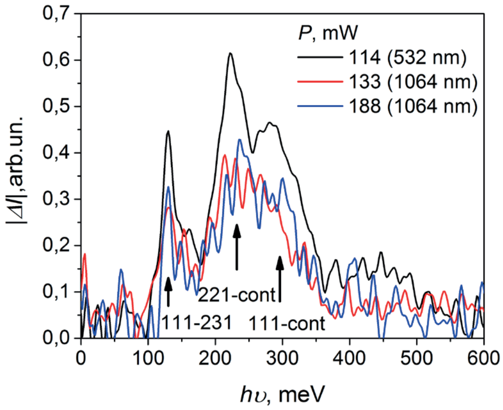

Fig. 2. Photoinduced change in the intensity of *p*-polarized radiation in undoped QDs for wavelength of pumping light λ = 523 nm (black curve) and λ = 1064 nm (blue and red curves) at $T = 4.2$ K

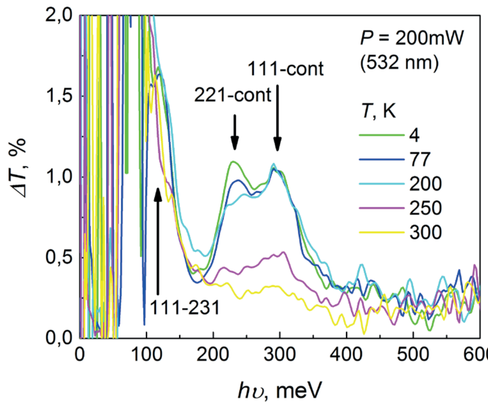

Fig. 3. Spectra of *p*-polarized transmission changes of undoped QDs under λ = 523 nm pumping at different temperatures

This behavior is associated with the temperature broadening of the excited level. As mentioned above, interlevel transitions 111 → 321, 231 contribute to the absorption spectra of light of both *p*- and *s*-polarization and are weakly identified in the equilibrium absorption spectra. Consequently, during the normalization the peak 111 → 321, 231 becomes indistinguishable in the noise. Similar measurements were carried out in a doped sample, i.e. at an initially higher concentration of holes in the QD. The obtained results demonstrate that at a given doping level, the peak corresponding to the transition from the ground state begins to dominate, while the peak corresponding to transitions from the excited state to the continuous spectrum is lost against its background.

The influence of pumping intensity on the photoinduced absorption spectra was also analyzed. All photoinduced absorption spectra were decomposed into three Gaussian contours. From a comparison of the dependences of the peak areas on the optical pump power, it is clear that the peak associated with transitions from the ground state to the continuous spectrum, with a photon energy of 280 meV, grows faster than the peak corresponding to the transitions from the excited state to the continuous spectrum with an energy of 230 meV. The peak 111 → 321, 231, with a photon energy of about 130 meV, remained virtually unchanged with increasing optical pump power. This is due to the fact that additional optical pumping uniformly increases the concentration of charge carriers at the 111 and 321, 231 levels, while only slightly increasing the number of charge carriers in the continuous spectrum above the QD.

Furthermore, time-resolved spectra of photoinduced absorption were obtained using direct interband high-power pulse pumping at a wavelength of 532 nm with a pulse duration of 10 ns (see Fig. 4). The time slice at the moment of laser radiation arrival corresponds to the previously mentioned spectra.

Based on the previously studied dynamics of photoinduced absorption with a fixed probe radiation energy of 300 meV, which corresponds to the 111 → cont transitions [19, 20], the dynamics of absorption decay can be described in terms of fast and slow components. In the present work, contrary to [19, 20], we studied the time dynamic features of the entire absorption spectrum. Fig. 1 schematically illustrates several relaxation paths of photoexcited charge carriers. The fast component τ_r is associated with the probability of direct τ_1 and indirect τ_2 interband transitions in real space, while the slow component τ_Σ is determined by the probability of charge carrier recombination τ_{Si} in Si and the probability of carrier capture in QDs τ_δ.

The dynamics of charge carriers in Ge/Si QDs under optical pumping with 532 nm radiation wavelength is described taking into account recombination in the QDs, capture of charge carriers from the bulk Si matrix, and ejection of carriers into the bulk from the QDs. The kinetic equation can be expressed as follows [20]:

$$\frac{dp}{dt} = -\frac{p}{\tau_r} + \gamma p_b(t)\left(N_{QD} - p\right) - \alpha p, \tag{1}$$

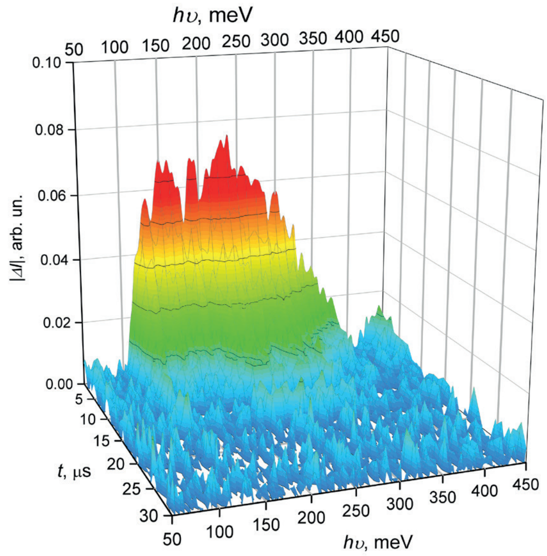

Fig. 1. Schematic representation of the band diagram of Ge/Si QDs in a silicon matrix. Optical transitions within QD states, processes of interband pumping and recombination of photoexcited charge carriers are shown by arrows

where p is the surface density of holes inside QDs, NQD is the number of localized states per unit area of the QD layer, $p_b(t)$ is the concentration of holes in the bulk Si, γ is the capture rate coefficient of holes in QD, α is the coefficient of thermal rate ejection of holes from QD.

This equation does not have an analytical solution in the general case, however, the rate of decay of photoinduced absorption can be empirically described by the sum of two exponential functions [20]:

$$p(t,T) = C_1 \exp\left(-\frac{t}{\tau_r(T)}\right) + C_2 \exp\left(-\frac{t}{\tau_\Sigma(T)}\right). \quad (2)$$

The fast τ_r and slow τ_Σ components of charge carrier relaxation were determined by approximating the time dependences of the photoinduced absorption decay using expression (2) at different temperatures of the sample with undoped QDs and a time resolution of 200 ns. From the obtained temperature dependences of $\tau_r(T)$ and $\tau_\Sigma(T)$, the amplitude of the peaks associated with transitions from the first and second states to the continuum with energies of 280 and 230 meV, respectively, decreases in accordance with expression (2) and can be described by two components. At $T = 4$ K, the times of the slow and fast components for the 111 → cont peak are 28 and 1.8 μs, respectively. For the 221 → cont peak, the component times are 2.7 and 0.2 μs, respectively. It is worth noting that the peak associated with the interlevel transition 111 → 321, 231 has a time component τ_Σ that, in this case could not be described by the used model. This prevents us to reliably determine the corresponding time. The fast decay time τ_r for interlevel transitions is 2.5 μs at 4 K, which corresponded to the same component in intraband transitions from ground 111 level.

Conclusions

Spectra of photoinduced absorption and transmission of polarized radiation were experimentally obtained in the mid-infrared spectral range for Ge/Si QDs. The measurements were conducted under direct (532 nm) and indirect in real-space resonant (1064 nm) interband optical pumping. Peaks associated with hole transitions from the ground and excited states to the continuous spectrum as well as a peak associated with interlevel hole transitions between the ground and excited states were detected.

A study of the dynamics of the decay of photoinduced absorption during pulsed interband pumping showed that the time dependence of high-energy peaks in the spectrum can be approximated by a biexponential function. The fast decay time of the relaxation curve is associated with the probability of interband recombination in the QD, while the slow time is associated with the capture and ejection of holes in the QD. Analysis of the time dependence of the low-energy peak indicated that recombination processes associated with interlevel transitions occur much slower than processes with transitions to a continuous spectrum above the QD.

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