JETP Letters, Vol. 73, No. 10, 2001, pp. 529–531. Translated from Pis'ma v Zhurnal Éksperimental'noĭ i Teoreticheskoĭ Fiziki, Vol. 73, No. 10, 2001, pp. 598–600.
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Spatial Separation of Electrons in Ge/Si(001) Heterostructures with Quantum Dots

A. I. Yakimov*, A. V. Dvurechenskiĭ, and A. I. Nikiforov

Institute of Semiconductor Physics, Siberian Division, Russian Academy of Sciences, pr. Akademika Lavrent'eva 13, Novosibirsk, 630090 Russia

** e-mail: yakimov@isp.nsc.ru*

Received April 10, 2001

It is shown experimentally that the excitation of interband optical transitions in arrays of Ge/*n*-Si(001) quantum dots leads to a decrease in the concentration of electrons in the conduction band. The phenomenon observed is due to the formation of negatively charged exciton complexes in Ge islands and represents the first experimental confirmation of the spatial separation of electrons in the silicon matrix surrounding the islands. *© 2001 MAIK "Nauka/Interperiodica".*

PACS numbers: 73.21.La; 71.35.-y

One of the promising methods for generating assemblies of quantum dots (QDs) is based on the use of kinetic effects in morphological changes at the surface of semiconductors during heteroepitaxy. At a large mismatch of the lattice constant, semiconductor layers will grow by the Stranski–Krastanov mechanism. The growth of a germanium film for the Ge/Si(001) system is first carried out layer by layer up to a thickness of four or five monolayers, and then a transition to the three-dimensional growth of pyramidal Ge nanoclusters (quantum dots) takes place. These nanoclusters provide partial relaxation of elastic strains in the heteroepitaxial structure [1].

The Ge/Si structures with Ge quantum dots relate to the second-kind heterojunctions in which electrons and holes reside in potential wells located on different sides of the heterojunction (Fig. 1), forming spatially indirect excitons. Such an unusual electronic configuration opens new possibilities both in fundamental research and for instrumental applications [2]. Here, the conduction band of one of the materials (Si) is close to the valence band of the other (Ge), which results in the spatial separation of carriers. In the given case, holes are localized in Ge nanoclusters, whereas there is a potential barrier for electrons in Ge. However, the localization of holes in Ge leads to a change in the behavior of the potential in the surrounding silicon matrix. A consequence of this change is the possibility of the formation of a potential well around Ge islands for electrons and the formation of electron states bound in this well [3]. The modeling of the electron structure of exciton complexes carried out in [3, 4] predicts that, on successively adding excitons to pyramidal Ge nanoclusters in Si(001), holes are concentrated in Ge in the vicinity of the base of a pyramid, and electrons are alternately localized in Si either in the vicinity of the apex of a Ge pyramid or in the vicinity of the boundary between Si and the wetting Ge layer (Fig. 1b). Though the physical nature of the spatial separation of electrons in silicon is clear (this separation minimizes the Coulomb repulsion of electrons), no experimental evidence of such an unusual electron configuration has been obtained so far.

In this work, the occurrence of the spatial separation of electrons presented in Fig. 1 has been confirmed experimentally. The idea of the experiment can be outlined as follows: It is known that the position of the chemical potential in the impurity band of doped semiconductors in the case of weak compensation is determined by the concentration of so-called 0- and 2-complexes [5]. If there is none of the ionized donors in the vicinity of a given acceptor, this object is named 0-complex. If two ionized donors are arranged on the opposite sides of an acceptor, this is a 2-complex. Shklovskiĭ and Éfros [5] showed that the formation of 2-complexes is favorable in energy; that is, a negatively charged acceptor can hold together two positively charged donors on its opposite sides. This statement can also be extended to the case of a charged exciton complex composed of a hole in a Ge island and two electrons located above and below the pyramid. Calculations in the self-consistent field approximation presented in [6] showed that, given the spatial separation of electrons, $N + 1$ electrons can be held in the vicinity of a Ge QD containing $N < 4$ holes, forming an artificial "ion."[1] Consider silicon of the n type containing Ge nanoclusters. In the case of interband illumination, electrons and holes are excited in pairs, a hole is captured in a Ge island, and an electron occupies the lowest

[1] Actually, the valley degeneracy factor in Si was not taken into account in [6]. Taking into account this factor leads to the conclusion that an "excess" electron can be held at $N < 8$.

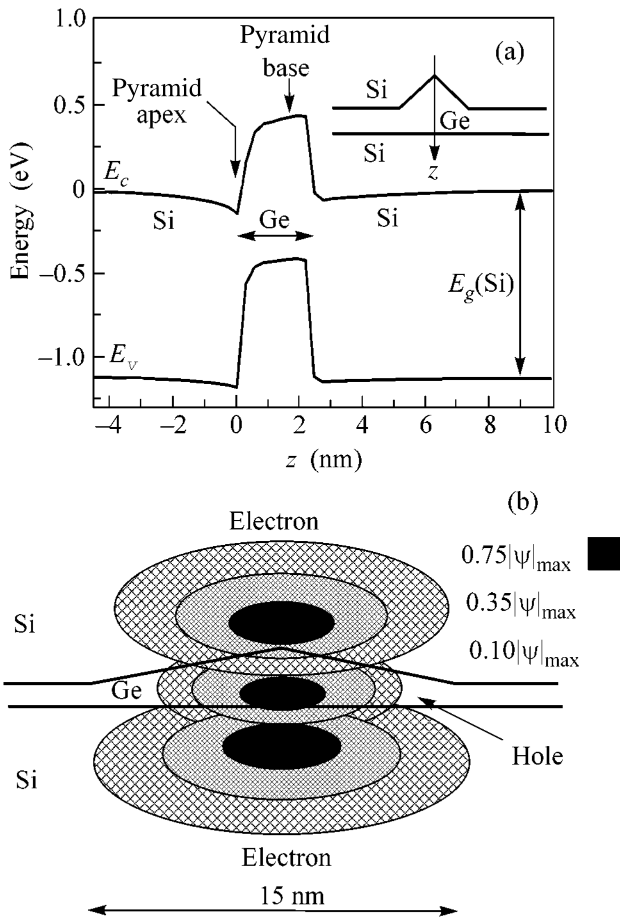

Fig. 1. (a) Calculated profile of conduction and valence bands along the z axis passing through the vertex of a Ge pyramid [4]. (b) Calculated structure of the wave functions of two electrons and a hole localized in the vicinity of a Ge pyramid [3]. Gradations of gray color correspond to regions at whose boundaries the wave functions drop down to levels of 75, 35, and 10%. The pyramid resides on an underlying continuous Ge layer and surrounded by silicon on all sides (see the inset in (a)). The pyramid base is 15 × 15 nm in size. The calculation was performed with allowance made for the nonuniform distribution of elastic deformations in the heterosystem within the Hartree approximation [3].

bound state in energy in the conduction band of Si in the vicinity of the apex of a germanium pyramid. Because one hole can hold two electrons at a QD, an equilibrium electron will be captured in the potential well located under the island, and the concentration of electrons in Si will decrease under conditions of illumination. Note that the concentration of free carriers usually increases upon illumination. Therefore, measuring experimentally the change in the concentration of free electrons under conditions of interband illumination leads to the conclusion that the electron configuration presented in Fig. 1 exists. Previously, indirect arguments about the photoinduced localization of equilibrium electrons in the potential of nonequilibrium holes were given in [6].

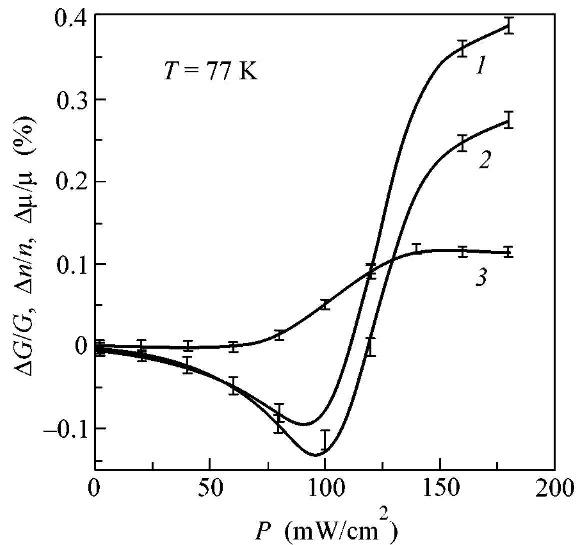

Fig. 2. Dependence of (*1*) the relative photoconductivity, (*2*) the relative change in the concentration of electrons in the Si conduction band, and (*3*) the Hall mobility on the intensity of interband illumination.

To verify the notions outlined above, we investigated epitaxial Ge/Si structures grown on a Si(001) substrate with a phosphorus concentration of $N_s \approx 10^{15}$ cm^{-3}. The samples consisted of ten layers of Ge islands separated with Si spacers 30 nm thick. The concentration of donors (Sb) in the epitaxial Si layer was 3×10^{16} cm^{-3}. Ge nanoclusters were shaped as pyramids with an average size of the base of 15 nm and a height of 1.5 nm. The layer density of nanoclusters in each layer was $\approx 3 \times 10^{11}$ cm^{-2}. A GaAs light-emitting diode (LED) whose radiation maximum was at a wavelength of ~0.9 μm was used as the light source. The intensity of the LED radiation was modulated with a frequency of 2 kHz. The photoconductivity (PC) and the Hall coefficient were measured by the van der Pau method at the modulation frequency using a lock-in nanovoltmeter. This allowed small variations of the resistance and the Hall emf to be measured upon illumination of the structures. In order to exclude the illumination of contacts, which may result in a spurious PC [7], the contacts themselves and the region around the contacts were protected with a nontransparent coating.

Experimental curves of the relative PC $\Delta G/G$, the relative change in the concentration of electrons $\Delta n/n$, and the relative change in the Hall mobility $\Delta\mu/\mu$ are shown in Fig. 2 as functions of the illumination power P at $T = 77$ K. In accordance with the notions outlined above, a decrease in the concentration of electrons is actually observed upon illumination in the range $P =$ 0–100 mW/cm^2. This decrease is accompanied with the appearance of a negative photoconductivity. The mobility remains virtually unchanged at $P < 70$ mW/cm^2 and weakly increases at more intense illumination. The growth of mobility is apparently associated with the

fact that a positively charged nucleus cannot hold an "excess" electron as the number of holes in the QD increases. Therefore, scattering from "ions" (N holes + $(N + 1)$ electrons) gives way to weaker scattering from dipoles (N holes + N electrons). The decrease in the binding energy of the "excess" electron in the field of N holes at large N can also be explained by the change in the sign of Δn and ΔG at $P > 100$ mW/cm^2.

Thus, it was found that the concentration of electrons in the conduction band of Ge/Si(001) heterostructures with germanium quantum dots decreases upon illumination that leads to interband transitions. The reason for this behavior is the formation of negatively charged "ions." This possibility of this formation is due to the spatial separation of electrons at Si/Ge heteroboundaries.

This work was supported by the Russian Foundation for Basic Research, project no. 00-02-17885.

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Translated by A. Bagatur'yants