Charge carrier kinetics in GeSi/Si quantum dots probed by mid-infrared radiation

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Abstract. The mid-infrared photo-induced absorption relaxation kinetics of the self-assembled array of GeSi quantum dots in Si matrix was studied in the conditions of short pulsed interband optical excitation. The measured absorption decay curves directly show the temporal evolution of the population of the QD ground states. The analysis of the experimental data allowed us to estimate characteristic recombination and capture times, and the electron localization energy in the vicinity of QD.

1. Introduction
GeSi/Si quantum dot structures are promising objects for the development of the quantum dot infrared photodetectors (QDIP) [1,2]. Germanium quantum dots (QDs) in silicon matrix self-assemble during the strain-driven epitaxy. The built-in strain affects the conduction and valence band profiles in the vicinity of the quantum dot interface. The details of the band alignment are important for the performance of the QDIPs. The mid-infrared optical spectroscopy is a powerful instrument for study of the energy structure of the charge carriers in QD structures, while kinetics of the absorption decay allows one to resolve the information about characteristic relaxation times. In the present work, we study the decay curves of the photo-induced absorption after a short excitation of the electron-hole pairs in GeSi/Si QDs in a wide temperature range.

2. Samples and methods
The charge carrier kinetics studies were carried out in a dense array of the surfactant-mediated GeSi/Si (001) undoped quantum dot structures. The details of growth process are described in Ref. [3].

Non-equilibrium charge carriers were created in structure by means of short 10 ns pulse of interband optical excitation from the solid state YAG:Nd laser with frequency multiplication with pulse energy up to 9 mJ. The temporal evolution of the population of the hole ground state inside the quantum dot was monitored measuring a decay of the intraband mid-infrared (mid-IR) optical absorption of the sample after the pulsed interband excitation. The wide-band globar coupled with prismatic monochromator was used as a source of tuneable mid-IR probe radiation. The intensity of the mid-IR radiation transmitted through the sample was measured with liquid-nitrogen-cooled MCT detector with about 100 ns rise time.
The photon energy of the mid-IR probe light was selected to match the energy of the transition between dot ground state and the continuum, as it is known from the previous steady-state photo-induced absorption and photoconductivity spectroscopy [4]. So the mid-IR optical absorption change under optical excitation is directly proportional to the number of holes in the ground state of QD.

3. Experimental results and discussion

The photo-induced absorption decay curves were measured in a wide range of the excitation levels at lattice temperatures $T_L$ from 80 K up to 300 K with a 10 K interval. Typical decay curves for high and low temperatures are plotted in figure 1 at probe photon energy 0.3 eV as a time dependencies of the optical transmission change $|\Delta T|$. The low-temperature (below 200 K) relaxation curves demonstrate two-stage behavior with a fast (few $\mu$s) and slow (tens of $\mu$s) characteristic decay times. A slow relaxation time tends to decrease with an increase in temperature, while the fast relaxation time slightly grows up. Inset in figure 1 shows the temperature dependencies of the photo-induced absorption at two moments of time inside the fast (3 $\mu$s after the excitation) and slow (25 $\mu$s after the excitation) decay stages.

![Figure 1](image_url)

**Figure 1.** Typical decay curves of photo-induced absorption at 0.3 eV for high and low temperatures after short pulsed excitation. Inset shows the temperature dependence of the optical transmission change at the moments of 3 $\mu$s and 25 $\mu$s after the excitation. Dashed curves show the biexponential fit.
We attribute the fast decay component to the direct and/or indirect in real space recombination [5] of the electrons and holes in the quantum dots. Then, the inverse fast time shows the probability of recombination proportional to the number of electrons localized in the vicinity of the quantum dot. It decreases with temperature due to the thermal electron delocalization.

In order to explain the experimentally observed biexponential decay we suggest to introduce the positive contribution to the QD depopulation rate that exponentially decays itself. This contribution can stand for the capture of the bulk holes in silicon matrix to the quantum dots. Then, the inverse slow time shows the probability of capture of the holes to the dot ground state.

The dashed curves in figure 1 show the result of biexponential fit of the whole set of experimental $|\Delta T|$ curves with additional conditions on the inverse fast and slow times that were assumed to be activation-like dependent on temperature with activation energies $E_e$ and $E_h$, respectively. The resulting temperature dependencies of the fast and slow times are plotted in figure 2. The corresponding activation energies $E_e = 25$ meV and $E_h = 70$ meV were derived.

![Graph](image)

**Figure 2.** Temperature dependencies of the fast ($\tau_f$, right axis) and slow ($\tau_s$, left axis) decay times derived from the biexponential fit of the experimental absorption decay curves.

The localizations of the electrons in the vicinity of the QDs with the binding energy $E_e$ arises due to the strain-induced band bending at the quantum dot interface. The 25 meV binding energy derived from the absorption decay curves analysis is comparable with the calculated energy of electron strain-induced localization [6].

The presented experimental results show the significant temperature dependence of the hole capture time. The capture probability rises with temperature with relatively large 70 meV activation energy. Again, this energy can be treated as a strain-induced valence band barrier at the QD interface for the bulk holes in silicon matrix. However, the QD capture time can be significantly dependent on
such factors as initial population and charge state of the QD, and it is necessary to exploit a set of master equations for detailed quantitative analysis of the QD state population kinetics.

4. Conclusion
In this work, we present the temperature evolution of the ground state population kinetics in the GeSi/Si quantum dot structures. The characteristic recombination and capture times and the electron binding energy were estimated. It was shown that non-equilibrium population of the hole ground state in the GeSi/Si quantum dots is highly temperature dependent.

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References
[1] Yakimov A I, Kirienko V V, Armbrister V A, Bloshkin A A, and Dvurechenskii A V 2015 *Appl. Phys. Lett.* **107** 213502
[2] Yakimov A I, Kirienko V V, Bloshkin A A, Armbrister V A, Kuchinskaya P A, and Dvurechenskii A V 2015 *Appl. Phys. Lett.* **106** 032104
[3] Tonkikh A and Werner P 2013 *Physica Status Solidi B* **250** 1795
[4] Sofronov A N, Vorobjev L E, Firsov D A, Panevin V Y, Balagula R M, Werner P, and Tonkikh A A 2015 *Superlattices and Microstructures* **87** 53
[5] Kamenev B V, Tsybeskov L, Baribeau J-M, and Lockwood D J 2005 *Phys. Rev. B* **72** 193306
[6] Yakimov A I, Dvurechenskii A V, Nikiforov A I, Bloshkin A A, Nenashev A V, and Volodin V A 2006 *Phys. Rev. B* **73** 115333