Defense Technical Information Center
Compilation Part Notice

ADP013172

TITLE: Spatially Indirect Excitons in Self-Assembled Ge/Si Quantum Dots

DISTRIBUTION: Approved for public release, distribution unlimited

Availability: Hard copy only.

This paper is part of the following report:

TITLE: Nanostructures: Physics and Technology International Symposium [9th], St. Petersburg, Russia, June 18-22, 2001 Proceedings

To order the complete compilation report, use: ADA408025

The component part is provided here to allow users access to individually authored sections of proceedings, annals, symposia, etc. However, the component should be considered within the context of the overall compilation report and not as a stand-alone technical report.

The following component part numbers comprise the compilation report:
ADP013147 thru ADP013308

Spatially indirect excitons in self-assembled Ge/Si quantum dots

A. I. Yakimov, A. V. Dvurechenskii, N. P. Stepina, A. V. Nenashev
and A. I. Nikiforov

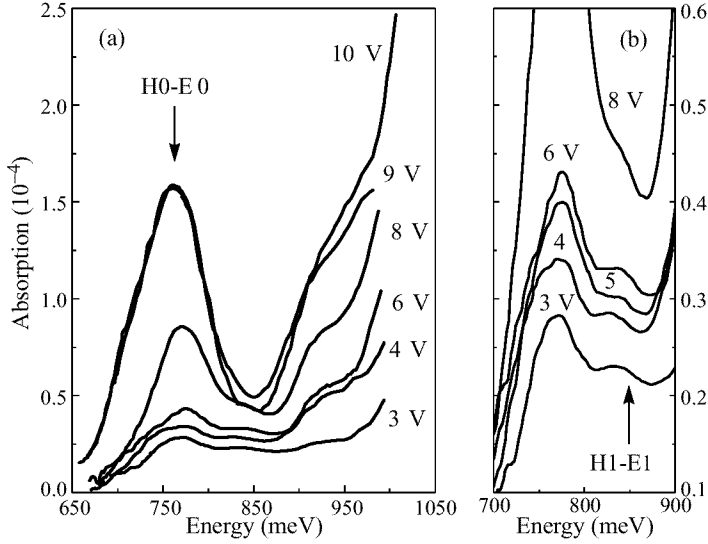Institute of Semiconductor Physics, 630090 Novosibirsk, Russia

Abstract. Using electron-filling modulation absorption spectroscopy, we study the effect of quantum dot charging on the interband excitonic transitions in type-II Ge/Si heterostructures containing pyramidal Ge nanocrystals. In contrast to type-I systems, the ground state absorption is found to be blueshifted when exciton-hole and exciton-exciton complexes are formed. For a positively charged dot, we argue that this is the consequence of dominance of the hole-hole and electron-electron interactions compared to the electron-hole interaction due to the spatial separation of the electron and hole. When two excitons are excited in the dot, the electrons are found to be spatially separated and have different single-particle quantization energies. This is the reason why the biexciton absorption is blushifted as compared to a single exciton.

The study of the excitonic properties in quantum-dot (QD) structures has drawn considerable interest in recent years. Most of the work has been reported in the type-I QD structures, where an electron and a hole are confined spatially in the same quantum well. Ge/Si(001) quantum dots exhibit a type-II band lineup. The large ($\sim$ 0.7 eV) valence-band offset characteristic of this heterojunction leads to an effective localization of holes in Ge regions, which represent potential barriers for electrons. When an electron-hole pair is photoexcited, the hole is captured by the Ge dot and creates a Coulomb potential resulting in a binding of an electron in the vicinity of the Ge dot. The spatially separated interacting electron and hole are usually referred to as 'spatially indirect exciton' [ ].

In this paper, we use electron-filling modulation absorption spectroscopy (EFA) to study effect of dot charging on the interband transitions in 10-nm-scale Ge/Si QDs. Ge dots are embedded into a n^+-p-p^+ Si diode, in which the number of holes in the QDs can be finely tuned by an external applied bias. When a state is occupied by a hole, no interband transition from this state is possible. When the hole is evacuated from the level, the interband transition is allowed. Modulating the holes in and out of the state by applying an ac bias voltage therefore induces corresponding changes in the infrared absorption. Thus the absorption signal measured under different bias conditions reflects directly properties of excitons at charged quantum dots.

The sample was grown by molecular beam epitaxy on a (001) oriented 4.5 Ω cm boron doped Si substrate. The Ge quantum dot layer with a nominal thickness of 10 ML was symmetrically embedded into a 1-μm thick p-Si region (B, 5×10^{16} cm^{-3}) at 300 °C. A buried back contact is formed by 50-nm B-doped p^+-Si (2×10^{18} cm^{-3}). The structure was finally capped with a 50 nm n^+-Si front contact (Sb, 1×10^{19} cm^{-3}). The dots are pyramidal with base orientation along [100] and [010] directions. The area density of the dots was estimated to be 3×10^{11} cm^{-2}. The average size of the dot base length was found to be about 15 nm, the height about 1.5 nm, and the dot uniformity approximately $\pm 20\%$.

To reveal the charge state of the dots we measure the capacitance-voltage characteristics. The dots are charged with holes at zero bias. The holes begin to escape at $V > 0.5$ V and the dots become totally depleted at $V > 8.5$ V. In the discussion that follows, we modulate
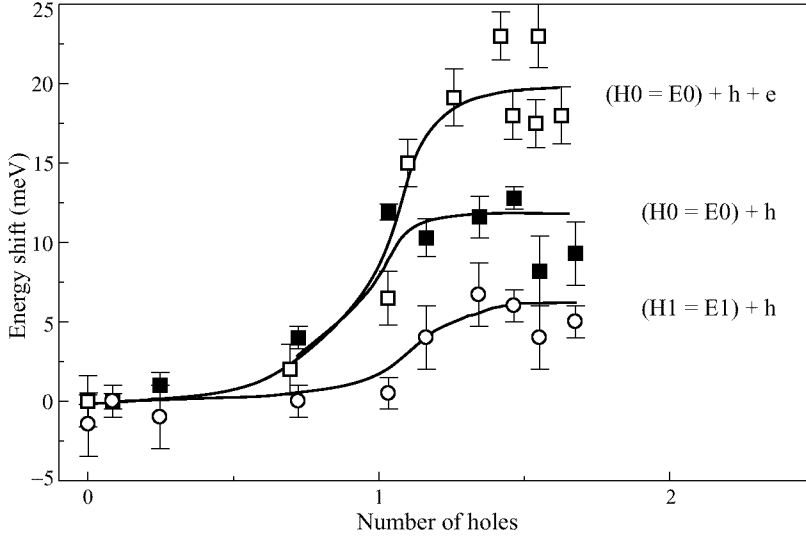
Fig. 1. (a) Room-temperature electron-filling absorption spectra at different reverse bias. (b) Expanded view of the spectra at low bias.

the reverse bias voltage between $V = 0$ V and $V = 2$–10 V with a frequency of 700 Hz. Differential aborption was measured at the voltage modulation frequency with the lock-in technique.

Figure 1 shows the EFA signal measured at different values of the bias V. Below the energy gap of Si, at energies 650–850 meV, we observe an absorption maximum with a broadening of $\sim$ 50–70 meV. An expanded view shows that this maximum can be well described by a sum of two Gaussian peaks. Symmetric line shape of the two peaks is characteristic of a bound-to-bound transition. We interpret the first absorption peak as an excitonic transition between the hole ground state (H0) in the Ge dots and the electron ground state (E0) confined in Si near the heterojunction. A second maxima at ≈ 860 meV is assigned to the excited-state excitonic transition (the H1–E1 transition). We assume that the broadening of the interband transitions is mainly due to the dispersion of the carrier confinement energies of dots with different sizes.

To obtain evidence to support the proposed origin of the EFA peak, we have studied the effect of additional interband optical excitation of the sample by a tungsten halogen lamp with a bandpass filter as source. When the sample is illuminated, nonequilibrium electrons and holes are photogenerated. The holes are captured by the dots while the electrons are accumulated near the dots forming the indirect excitons. At high pump intensities, the hole and electron ground states become fully occupied and the Pauli exclusion principle forbids the H0-E0 transition. We find that the experimental EFA signal is actually suppressed by the optical pumping.

One of the main results is that the excitonic transitions show a substantial stepwise blueshift with decreasing reverse bias. A qualitatively similar effect is seen with increasing the pump excitation density at fixed V. This result differs drastically from what has been observed for direct excitons, in which case charging leads to a redshift of the excitonic transition [ ]. From the oscillator strength (0.5) obtained in [ ] and the measured integrated absorption we calculate the number of holes per dot, N, at different biases in the dark and at different pump intensities. The energetic position of the excitonic transitions is shown in

Fig. 2. Shift of the H0–E0 (solid squares) and H1–E1 (open circles) transitions as a function of the hole occupation per dot. The open squares show the shift of the ground state transition at different pump intensities at fixed bias voltage ($V = 9$ V).

figure 2 as a function of N. It should be noted that the transition energy increases sharply when the first hole enters the ground state and then is approximately insensitive to further increase in the hole concentration.

When a H0–E0 exciton is created in a positively charged dot, an exciton-hole complex is formed consisting of two holes in the dot and an electron confined near the dot. There are two additional contributions to the energy of the exciton-hole complex as compared to e-h excitation in a neutral dot. The first is a positive Coulomb energy due to correlation between the two holes in the dot, E_{hh}, and the second is a negative contribution from the Coulomb attraction between the excited electron in the nearby silicon and the second hole on the dot, E_{eh}. Here we neglect the exchange interaction between the two holes since they have antiparallel spin orientation. For spatially direct excitons, the electron-hole interaction dominates and the resulting shift $\Delta E_{h-ex} = E_{hh} - E_{eh}$ is negative [ ]. Hence the expected reduction of the overlap factor for type-II excitons as compared with type-I systems yields a smaller magnitude of the electron-hole interaction energy E_{eh}. As a result, the energy of the exciton-hole interaction referenced to a neutral exciton energy can be positive. Taking the experimentally observed shift of 11 meV and $E_{hh} = 36$ meV [ ], the ground state exciton binding energy is estimated to be $E_{eh} = 25$ meV. Similar estimate gives $E_{eh} \approx 15$ meV for the exciton in excited state.

As can be seen from Fig. 2, optical pumping affects the transition energy more strongly than the bias voltage. This stems from the fact that illumination creates both holes and electrons while the field effect only induces holes in the dots. Under illumination we have two interacting excitons in the dot: the first is generated by the pump illumination, the second is excited by the infrared probing light. As compared to a single exciton, the transition energy now increases by $\Delta E_{ex-ex} = E_{ee} + E_{hh} - 2E_{eh}$, where E_{ee} is the energy of repulsive interaction between two electrons confined near the dot. For $\Delta E_{ex-ex} = 20$ meV, $E_{hh} = 36$ meV and $E_{eh} = 25$ meV, we obtain a suprising result $E_{ee} = 34$ meV. It is quite

improbable that E_{ee} could be so close to E_{hh} in a system where the hole states are more localized than the electron states. To resolve this problem we make self-consistent calculations of the expected electronic structure [ ]. The calculations show that the two electrons in the exciton-exciton complex are *spatially separated*. Electron-electron repulsion causes the second electron to localize below the dot base. As a result, the e-e interaction energy turns out to be only 19 meV, i.e., about two times less than the energy of the h-h interaction. Moreover, the single-particle energy of the second electron is larger than that for the first one, and the resulting shift of the absorption E_{ex-ex} turns out to be positive and equals to 10.2 meV.

References

[1] A. I. Yakimov, N. P. Stepina, A. V. Dvurechenskii, A. I. Nikiforov and A. V. Nenashev, *Phys. Rev. B* **63**, 45 312 (2001).

[2] R. J. Warburton, C. S. Dürr, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro and P. M. Petroff, *Phys. Rev. Lett.* **79** 5282 (1997).

[3] A. I. Yakimov, A. V. Dvurechenskii, A. I. Nikiforov and O. P. Pchelykov, *JETP Lett.* **68** 135 (1998).