# Dynamics of mid-infrared light absorption related to photo-excited charge carriers in Ge/Si quantum dots

**O I Kirilenko[1], R M Balagula[1], A N Sofronov[1], V Yu Panevin[1], L E Vorobjev[1], D A Firsov[1] and A A Tonkikh[2,3]**

[1] Peter the Great St. Petersburg Polytechnic University, Polytechnicheskaya 29, St. Petersburg 195251, Russia

[2] Max Planck Institute of Microstructure Physics, Weinberg 2, Halle (Saale) 06120, Germany

[3] Institute for Physics of Microstructures RAS, GSP-105, Nizhny Novgorod 603950, Russia

**Abstract**. Mid-infrared optical absorption spectra of Ge/Si quantum dot structures in polarized light were obtained under conditions of interband optical excitation. Mid-infrared absorption changes in comparison to equilibrium conditions for certain light polarization in spectral range of 0.25 – 0.6 eV. The sign of the effect is found to be different in various spectral ranges. Transients of photoinduced absorption contain fast and slow components. The fast decay component of the absorption is related to the direct recombination of localized holes and electrons in quantum dots while a slow component is determined by significantly less probable processes of indirect in real space recombination.

## 1. Introduction

Nanostructures with quantum dots are being studied by a number of scientific groups around the globe. Intensive studies of optical and photoelectric properties of Ge/Si quantum dots are motivated by a possibility to develop mid- and near-infrared optoelectronic devices based on these structures. Lately a noticeable progress has been made in technology aspects of Ge/Si quantum dots growth [1]. There is a number of papers dedicated to studies of near-IR photoluminescence dynamics [2,3], and the results contain a lot of original information explaining recombination processes in quantum dots. The present work is focused on the optical properties of Ge/Si quantum dots in the mid-infrared spectral range under conditions of non-equilibrium population of the dot states with photo-excited charge carriers and also presents recent results on photoinduced absorption dynamics in GeSi quantum dot structures

## 2. Experimental setup and samples

Structure with MBE-grown Ge/Si quantum dots on silicon (100) substrate was studied. Structure contains 10 periods of quantum dot layers formed by deposition of 7 Ge monolayers with 0.14 ML/s rate at $600^0$C. Quantum dot layers were separated from each other with 15 nm Si interlayer. Structure was isolated from substrate with epitaxial 100 nm silicon buffer layer and was covered with 100 nm top layer. A surfactant (Sb) was used during the growth process in order to increase the density of the QD array as described in [4]. No doping operations were performed during structure growth.

QD structure was characterized using transmission electron microscopy and atomic force microscopy. The first method allowed us to determine the shape of quantum dots as a truncated pyramid with average height of 2.7 nm, and average base size of 14 nm. Results of atomic force microscopy of specially grown samples with no surface silicon layer showed the QD density at the level of $2 \cdot 10^{11}$ cm$^{-2}$. The material of the quantum dot is $Ge_xSi_{1-x}$ solid solution with approximate germanium content 60-65%.

Optical measurements in mid-IR were performed with the samples prepared in multi-pass geometry with opposite sample sides ground at $45^0$ angle so the light beam could pass the QDs layers multiple times

due to total internal reflection in the sample. This geometry allows one to study optical transmission of the sample for different light polarizations with respect to growth direction.

Charge carriers in the undoped sample were created only using interband illumination of the top surface of the sample with solid-state 532 nm laser.

Experimental setup for steady-state photoinduced absorption spectra measurements is the variation of steady state pump-probe technique. Absorption spectra in mid-infrared range were measured with automated prismatic monochromator. Globar was used as a source of probe radiation. Intensity of the probe radiation after the sample was measured with InSb photodetector. Sample was mounted in the liquid nitrogen-cooled cryostat with ZnSe optical windows. ZnSe metal grid polarizer was placed in front of the entrance window. Both probe and pump radiation were mechanically chopped at different frequencies, and photodetector signal was measured simultaneously with two SR830 Lock-in amplifiers. One of them measured a signal proportional to the equilibrium optical transmittance $T$ of the sample, while the second one measured a signal proportional to the variation of the optical transmittance $\Delta T$, related only to non-equilibrium photo-excited charge carriers.

Dynamics of photoinduced absorption was registered with digital oscilloscope after a short 10 ns pump radiation pulse at the spectral maximum of p-polarized light absorption.

## 3. Experimental results

Typical measured steady-state spectra of photo-induced absorption related to photo-excited charge carriers captured to the QDs are shown in Figure 1 (a) for p- and s-polarization of probe radiation at lattice temperature $T_L = 79$ K. Figure 1(b) presents spectra for probe light polarized along the growth direction (z) at two different temperatures. These spectra were calculated from measured data taking into account that polarization vector for *p*-polarization contains equal *xy*- and *z*-components (while *s*-polarization is in fact 100% *xy*).

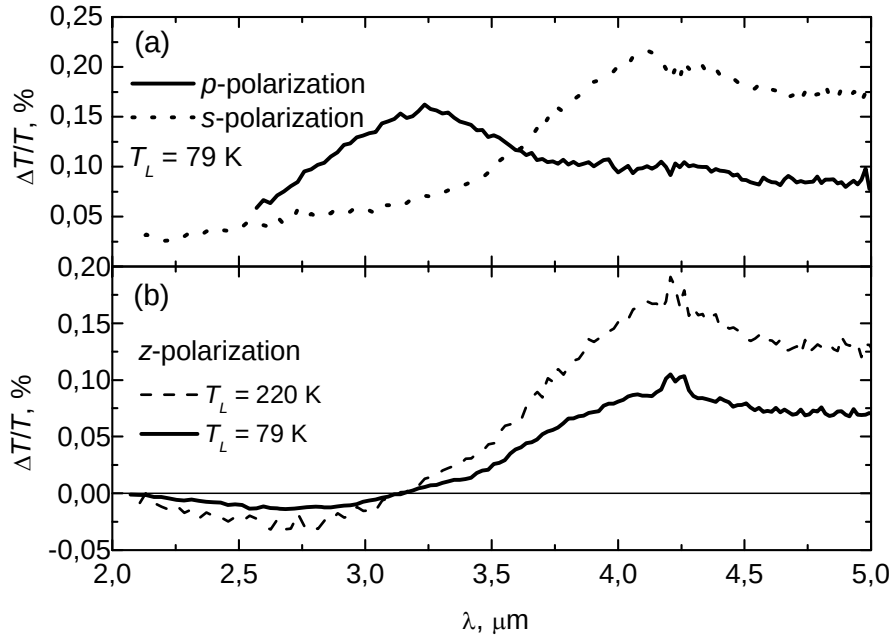


**Fig.1.** Typical spectra of photo-induced absorption for p- and s- infrared light polarization at $T_L = 79$ K (a); calculated spectra for probe light polarized along the growth direction at 79 K and 220 K (b). Pump intensity $I = 20$ mW

Since the optical response of the sample related to the quantization of the hole energy spectrum in the growth direction (which is the main confinement factor due to small "height" of the QDs with respect to its lateral dimensions) is sensitive only to z-polarized light, one can treat the spectral features presented in Figure 1(b) as related to optical transitions of the non-equilibrium holes between the energy states of the QD. The noticeable feature of the $\Delta T/T$ spectrum for z-polarized probe is the change of the sign of the $\Delta T$.

Positive value of $\Delta T$ corresponds to the increase of absorption at the certain spectral range close to 4 μm. Absorption increase is related to the contribution of optical transitions of non-equilibrium holes from the quantum dot ground states to the continuum of states, and the energy of these transitions is about 300 meV. Negative value of $\Delta T$ corresponds to the decrease of absorption at the presence of non-equilibrium charge carriers at the spectral range close to 2.75 μm. This fact can be associated with the suppression of interband-like processes such as generation of hole inside the dot and bound electron outside of the dot under conditions of full occupation of appropriate discrete states with charge carriers photo-generated by pump radiation. Figure 2 shows the optical transition scheme. Transitions marked with solid arrow 1 are responsible for absorption increase at long-wavelength part of $\Delta T/T$ spectrum. Suppression of transitions with energy of about 450 meV marked with dashed arrow 2 is responsible for absorption change drop below zero.

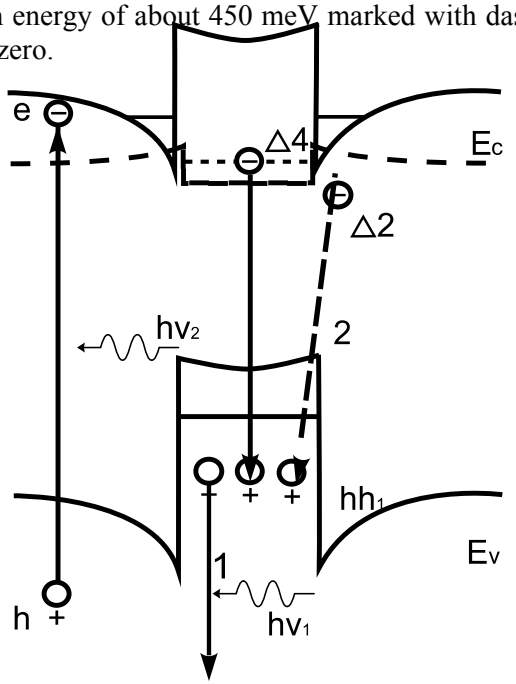


**Fig.2.** Band diagram of the Ge/Si QD and optical transition scheme.

The dynamics of the processes involving transitions from ground states was investigated at the probe radiation wavelength corresponding to the maximum of the positive part of $\Delta T/T$ spectrum. The absorption relaxation curves were obtained in the wide lattice temperature range $T_L$ = 90…290 K. Typical decay curve at the intermediate temperature of 190 K is presented in Figure 3.

Observed $\Delta T/T$ decay curves demonstrate two-stage behavior with a pronounced fast and slow components with a characteristic decay times that differ by almost one order of magnitude at 90 K. Heating of the sample leads to the decrease of the slow component decay time so at room temperature it can not be isolated from the fast one.

Such two-stage kinetics was observed earlier in interband photoluminescence studies [2,3]. Since the measured $\Delta T/T$ decay curves in essence reflect the dynamics of the population of the QD ground state with non-equilibrium charge carriers, the microscopic mechanism of the photoinduced absorption relaxation should be similar to the one of the interband photoluminescence. Non-uniform mechanical strain distribution in the QD structure can reduce the degeneracy of the electron Δ-valleys in silicon around the QD [7] and form a complicated potential profile at the quantum dot interface similar to the one presented in Figure 2. Two Δ2 valleys stretched along z axis form a potential well for electrons at the silicon side of the QD Ge/Si interface while four Δ4 valleys stretched in the xy-plane can lead to weak electron localization inside the quantum dot. Thus, two different ways of recombination are possible in such structures. Direct recombination of localized in Δ4 states electrons and holes in quantum dots can be associated with a fast decay component. Indirect in real space recombination of electrons in Δ2 states with holes inside the dot has significantly less probability due to weak overlap of the involved states and can be responsible for a slow decay component.

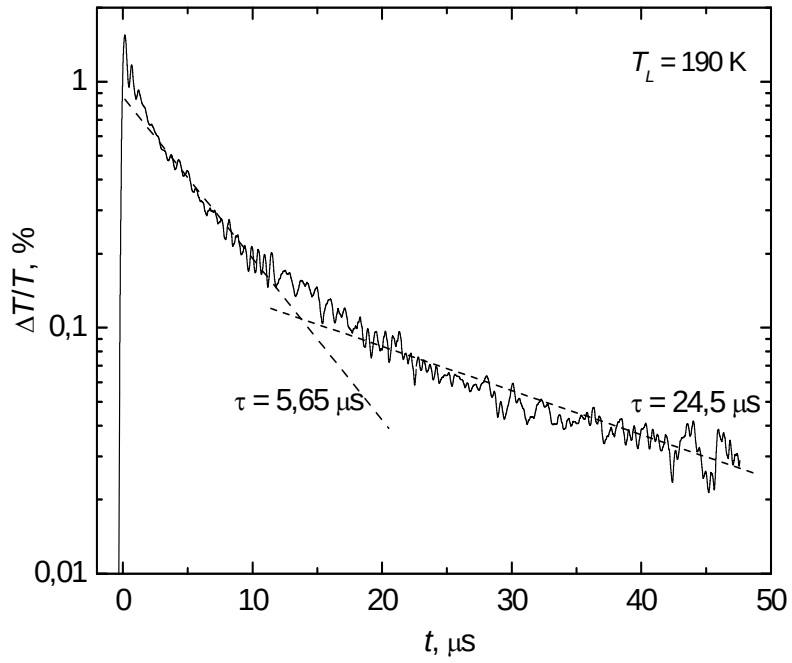


**Fig. 3.** Typical photoinduced absorption relaxation curve at $T_L$ = 190 K. Characteristic time values derived from the experimental data are shown for each decay component.

## 4. Conclusion

In conclusion, in this work the photo-induced mid-infrared absorption spectra were measured in Ge/Si quantum dot structures. The analogue of the Burstein-Moss effect is observed. Dynamics of photoinduced light absorption was studied under conditions of pulse interband optical excitation. Two distinct temporal components were isolated. The underlying mechanisms were proposed.

## Acknowledgments

This work has been supported by the Russian Foundation for Basic Research (Project № 13-02-12203) and the Ministry of Education and Science of the Russian Federation.

## References

[1] Tonkikh A A, Zakharov N D, Novikov A V, Kudryavtsev K E, Talalaev V G, Fuhrmann B, Leipner H S and Werner P 2012 *Thin Solid Films* **520** 3322
[2] Julsgaard B, Balling P, Lundsgaard Hansen J, Svane A and Nylandsted Larsen A 2011 *APL* **98** 093101
[3] Kamenev B V, Tsybeskov L, Baribeau J-M, Lockwood D J 2005 *Phys. Rev.* B **72** 193306-1-4
[4] Tonkikh A A, Zakharov N D, Talalaev V G, Werner P 2010 *Phys. Status Solidi* RRL **4**, 224.
[5] Panevin V Yu, Sofronov A N, Vorobjev L E, Firsov D A, Shalygin V A, Vinnichenko M Ya, Balagula R M, Tonkikh A A, Werner P, Fuhrman B, Schmidt G. 2013 *Semiconductors*, **47**, 1574.
[6] Talochkin A B, Chistokhin I B 2011 *Semiconductors*, **45**, 907
[7] Kurdi M El, Sauvage S, Fishman G and Boucaud P 2006 *Phys. Rev.* B **73** 195327..