Multi-particle effects in far-IR optical transmission spectra of Ge/Si quantum dots

D A Pashnev, R M Balagula, A N Sofronov, D A Firsov and L E Vorobjev

Peter the Great St.Petersburg Polytechnic University, Polytechnicheskaya 29, St.Petersburg 195251, Russia
e-mail: pashnevDaniil@gmail.com

Abstract. Photoinduced and equilibrium optical transmission was studied in undoped and \( \delta \)-doped Ge/Si quantum dot structures in the far-infrared spectral range. Multi-particle excitations were observed in the absorption spectra of the structures with different doping levels. Photoinduced absorption spectra demonstrate the thermal distribution of the photoexcited holes over the single-particle states of the quantum dot ensemble in contrast with the equilibrium absorption spectra.

1. Introduction

The semiconductor epitaxial structures with quantum dots draw attention in the present time due to the specifics of the energy spectrum of charge carriers, which is discrete, and, therefore, the possibility of their use in optoelectronic devices. Structures with quantum Ge/Si dots are especially promising if we consider the possibility of their integration in advanced silicon technology.

Novel detectors of both near and mid-infrared radiation, operating via interband and intraband optical transitions of holes between the quantum-dot levels, can be created on the basis of systems with Ge/Si quantum dots. Detectors in the mid-IR range [1, 2] have advantages over similar devices based on quantum wells, such as the absence of strict selection rules for the radiation polarization, which leads to the possibility of using the simplest geometry of the normal incidence of radiation on the surface of the photosensitive device.

As most of the self-assembled quantum dot systems, the Ge/Si quantum dots (QDs) have significant size dispersion. It leads to the inhomogeneous broadening of the energy states of the quantum dot ensemble. In such conditions, the exact distribution of charge carriers over the dot ensemble is a significant issue. In the classic work [3], the nonthermal distribution is assumed in the framework of the random population approach for quantum dot systems based on the master equations. Under such conditions, at low temperatures dots with different sizes (and ground state energies) have the same population since there is a lack of interdot coupling. However, in structures with a dense array of quantum dots, it is not always the case, and the lateral correlation can exist. Indeed, the authors of paper [4] successfully applied the thermal (Fermi) distribution for the description of the temperature dependencies of the intraband absorption in the InAs/GaAs quantum dot system. However, the inhomogeneous broadening was not taken into account in the model used in reference [4].

In doped quantum dot structures, the dots with different charge states (different number of electrons or holes inside) can exist. In this case, one should consider the distribution of charge carriers
over the alternative multi-particle configurations rather than over the single-particle states come from the quantum confinement.

In the present work, we study the optical transmission spectra of the dense array of Ge/Si quantum dots in the far infrared spectral range close to the very first interlevel resonance in the valence band. In this spectral range, the specific conditions are realized, when the optical transition energy is comparable with the inhomogeneous broadening of the interlevel resonance. We focused on both the equilibrium absorption spectroscopy and the steady-state non-equilibrium absorption spectroscopy under the interband optical excitation of the electron-hole pairs (so-called photoinduced absorption spectroscopy).

2. Experimental setup and samples
The MBE-grown structures with 10 periods of self-assembled GeSi/Si QDs were studied in this work. A surfactant (Sb) was used during QD growth process in order to increase the QDs density as described elsewhere [5]. We studied both the undoped structure and p-doped in a $\delta$-layer structures with boron surface densities of $4 \times 10^{11}$, $8 \times 10^{11}$, and $1.2 \times 10^{12}$ cm$^{-2}$. The QDs have a pyramidal shape with an average height of $h = 2.7$ nm and the average base size of $b = 14$ nm. The surface density of QDs is $2 \times 10^{11}$ cm$^{-2}$.

We used the multipass sample geometry similar to the one used in our previous studies of the steady-state and transient photoinduced absorption in this structures in the mid-infrared spectral range [6]. The samples for optical measurements were mounted in a liquid nitrogen cooled cryostat with the controllable temperature $T_L$ in the range between 80 and 320 K. The cryostat was equipped with a pair of polyethylene optical windows for the far-infrared probe radiation and a quartz optical window for the pump illumination.

The spectra of optical absorption in the far-infrared spectral range were recorded with a Bruker Vertex 80v Fourier transform spectrometer. A globar was used as a source of broadband infrared radiation. The mylar beamsplitter ensured the measurements in the spectral range of 100 - 500 cm$^{-1}$ (approximately 10-60 meV). The intensity of the probe radiation, which has passed through the sample, was measured with a liquid He-cooled silicon bolometer.

A solid-state CW YAG:Nd laser with a wavelength of 532 nm and the maximal output optical power of 100 mW was used as a source of the pump illumination creating additional charge carriers in the samples. The photoinduced absorption spectra were recorded in a step-scan spectrometer mode with a SR830 Lock-In amplifier, which was modelocked to the chopping frequency of the pump radiation.

3. Experimental results and discussion
Let us consider first the equilibrium optical transmission spectra of the doped QDs that were measured at 80 K and 300 K. They are plotted in figure 1 for three structures with a nominal doping level (the ratio of the acceptor and QDs surface densities) of 2, 4, and 6 holes per dot and are surprisingly similar for all three structures.

Within the single-particle approximation, one should expect a completely different behaviour of the absorption peak related to the inhomogeneously broadened interlevel resonance of QDs with the increase of the total number of holes in the system. In an array of quantum dots with size dispersion, holes first populate the larger dots due to the lower ground state energies. The increase of the number of holes leads to population of smaller and smaller dots, where the interlevel resonance energy increases. As a result, for non-interacting holes one should expect the interlevel absorption spectrum to widen into a short-wave-length region with an increase of the nominal doping level, while the long-wave-length edge of the spectrum corresponds to the Gaussian distribution of the dots’ sizes.

Consideration of the Coulomb interaction of the holes changes this picture dramatically. A charged dot with more than one hole inside should be treated as a dot with a quasi-particle that consists of several interacting holes (sometimes referred as 0D-plasmons for a large number of particles). Addition of one more hole changes (increases) the ground state energy of this quasi-particle (mainly
due to the Coulomb repulsion), but the energy differences between the ground and low excited states remain the same. This fact was firstly mentioned by W. Kohn in the work [7] for cyclotron resonance of 3D electron gas with interaction, and later generalized for quantum dots (see, for example, the recent work [8] and references therein) with a parabolic confinement potential (the so-called “parabolic” quantum dot model, that can be applied to real QDs with finite potential barriers for description of the lowest excited states since their energies are usually significantly lower than the barrier height).

Thus, the interlevel resonance energy of charged dot does not depend on the number of holes inside, but is determined only by the confinement, i.e. by the geometry of the dot. However, the ground state energy of the confined few-hole quasi-particle does depend on the number of holes in this quasi-particle. In a broadened array of quantum dots with a low nominal doping, a certain hole can find an empty dot with ground state energy less than the one of the already charged dot. As a result, the distribution of the holes over the dot ensemble (which, in fact, is the thermal distribution over the alternative-multi-hole configurations) is more intricate than the thermal one (over the single-particle states). Increase of doping leads just to the increase of the hole number in certain quasi-particles (with the lowest energies), and the inter-(quasi-particle)-level absorption spectrum does not change. Experimental spectra of doped structures demonstrate exactly the same behaviour.

The experimentally observed temperature broadening of the far-IR transmission spectra (see fig 1, where the transmission data at 300 K are plotted with the dashed lines) proves again the lateral correlation in the distribution, because it clearly corresponds to redistribution of the holes between the dots with the heating of the structure.

**Figure 1.** Transmission spectra of doped samples with a nominal doping of 2 (a), 4 (b) and 6 (c) holes per dot at $T_L = 80$ K (solid lines) and $T_L = 300$ K (dashed lines). Scale is the same for all panels.

**Figure 2.** Photoinduced absorption spectra of the undoped structure (curve 1) and the structures with a nominal doping of 4 (curve 2) and 6 (curve 3) holes per dot at $T_L = 80$ K. Excitation intensity is the same for all curves and equal to 100 mW at a wavelength of 532 nm.
The photoinduced absorption spectra for the undoped structure and the two most heavily doped structures, measured at 80 K under the same excitation intensity, are shown in figure 2 as a ratio of the transmission change $\Delta T$ under interband photoexcitation and the equilibrium transmission $T$. In the case of photo-excitation, the capture of an additional hole into the QD does not change the charge state of the dot due to the simultaneous capture of an electron to the strain-induced 3D potential wells at the Si-side of the QD interface (see, for example, [9] for details on the Ge/Si QD interface band alignment and profiles). Thus, we effectively “turn off” the Coulomb interaction of the holes inside the dot. At the same time, the absorption spectra correspond exactly to the single-particle behavior discussed above. In the undoped sample, photoexcited holes occupy the dots with the lowest ground state energies (large dots). At the same excitation level, photoexcited holes in the doped sample occupy smaller dots with higher interlevel transition energies, so the photoinduced absorption peak widens to the short-wave-length region. Obviously, the extrinsic holes in doped samples do not contribute to the photoinduced change of the transmission, and we observe zero $\Delta T$ signal at $\hbar \omega < 30$ meV, where the equilibrium absorption exists.

4. Conclusion

In this work, we experimentally observe simultaneously a non-interacting holes’ response and a 0D multi-hole excitations’ response in far-IR equilibrium and photoinduced optical transmission spectra of Ge/Si quantum dot structures with size dispersion. The energy of the transition from the ground to one of the first excited states have been found to be independent on the number of holes in the multi-hole quasi-particle and equal to 30 meV. The qualitative differences of the hole distribution over the dot ensemble are indirectly registered in the absorption spectra. The temperature evolution of the few-particle excitation spectra proves the presence of the lateral correlation in the distribution of charge carriers over the dot ensemble.

Acknowledgements

This work was supported by grant of the President of the Russian Federation for young Candidates of sciences (MK-6064.2016.2) and Ministry of Education and Science of the Russian Federation (state assignment).

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] Grundmann M and Bimberg D 1997 Phys. Rev. B 55 9740
[4] Bras F, Boucaud P, Sauvage S, Fishman G and Gérard J.-M. 2002 Appl. Phys. Lett. 80 4620
[5] Tonkikh A, Zakharov N, Talalaev V and Werner P 2010 physica status solidi (RRL) Rapid Research Letters 4 224
[6] Sofronov A, Vorobjev L, Firsov D, Panevin V, Balagula R, Werner P and Tonkikh A 2015 Superlattices and Microstructures 87 53
[7] Kohn W 1961 Phys. Rev. 123 1242
[8] Hayrapetyan D B, Kazaryan E M and Sarkisyan H A 2016 Physica E 75 353
[9] Kurdi M El, Sauvage S, Fishman G and Boucaud P 2006 Phys. Rev. B 73 195327