Mid-infrared light absorption by photo-excited charge carriers in Ge/Si quantum dots

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Abstract. Mid-infrared optical absorption spectra of Ge/Si quantum dot structures in polarized light were obtained under conditions of additional interband optical excitation. Photo-excited electron-hole pairs captured to quantum dots cause mid-infrared absorption changes for certain light polarization in spectral range of 0.25 – 0.6 eV. The sign of the effect is found to be different in different spectral ranges. There is an increase of absorption at the long-wavelength edge of the spectrum and a decrease of absorption at the short-wavelength edge. Absorption increase is considered to be related to contribution of optical transitions of non-equilibrium holes from the quantum dot ground states, while absorption decrease is associated with suppression of interband-like processes with generation of hole inside the dot and bound electron outside of the dot under conditions of full occupation of appropriate discrete states.

1. Introduction

Ge/Si quantum dot structures are attractive objects for a number of scientific groups around the world. Intensive studies of optical and photoelectric properties of Ge/Si quantum dots are explained by a possibility to develop the mid- and near-infrared optoelectronic devices based on these structures. One of the proposed advantages of this material system is the possibility of integration into the existing silicon technology.

In the recent several years a noticeable progress has been made in technology aspects of Ge/Si quantum dots growth (see, for example, [1] and reference therein), in theoretical studies of specific charge carriers quantization features [2] and experimental studies of near-infrared [3, 4] and mid-infrared [5, 6] optical properties, and in device development [7].

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.
2. Experimental setup and samples

Structure with self-assembled MBE-grown Ge/Si quantum dots on Si (001) 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°C. Quantum dot layers were separated from each other with 15 nm Si interlayer. The structure was separated from substrate with an epitaxial 100 nm silicon buffer layer and was covered with a 100 nm cover layer (see figure 1(a)). A surfactant (Sb) was used during the growth process in order to increase the density of the QD array as described in [8].

The form and dimensions of QDs were estimated using transmission electron microscopy. Quantum dots have a pyramidal shape with the average height of 2.7 nm, and the average base size of 14 nm. The results of atomic force microscopy of specially grown samples with one QD layer and no surface silicon layer showed that the QD density was $2 \times 10^{11} \text{ cm}^{-2}$. The approximate content of germanium in a resulting quantum dot material is 60–65%. No special doping was performed during structure growth.

The samples in common multi-pass geometry were prepared from the structure for optical measurements in the mid-infrared spectral range (see figure 1(b)). Two opposite sample sides were mechanically processed so the angle between the incident beam (normal to the side) and the top surface of the sample was 45°. The light beam passed the structure multiple times due to internal reflection in the sample. This geometry also allows us to use different light polarization with respect to growth direction.

Since the structure under investigation was not specially doped, the charge carriers were created only using additional illumination of the top surface of the sample with a solid-state 532 nm laser.

![Figure 1](link)

Figure 1. Schematics of the structure, dot shape (a), and experimental sample geometry (b). Vector diagram shows the orientation of probe radiation wave vector $\mathbf{k}$ and two possible polarization vectors $\mathbf{e}_s$ for $s$- and $p$-polarization. The last one has the non-zero component in the direction $z$ of growth axis.

Experiment scheme in general is similar to the variation of steady state pump-probe technique. The absorption spectra in the mid-infrared range were measured with an automated experimental setup based on a prismatic monochromator with a globar as a source of broadband infrared radiation. A spherical mirror based optical scheme formed a thin strip of monochromatic infrared light (probe radiation) at the cant side surface of the sample and focused light from the opposite side of the sample to the photosensitive area of liquid nitrogen cooled InSb photodetector. Probe radiation polarization was selected with a metal grid ZnSe polarizer. The sample was mounted in a liquid nitrogen cryostat with a pair of ZnSe optical windows in the direction of probe light propagation and a quartz optical window in the orthogonal direction to allow the excitation of sample with interband laser radiation (pump radiation). Probe radiation was mechanically chopped at 900 Hz, simultaneously exciting laser radiation was chopped at 90 Hz. The photodetector signal was measured simultaneously.
with two SR830 Lock-In amplifiers. The first one was locked to the frequency of infrared radiation modulation (900 Hz) and measured a signal proportional to the equilibrium optical transmittance $T$ of the sample. The second one was locked to the excitation modulation frequency (90 Hz) and measured a signal proportional to the variation of the optical transmittance $\Delta T$, related only to non-equilibrium photo-excited charge carriers.

3. Experimental results

It is well known that Ge/Si material system forms heterointerfaces of second type with a quantum well for holes in the valence band and a barrier for electrons in the conduction band. A pump radiation generates non-equilibrium electron-hole pairs with a relatively large energy. After fast relaxation processes the holes are captured inside the quantum dots, while the electrons tend to be localized around the dot as discussed later in this section. This non-equilibrium population of quantum dot energy spectrum states can effect the optical absorption of the probe radiation. A complicated shape of the quantum dot and a noticeable size difference of the dot in the plane of structure and in the growth direction allow one to expect a certain selection rules of the intraband absorption for in-plane $xy$-polarization and $z$-polarization along the growth direction. In experiment, we measured $\Delta T/T$ spectra for $s$- and $p$-polarization of probing radiation. The typical measured spectra of photo-induced absorption for these two polarizations are shown in figure 2.

Since the polarization vector for $p$-polarization in experiment contains equal $xy$- and $z$-components (while $s$-polarization is in fact $xy$- one, see figure 1(b)), one can obtain the value of $\Delta T/T$ for $z$-polarized light with simple conversion $(\Delta T/T)_z = 2((\Delta T/T)_p - (\Delta T/T)_s)/2).$ The resulted spectra are shown in figure 3.

![Figure 2](image_url). Measured spectra of photo-induced absorption for $p$- and $s$- infrared light polarization at $T = 79$ K and pump intensity $I = 20$ mW.
Figure 3. Spectrum of photo-induced absorption for infrared light polarized along the growth direction calculated from measured data at $T = 79$ K and pump intensity $I = 20$ mW. Inset shows the optical transitions scheme. Transitions marked with solid arrow 1 are responsible for absorption increase at long-wavelength part of spectrum. Suppression of transitions marked with dashed arrow 2 is responsible for absorption change drop below zero.

One can see that presence of nonequilibrium charge carriers leads to an increase in the optical absorption for $z$-polarized light in a wide spectral range with the centre at 0.3 eV and to a decrease in the absorption at photon energies about 0.45 eV. The absorption at 0.3 eV is related to hole optical transitions from the ground state of quantum dot to continuum of states above the barrier (see inset on figure 3, where this transition is depicted with solid arrow 1). The spectral position of this absorption feature correlates well with the previously measured mid-infrared photoconductivity spectra in similar doped structure [9]. Inhomogeneous broadening of observed absorption peaks is related to the variation of quantum dots geometry.

The decrease of absorption at 0.45 eV can be related to the dynamic analogue of bulk Burstein–Moss effect. Processes of photogeneration of electrons around the dot and holes inside the dot by probe radiation become impossible due to the full population of the final states for these transitions by charge carriers generated by pump radiation. This type of transition is shown at inset in figure 3 with dashed arrow 2. Again, the spectral position of this effect is in good agreement with the results of interband photoconductivity measurements in Ge/Si quantum dot structures [4].

The observed polarization of the interband absorption can be related to the complicated potential profile of the silicon conduction band at the quantum dot heterointerface. In particular, non-uniform distribution of mechanical strain in a multilayer structure leads to the energy shift of only two X-valleys of conduction band aligned along the structure growth direction, while the other four valleys remain unshifted [2]. This leads to the angle-dependent potential profile, and electron localization arises only at certain side of the dot, but not uniformly around the dot.
The temperature dependence of the $\Delta T/T$ value at the spectral maximum was also measured. The experimental data demonstrate a significant suppression of the observed effect with the increase of temperature. At the temperatures above 250 K the variation of the optical transmittance related to non-equilibrium charge carriers becomes negligible.

4. Conclusion
In conclusion, in this work the photo-induced mid-infrared absorption spectra were measured in Ge/Si quantum dot structures. An increase of absorption under interband optical excitation related to the contribution of optical transitions of non-equilibrium holes from the quantum dot ground state is observed for light polarized along the growth direction. At the same time, at shorter wavelengths the decrease of absorption is observed for the same light polarization. This effect is considered to be caused by the suppression of interband-like optical transitions between bound states in the valence and conduction bands and can be described as a dynamic analogue of the Burstein–Moss effect.

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