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Localization of electrons in dome-shaped GeSi/Si islands

A. I. Yakimov,[1,2,a)] V. V. Kirienko,[1] A. A. Bloshkin,[1] V. A. Armbrister,[1] P. A. Kuchinskaya,[1] and A. V. Dvurechenskii[1]
[1]*Rzhanov Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Science, prospekt Lavrent'eva 13, 630090 Novosibirsk, Russia*
[2]*Tomsk State University, 634050 Tomsk, Russia*

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We report on intraband photocurrent spectroscopy of dome-shaped GeSi islands embedded in a Si matrix with n^+-type bottom and top Si layers. An in-plane polarized photoresponse in the 85–160 meV energy region has been observed and ascribed to the optical excitation of electrons from states confined in the strained Si near the dome apexes to the continuum states of unstrained Si. The electron confinement is caused by a modification of the conduction band alignment induced by inhomogeneous tensile strain in Si around the buried GeSi quantum dots. Sensitivity of the device to the normal incidence radiation proves a zero-dimensional nature of confined electronic wave functions. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4906522]

Ge/Si quantum dots (QDs) formed by strain-driven epitaxy are of great interest due to their compatibility with the well-developed Si readout circuitry. Ge QDs in Si matrix form a type II band alignment. The large ($\sim$0.7 eV) valence-band offset characteristic of this heterojunction leads to an effective localization of hole in Ge regions, which represent potential barrier for electrons. Most of the demonstrations of electronic and optical phenomena associated with zero-dimensional nature of carrier states were achieved with *p*-type Ge/Si QD heterostructures. So far, little work has been done for the *n*-type Ge/Si QDs which are especially important for realization of spintronics and quantum computation. The simple consideration of energy bands disregards possible modification of the band structure due to elastic strain in the dots and the surrounding matrix. However, the strain fields appear due to the $\sim$4% lattice mismatch between Si and Ge. The strain causes splitting of the sixfold-degenerate Δ valleys into the fourfold-degenerate in-plane Δ_{xy} valleys and the twofold-degenerate Δ_z valleys along the [001] growth direction in Si. As it has been shown in calculations for Ge hut-shaped islands, tensile and compressive strain fields induced by the dots in the nearby Si can lead to the formation of three-dimensional (3D) potential wells for electrons both near the QD apexes[1–3] and close to the dot base edges.[2,4] The most favorable place of spatial electron position is determined by the shape, size, and chemical composition of the dots. The experimental evidence for the electron localization in Ge/Si heterostructures with small Ge huts has been obtained by using the photocurrent (PC) spectroscopy[3] and electron spin resonance technique.[4] The typical lateral size of the Ge huts was about 20 nm, the dot density equals to $\sim 10^{11}$ cm^{-2}, and the average Ge fraction was close to 90% (Ref. 3).

Depending on the substrate temperature and the amount of deposited Ge, different island morphologies can be observed. The smallest facetted Ge islands are square-based pyramids[3] or rectangular-based huts[5–8] which are formed at low growth temperatures ($\leq$550 °C) and at small Ge coverage. At temperatures higher than 600 °C, the majority of the islands are dome-shaped with decreased island density, increased both the island size[9,10] and the island aspect ratio.[11] From the point of view of applicability of the system in quantum computations, the dome-shaped QDs have two main advantages with respect to the Ge huts. First, the density of the domes is by two or three orders of magnitude less than density of the huts[12] providing selective access to individual QDs for implementation of one- and two-qubit operations. Second, characteristic island size distribution for dome-shaped QDs is known to be much smaller than that for the huts,[13] thus giving possibility to fabricate uniform arrays of logic gates. The question is whether the 3D electron confinement can be achieved in dome-shaped GeSi islands. The point is that the dome clusters are formed at elevated substrate temperatures and, therefore, the average Ge content of the islands is much less, 100%, resulting in a reduced conduction band offset between Si and GeSi and in a smaller strain in the Si barrier compared to Ge-reach hut clusters.

Rezaev *et al.*[14] have analyzed the effect of GeSi island form and composition on the electron confinement in the Si regions surrounding QDs. For GeSi dome-shaped clusters with the dome-width of 87 nm, dome-height of 19 nm, and realistic Ge distribution (all numerical parameters have been taken from Ref. 10) the ground-state electron binding energy as large as 120 meV has been evaluated. Although the average Ge fraction in this structure is only 47%, the wide spread of strain field in Si due to the high aspect ratio of domes gives rise to a broad potential well for electrons and provides a deep confined level. The very important finding of Ref. 14 is that upon a truncation of the island and consequent height reduction during the capping process, the electron localization energy is rapidly decreased and tends to zero. Thus the essential point for successful experimental verification is to preserve the GeSi island size during formation of Si capping layers. The Si overgrowth at temperatures close to the Ge growth temperature results both in Ge–Si intermixing and in changing of island size and shape,[15–19] in particular, due to

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[a)] yakimov@isp.nsc.ru

dissolving and removal of the Ge-rich dot apex.[11] A way to circumvent this detrimental phenomenon has been proposed by Stoffel *et al.*[17] and Wu *et al.*[18] It has been demonstrated that atomic processes associated with the island shape changes are kinetically limited at a temperature bellow 400 °C (Ref. 18). This means that no significant change in size and shape of domes is expected after Si capping below this temperature. The reduction of the Si overgrowth temperature down to 400 °C is the key technological approach employed in our work to obtain buried dome-shaped GeSi/Si islands with the robust electron confinement.

In this letter, we report a far-infrared PC spectroscopy study of GeSi dome-shaped QDs embedded in a Si matrix. For the experimental work, we choose the same growth conditions as in Ref. 10 to fabricate just the same GeSi/Si islands that have been analyzed theoretically in Ref. 14. Figure 1(a) shows schematically the structure of the sample discussed in this paper. The sample was grown by solid source molecular beam epitaxy on a (001) oriented arsenic doped n^+-Si substrate with resistivity of 0.005 Ω cm. The active region of the device was composed of seven stacks of undoped GeSi islands separated by 60 nm Si barriers. Each GeSi QD layer consisted of a nominal Ge thickness of 6.5 monolayers (ML) and formed by self-assembling in the Stranski-Krastanov growth mode at 620 °C and at a growth rate of 0.01 nm/s. The Si spacer layers were deposited at a temperature of 400 °C, with the temperature ramps before and after QD growth. Cross-sectional image obtained by transmission electron microscopy (TEM) clearly demonstrates that the most part of the GeSi islands exhibit a high correlation along the growth direction (Fig. 1(b)). Similar to the case investigated by Thanh *et al.*,[20] the vertical ordering already occurs for the second deposited layer, demonstrating good structural quality and the planarity of the Si cap layers (Fig. 1(c)). The multilayer heterostructure was sandwiched in between the 100-nm-thick intrinsic Si buffer and cap layers fabricated at 620 °C. Finally, a 50-nm-thick n^+-Si top contact layer with antimony doping of $\sim 10^{19}\,\mathrm{cm}^{-3}$ was deposited. In such *n–i–n* device, the electrons in the active QD region are contributed from the highly doped contacts layers.[21,22] The average Ge content of about 50% in the islands was determined from Raman scattering experiments (Fig. 2) using an approach described in Ref. 23.

Surface morphology was controlled by atomic force microscopy (AFM) (Fig. 1(d)). At the growth temperature used, dome shaped islands are commonly observed.[9,10] The dots are about 18 nm in height and 84 nm in diameter with the standard deviation ±5%. The QD areal density is $3 \times 10^9\,\mathrm{cm}^{-2}$. The island form, diameter, height, composition, and density are in a good agreement with the data published in Ref. 10.

Photoluminescence (PL) measurements were carried out at $T = 5\,\mathrm{K}$ using a Bruker Vertex 70 Fourier transform

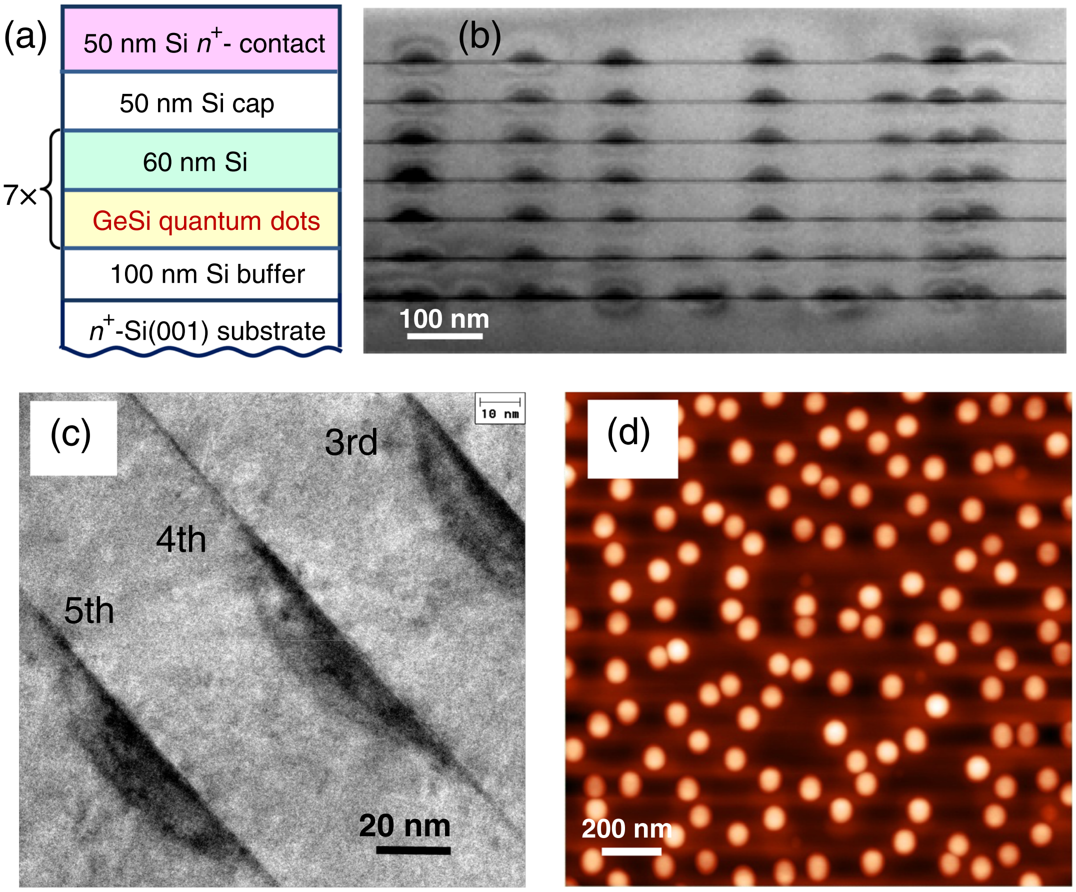

FIG. 1. (a) Layer sequence of the GeSi/Si heterostructure. (b) Cross-sectional TEM micrograph of vertically aligned GeSi QDs. (c) High-resolution TEM image of GeSi dome clusters in the third, fourth, and fifth Ge layers capped with Si at 400 °C. (d) AFM image ($2\,\mu\mathrm{m} \times 2\,\mu\mathrm{m}$) of the topmost uncapped Ge layer.

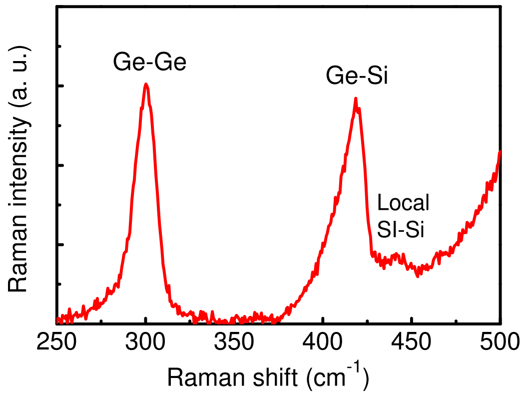

FIG. 2. Room-temperature Raman spectrum of the sample under study.

infrared (FTIR) spectrometer in a rapid scan mode. The FTIR system uses a calcium fluoride beamsplitter and a room temperature InGaAs detector with cutoff at 0.7 eV. The individual PL spectra were corrected with respect to the spectral sensitivity of the InGaAs detector. PL excitation was provided by a 532 nm line of a diode yttrium aluminium garnet (YAG) laser, under an excitation power of 100 mW. For vertical PC measurements the samples were processed in the form of circular mesas with diameter 3 mm by using wet chemical etching and contacted by AuSb metallization. The normal-incidence photoresponse was obtained using a FTIR system with a spectral resolution of 5 cm^{-1} equipped with a KBr beamsplitter and a globar light source operating in a step-scan mode along with a SR570 low noise current preamplifier and a SR850 lock-in amplifier. The PC spectra were calibrated with a DLaTGS detector. The device was mounted in a cold finger inside a helium flow cryostat (Oxford Optistat CF-V) with ZnSe windows.

The 5 K PL spectrum from the sample is shown in Fig. 3. A group of narrow PL lines at 1117, 1105, and 1058 meV are assigned to the no-phonon (NP) and transverse optical (TO) phonon replicas in the heavily doped Si substrate and undoped Si epilayers.[24] The two PL lines at 985 and 928 meV originate from the NP and TO phonon replica of the wetting layer. The broad PL band located around 0.75 eV is attributed to the recombination of holes localized in the Ge islands and electrons confined within the tensile-strained Si around the dots. It seems interesting that the entire spectrum from multilayer heterostructure reproduces well the single-layer spectrum from Ref. 25, thereby giving evidence for a good size uniformity of GeSi nanoislands in different layers.

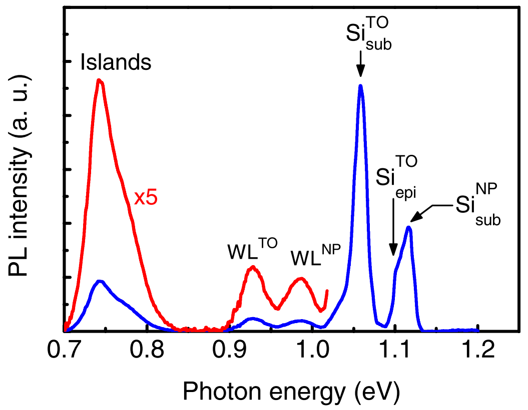

FIG. 3. PL spectrum of a sevenfold stack of 6.5 ML GeSi dots separated by 60 nm Si.

Figure 4 shows the normal incidence photocurrent spectra of the heterostructure under study. The sample exhibits a broad photoresponse ranging from 85 to 160 meV. The broad nature of the photoresponse and the asymmetric PC line shape with the flat slope on the high energy side suggest that the photocurrent could be due to a bound-to-continuum transition.[26–30] Two distinct PC peaks is observed at ∼100 and ∼130 meV, which we attribute to transitions from two occupied electron states to the unstrained Si continuum. The position of the high-energy peak agrees well with the ground-state electron binding energy $E^{\mathrm{D}}_{\mathrm{s1}} = 120$ meV (Ref. 14). The ground state is twofold degenerate; the appropriate electron wave function has an *s*-like spatial symmetry and is concentrated in Si near the dome apex. To estimate the average number of electron confined near each QD $\langle N_e \rangle$, we used a simple model described in detail in Ref. 31. The calculations were based on solving the system of the Poisson and electroneutrality equations as well as on statistical distribution of electrons over the energy levels in the system under study. For the chosen doping concentration and the heterostructure layout, we find $\langle N_e \rangle \approx 6$. It is expected that the first excited state is a *p*-like fourfold-degenerate state with the energy level $E^{\mathrm{D}}_{\mathrm{p}} = 100$ meV below the continuum.[14] Thus, we may conclude that the first excited state is occupied with about four electrons, and the peak at 100 meV likely comes from a bound-to-continuum transition with participation of the $E^{\mathrm{D}}_{\mathrm{p}}$ electron state. The twofold difference in the amplitude of the two peaks can be explained by the different occupation of the $E^{\mathrm{D}}_{\mathrm{s1}}$ and $E^{\mathrm{D}}_{\mathrm{p}}$ levels.

In summary, we have investigated the intraband photocurrent within the conduction band of dome-shaped GeSi/Si islands. The Si overgrowth at temperatures as low as 400 °C was used to reduce Ge-Si intermixing and to preserve the island shape and size from the effect of a further high temperature deposition. The fabricated islands are about 18 nm in height and 84 nm in diameter, the average Ge fraction in the

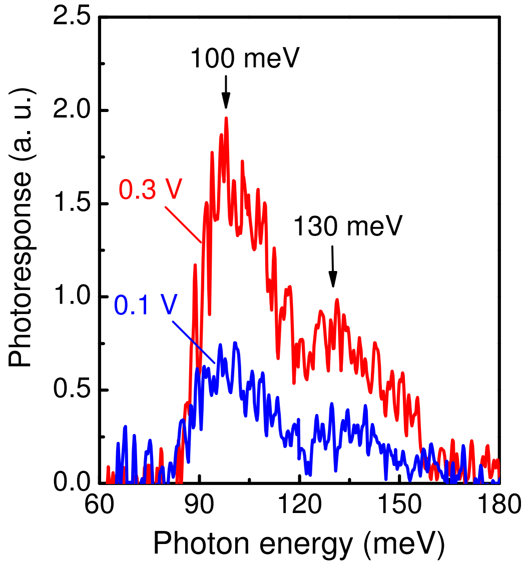

FIG. 4. Photocurrent spectra measured under normal incidence infrared radiation at $T = 5$ K and applied bias of 0.1 and 0.3 V.

dots is about 50%. A normal incidence photoresponse in the 85–160 meV energy region has been observed and ascribed to the optical excitation of electrons from states confined in the strained Si nearby the dot apexes to the continuum states of unstrained Si. The observed peak positions in the PC spectra agrees well with the theoretical results. The ability of the device to operate in the normal incidence mode can be regarded as a proof of the zero-dimensional nature of detected electron states, unlike *n*-type two-dimensional systems which are not sensitive to in-plane polarized radiation.[32]

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