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QUANTUM EFFECTS IN MULTILAYER Si-Ge NANOHETEROSTRUCTURES

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The lateral photoconductivity spectra and photofield electron emission were used to investigate multilayer Ge/Si heterostructures with Ge quantum dots. Earlier we have revealed a close connection between elastic strain in Ge quantum dots originating due to the lattice mismatch during the epitaxial growth and additional energy level forming in the strained Si-Ge heterojunction region. Thus, it appeared to be possible observing intraband transitions in Ge quantum dots that are absent in two-dimensional Si-Ge heterostructures using such simple and informative methods. While an influence of the number of Ge quantum dot layers on lateral photoconductivity spectra is not essential, the photofield electron emission characteristics showed considerable shift to middle infrared area, as the number of Ge quantum dot layers increased. It was revealed that size and composition parameters of Ge quantum dots correspond to energy levels in the valence band of the latter with the energy distance between them about 0.32 and 0.34 eV with a high accuracy. The results of our investigation make it possible to expect their possible application in new nano- and optoelectronic devices.

Introduction

With the progress in nanotechnologies new possibilities were opened for easy creation of nanosize objects, in particular Ge-Si heterostructures with Ge quantum dots, which attract much attention of researchers all over the world as systems that allow introduction of band gap engineering into silicon technology. This opens promising ways for the development of new Si-based high-frequency and optoelectronic devices, for instance photoemission detectors for infrared (IR) range that have been already elaborated [1 – 3]. Due to their geometry, ensembles of nanosized Ge quantum dots make possible the realisation of high electric fields ~ 10^7 V/cm near the dot apex at moderate voltages ~10^3–10^4 V, which causes an intense field electron emission.

The field emission from semiconductor cathodes was widely investigated by several research groups beginning from 1960's [4 – 8]. To our knowledge, the first field emission measurements from Ge nanoclusters grown on Si were reported in 2000 by Tondare et al. [9]. The first works on this system recorded no peculiarities in the current-voltage (*I*-V) curves of the field emission. We also investigated such systems [10, 11] and revealed interesting features – reproducible peaks of current in the *I*-*V*-curves of the field emission from Ge nanocluster structures on Si(100). The number of the peaks was found to depend on the cluster size. We connected the peaks with the effects of energy quantization in the Ge quantum dots. Moreover, the field emission current showed a considerable photosensitivity in the wavelength range from 0.4 to 10 μm. For the photodiodes that were used, it was found that the field emission current increased at room temperature by a factor of 5 to 3 under the irradiation by

light with the wavelength of 2 and 10 μm, respectively [12]. Besides that, lateral photoconductivity spectra from the same structures that we investigated independently revealed photocurrent in the mid-infrared range caused by optical transition from localized states in the valence band of QDs [13, 14].

It should also be noted that considering photoelectric and electronic properties of Ge-Si heterostructures with Ge quantum dots one has to take into account the morphology of such systems. They are generally referred to the second type heterostructures, in which holes are localized in the quantum dots, while electrons are supposed to be free in the conduction band. A large number of experimental works were devoted to investigation of the energy-band structure in a quantum dot. It is supposed that the positions of energy levels in a quantum dot are determined by its size, shape and composition, as well as the value of inhomogeneous strain inside it caused by the lattice mismatch. Morphological investigations may make possible the optimization of epitaxial conditions and growth mode of the heterostructure formation [15]. Another important parameter that essentially influences optical and photoelectric properties of Ge/Si heterostructures is the value of the valence band offset. The average valence band offset of $Si/Si_{1-x}Ge_x$ heterojunction was estimated as $0.54 \cdot x$ (eV), where x is Ge concentration. This value describes rather accurately the properties of heterojunctions of different compositions. The significant valence band offset between silicon and $Si_{1-x}Ge_x$ alloys determines a wide range of spectral sensitivity variation of photodetectors with the SiGe quantum dots [16].

We present here our experimental results on lateral photoconductivity and photofield electron emission from multilayer Ge/Si heterostructures with Ge quantum dots. They seem to reveal quantum size effects which mirror the nature of electron transport in such structures. Thus one of the main purposes of the present work was to search for a correlation between the quantum regularities of the photofield electron emission and the lateral photoconductivity in Ge/Si heterostructures with Ge quantum dots.

Experimental methods

The molecular beam epitaxy (MBE) technique ("Katun'-B" set-up, produced in Novosibirsk, Russia) was used to prepare multilayer Ge-Si(100) nanocluster arrays with the islands of various sizes and surface density. The (100) oriented wafers of n-Si with 7.5 and 20 Ohm·cm resistivity and diameter of 76 mm were used as substrates. In order to prepare multilayer quantum dot systems with regular nanoisland distribution over the substrate surface, we had earlier proposed to use a system of $Si_{1-x}Ge_x$ intermediate layers with a sub-critical thickness [11]. The Ge mole fraction x was gradually increased from layer to layer grown at gradually decreasing substrate temperature started from T_s=500 °C. The growth process, in particular the moment of the $2D \rightarrow 3D$ transition in the Stranski-Krastanov growth regime, was controlled *via* RHEED (reflection high energy electron diffraction). To study the surface morphology, atomic force microscopy (AFM) measurements were carried out using an MFP-3D AFM from Asylum Research with a closed loop scanner. Standard Si cantilevers with tips having a half opening angle of 10° were employed as probes. Fig. 1 represents an AFM image of the top nanoisland layer of the investigated samples. It shows that the nanoclusters are shaped as tetrahedral pyramids with the base about 40 nm and height about 2 nm. The average cluster density was about 10^{10} cm^{-2}. The growth of each Si intermediate layer was continued until a high-contrast Si(100)2×1 RHEED pattern was produced typical of clean Si. Thus, the multilayer Ge-Si(100) nanocluster arrays with three, five and eight layers of SiGe quantum dots alternated by Si layers of the thickness about 3.5 nm were grown at the temperature T_s=500 °C. The Ge fraction in the investigated structures was estimated by secondary ion mass-spectroscopy (SIMS) technique. This technique was applied to monitor sequentially the Ge fraction within the Si/Ge layers with QDs in the growth direction with a step of 1 nm. Taking

into account the value of QD surface density and the average size of QDs, the average Ge fraction in the QD ensemble was estimated at the range from 75 to 85 %.

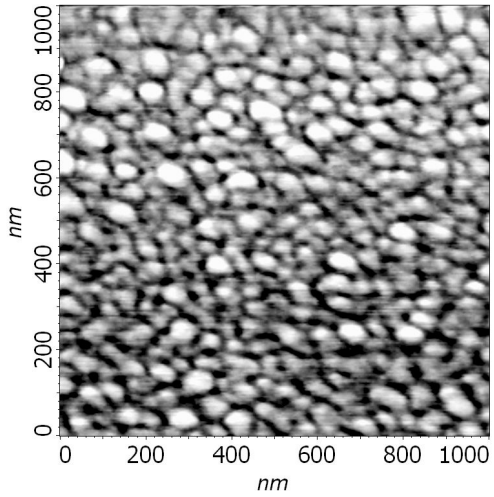
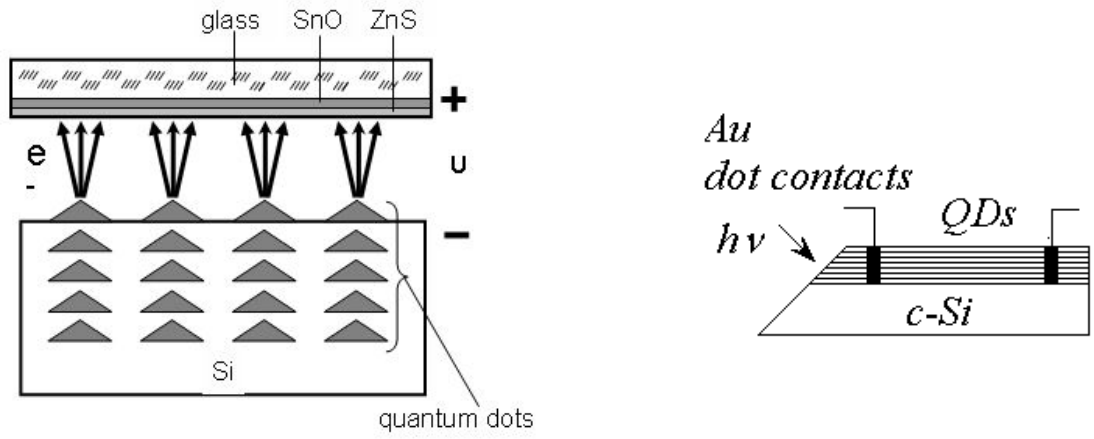


Fig. 1. A 1×1 μm^2 AFM image of the QDs grown at 500 ^{o}C. The average height and base diameter of dots are about 2 nm and 40 nm, respectively.

The measurements of the field and photofield electron emission were carried out in a diode cell, in which the distance between the electrodes was 300 μm (fig. 2, *a*). In order to visualize the spatial emission distribution, a ZnS cathodoluminescent screen was deposited onto a glass plate having a SnO_2 conducting layer which served as the anode. The details of the experiment were presented in [12].



a *b*

Fig. 2. (*a*) Diode cell for measurements of the field and photofield electron emission. (*b*) Sample for photoconductivity measurements.

To investigate lateral photoconductivity of SiGe quantum dot multilayer structures, two Au ohmic contacts 1 mm in diameter and placed 10 mm apart were fused into the surface with the SiGe QD layers [14] (fig. 2, *b*). As a result, the contacts were brought to each QD layer and to Si substrate. The conduction current-voltage characteristics of all the investigated structures were linear in the voltage range from U = –50 V to +50 V at the temperatures 77 – 290 K. Spectral dependences of lateral photoconductivity were measured using infrared spectrometer in the $h\nu$ range from 0.3 to 1.1 eV at U = 5 V. The photocurrent was measured using a current amplifier and the standard lock-in detection technique. The measured spectral dependences were brought to a constant quantum quantity using a nonselective pyroelectric receiver.

Results and discussion

Field and photofield electron emission

The investigated structures showed a stable field emission at T=290 K and a *macroscopic* electric field ~ 10^5 V/cm. It should be kept in mind that that the local electric fields, which render the surface potential barrier sufficiently transparent to ensure the field emission, are much higher (~10^7 V/cm). It is difficult to give their accurate evaluation, since they are determined by details of geometrical and compositional heterogeneities on the nanometer scale [17]. Curve 1 in fig. 3 is an *I-V* characteristic of photofield emission from a heterostructure with SiGe quantum dots of the height ≈ 2 nm. The sample was irradiated with a 10 Wt light halogen lamp placed at 20 cm from it. The dark field emission current measured without irradiation was several orders smaller (fig. 3, curve 2). Neither field emission nor photofield emission current was recorded for the Si samples without quantum dots in the investigated voltage range 2.0 – 4.3 kV. It is thus supposed that the observed photo-induced current is produced by emission of electrons from the quantum dots.

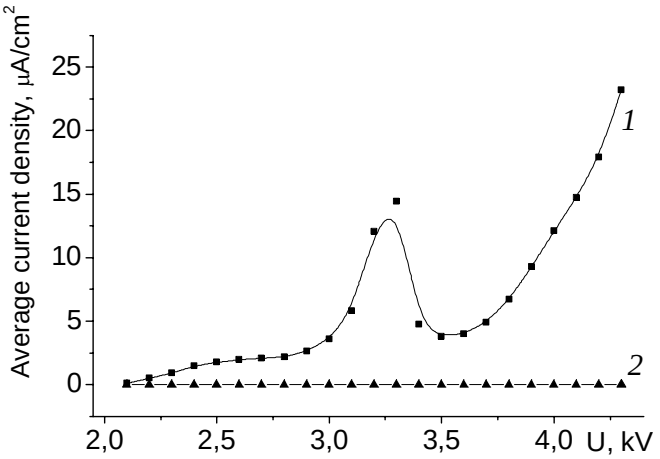


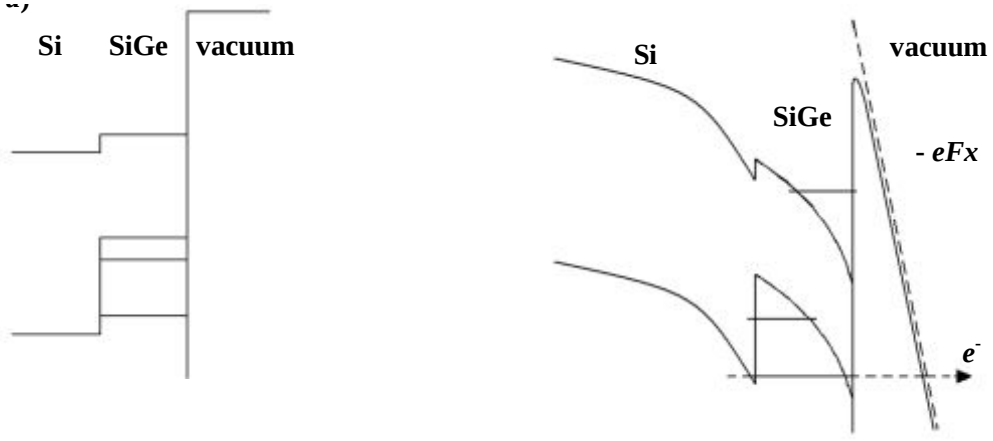
Fig. 3. (*1*) The current-voltage curve of photofield emission from a Si/Ge heterostructure with 8 layers of quantum dots of the height ~3 nm. (*2*) The dark field emission current-voltage for the same heterostructure.

The particular sample whose *I-V* curves are given in fig. 3 showed practically no dark current in the voltage range studied. However, in our previous works[11] we presented results recorded for other Si/Ge quantum dot heterostructures which did generate rather intense field emission currents in the dark (~10^{-7} to 10^{-5} A at the anode voltages of ~10^2 to 10^4 V and the total sample area of ~10^{-1} cm^2). It was found [11] that the structures with the quantum dots ≈ 3 to 5 nm high had the field emission current-voltage curves with a few distinct maxima (peaks). The numbers of the peaks in the field emission *I-V* curve increases with QD size, so that eventually no clear current peaks can be observed for the QDs higher than ≈ 10 nm. This effect was attributed to resonant electron tunneling through quantized energy levels in the dots.

We suppose that the current peak in the *I-V* curve of photofield emission (fig. 3, curve 1) may also be connected with energy quantization, in particular the presence of discrete energy levels in the valence band of the quantum dots. In the absence of external electric field, Si/Ge heterojunction is referred to the second type heterostructures, in which a potential well is formed for one type of carriers. Namely, the valence band of Ge nanoislands is a potential well for holes causing a localization of states. Possible energy-band diagrams for Si/SiGe heterojunction in the absence and presence of electric field are suggested in fig. 4, *a* and fig. 4, *b*, respectively. It should be stressed that the potential well near the surface is in any event highly asymmetric. Besides, the shape and transparence of the barrier Si/QD/vacuum

depends drastically on the strength of applied electric field. That is why the considered Si/QD/vacuum "heterostructure" can neither be unambiguously classified as a first type nor as a second type heterojunction. We have thus to treat it as a special type of heterojunction whose properties depend upon the value of the applied electric field and require a separate approach.

In particular, the shape and width of the potential well for holes in the valence band of SiGe nanoislands changes essentially when the electric field is applied to the surface. As a consequence, the discrete energy values E_n change too. At some value of the applied electric field, the energy position of a quantization level may coincide with the top of the Si valence band, so that resonance tunneling of electrons can become probable from this band into vacuum *via* the energy quantization level in the SiGe quantum dot (fig. 4, *b*). As the electric field is increased further, the potential well becomes shallower and resonance tunneling can proceed *via* the next energy level. The observation of a current peak for the structure with the QDs about 2 nm high may indicate that at least one localized energy state exists in the valence band of SiGe nanoisland in the absence of electric field.



a b

Fig. 4. An energy-band diagram of Si/SiGe heterojunction without (*a*) and in the presence of electric field F (*b*).

In our previous works [11], we considered another potential diagram for the field emission from Si/Ge dots corresponding to the case of a strong downward band bending and resonance tunneling through the discrete energy levels that can arise in the QD conduction band. It should be admitted that the real potential configuration near the QD surface still remains undetermined due to its complex dependence on a number of poorly evaluated factors, such as exact QD geometry, composition and strain distribution, field penetration, the nature and properties of surface states.

The essential increase of field emission current under the irradiation observed for the structures with Ge quantum dots may be caused by optical transitions in the quantum dots involving energy quantization levels. In low-dimensional Si/Ge heterostructures with Ge quantum dots, there exists a possibility of electronic transitions in near infrared range from energy quantization levels in the valence band of Ge quantum dots into the conduction band of Ge quantum dots and conduction band of Si surrounding [18].

An Estimation of the energy level positions in the valence band of Ge nanoislands

The number of energy levels in the valence band of Ge nanoislands depends on Ge nanocluster size, shape, elastic strain value and the corresponding potential well depth. When treating the QD approximately as a box with a finite confinement potential, with a thickness L_z and lateral dimension $L_x = L_y$, the energy level $E_{nml} = E_{nx} + E_{my} + E_{lz}$ and the wave

function $\psi_{nml} = \psi_{nx}\psi_{my}\psi_{lz}$ are three-dimensional. The corresponding energy values E_{nx}, E_{my}, E_{lz} and functions ψ_{nx}, ψ_{my}, ψ_{lz} are the solutions of one-dimensional equations. If one considers the simplest model of a rectangular finite potential well, the position of energy levels is determined by solving the transcendental equation [19]:

$$KL = n\pi - \arcsin\left(\frac{K_n}{G}\right), \tag{1}$$

where $K_n = \sqrt{2m_h^* E_n}/\hbar$, $G = \sqrt{2m_h^* V_0}/\hbar$, $n = 1,2,3...$, m_h^* is the effective mass of holes in the potential well with the depth V_0, and L is the width of the potential well. It should be noted that this estimation is approximate, since the real shape of Ge nanoislands, as well as their inhomogeneous composition are not considered.

The depth of potential barriers is taken equal to value of valence band offset. It is generally assumed that the valence band offsets for heavy and light hole states of the strained Ge/Si heterojunction depend linearly on the Ge mole fraction and residual elastic strain values ε_{xx} and ε_{zz}. Uniaxial strains entail lifting of the valence band degeneracy in the SiGe nanoislands. As a result, light and heavy hole states of the valence band are shifted in opposite directions, and the shifts can be determined using the relation for the average valence band offset ΔE_v^{av} in the following way [20]:

$$\Delta E_{LH} = -\frac{1}{6}\Delta_0 + \frac{1}{4}\delta E + \frac{1}{2}\left[\Delta_0^2 + \Delta_0\delta E + \frac{9}{4}(\delta E)^2\right]^{1/2}, \tag{2}$$

$$\Delta E_{HH} = \frac{1}{3}\Delta_0 - \frac{1}{2}\delta E, \tag{3}$$

where the value δE for elastic strain in the direction [001] can be given as:

$$\delta E_{001} = 2b(\varepsilon_{zz} - \varepsilon_{xx}). \tag{4}$$

The values of spin-orbit splitting of the valence band Δ_0, deformation potential b for SiGe nanoislands are determined using a linear interpolation of the data for clean Si and Ge.

According to our experimental results and estimations in [21], residual elastic strain value in the investigated SiGe quantum dot multilayer structures amounts to $\varepsilon_{xx} \approx 0.026\ldots0.027$. The best correspondence of theoretical calculations and experimental results concerning the dependence of the valence band offset of a compressively strained Ge on Si gives value $\Delta E_V^{av} = 0.54$ eV for average value and $\Delta E_V^{hh} = 0.74$ eV value for heavy hole discontinuity.[16] The estimations showed that the valence band offsets for the strained interface $Si_{0.2}Ge_{0.8}/Si$ come to $\Delta E_v^{av} = 0.43$ eV and $\Delta E_{hh} = 0.59$ eV.

In particular, for a Ge quantum box with the width $L_z = 2$ nm, lateral dimensions $L_x = L_y = 40$ nm, and $m_{hh}^* = 0.34 \cdot m_0$, one has $E_{111} = 0.132$ eV, $E_{121} = E_{211} = 0.135$ eV, $E_{112} = 0.472$ eV, $E_{212} = 0.475$ eV.

The proposed model is a simplified one allowing only a rough estimate of the number of the localized states in the valence band of the QDs. It is evident that taking into account the real shapes of the nanoislands, as well as inhomogeneous strain values and Ge fraction

distribution inside the clusters will give more precise positions of the energy levels in the valence band.

Spectral dependences of lateral photoconductivity

Spectral dependences of lateral photocurrent of Ge/Si heterostructure with quantum dots were measured at normal incidence and side illumination at 77 K (fig.5). The photocurrent is caused by intraband transition between localized states in the valence band of nanoislands.

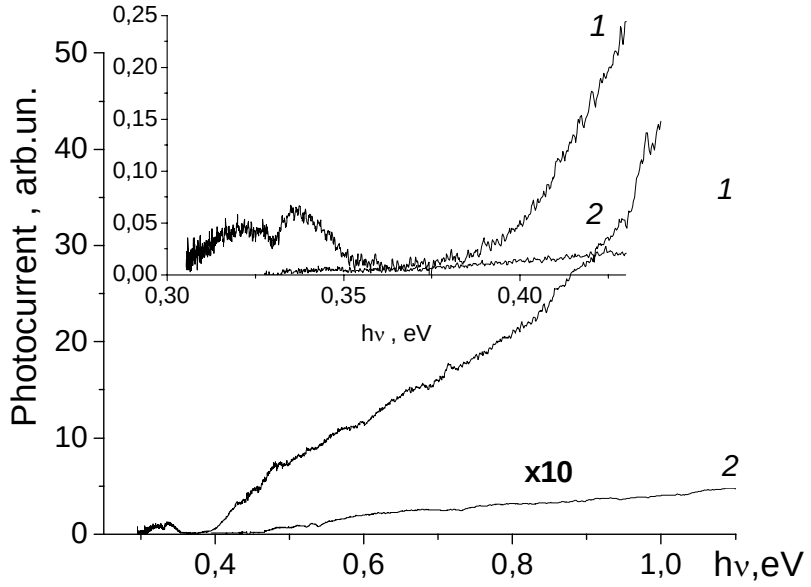


Fig. 5. Spectral dependence of lateral photoconductivity of Ge/Si heterostructure with Ge quantum dots measured at side excitation (*1*) and normal incidence (*2*) at 77 K. The inset shows in details the low-energy part of the spectra.

In the case of side excitation, when unpolarized light spreads along the basis of Ge nanoislands, there exists a component of the vector **E** oscillating along the growth direction (*z*-component), in which the confinement is the most pronounced. As it is known, intraband transitions in potential wells may be caused only by *z*-component of the vector **E**.

The optical absorption coefficient for the transition from level *i* to *f* in a QDs is proportional to the matrix element:

$$\alpha \propto \left| \left\langle \psi_i \left| \mathbf{p} \right| \psi_f \right\rangle \right|^2, \tag{4}$$

where $\mathbf{p}$ is the momentum operator for corresponding transition. The integral defining the transitions between the levels has the form:

$$P_{mn} = -i\hbar \int\int\int \psi_i^* \frac{\partial}{\partial x_k} \psi_f \, dxdydz, \tag{5}$$

where $x_k = x, y, z$. Polarization selection rules will be specified by nonzero terms of this integral. Only transitions with $\Delta n = 1$, i. e. between the first and second levels are dominant [22]. The transitions $E_{111} \to E_{112}$, $E_{121}, E_{211} \to E_{122}, E_{212}$ and $E_{221} \to E_{222}$ are allowed with polarization along the growth axis.

Selection rules can change if a potential well has a finite depth and effective masses of carriers in the well and barrier layer are different. As a consequence, electron transitions under the irradiation of light polarized in the lateral structure direction become possible (under the

action of *x*- and *y*-components of the vector **E**). As regards GaAs/AlGaAs quantum wells, the absorption coefficient for the light polarized along quantum dimensional layers is several orders smaller than for the light with *z*-polarization [23]. However, selection rules are modified in quantum dots making it possible to observe intraband transitions at one band also at normal incidence of exciting irradiation. As a result, the photocurrent value in our photoconductivity experiments depended essentially on the way of irradiation of the heterostructure. The photocurrent in the range from 0.3 to 1.0 eV at normal incidence of exciting irradiation was much smaller (fig. 5, curves 2).

In the case of side excitation in the spectral range from 0.3 to 0.37 eV, two peaks of current were observed at 0.32 eV and 0.34 eV and the photocurrent increased monotonically as the quantum energy increased in the range $h\nu$ >0.38 eV. The peaks of current observed in the spectral range from 0.3 to 0.37 eV can be attributed to hole transitions between the levels in the valence band of nanoislands. It can be seen that these values, by order of magnitude, fall within the range of excitation energies which follow from the level positions estimated above for SiGe quantum dots. The photoresponse in the range $h\nu > 0.38$ eV may be caused by hole transitions from the ground state of the valence band of SiGe nanoislands to two-dimensional continuum states of the valence band of a wetting layer or intermediate layers of Si surrounding [17, 24, 25]. Nonequilibrium carriers excited by such transitions may contribute to the observed lateral photocurrent, as far as no potential barriers exist for electron transport in the lateral direction. However, it is impossible to distinguish a contribution of bound-to-continuum transitions to the states existing in a wetting layer or Si intermediate layers in the conditions of given experiment.

Conclusions

The current density *J* of field emission depends on the product of supply function *S(E)* and the transmission factor of the potential barrier *T(E)*. The observation of peak in *I-V* curves of photofield emission indicates to existing of quantized energy levels in QDs that causes the dependence of barrier transmission of Si/QD/vacuum barrier on applied voltage value. It may reflect resonant tunneling of electrons via energy levels in QD potential well. The investigation of lateral photoconductivity in Si/Ge heterostructures with SiGe nanoislands revealed the presence of localized states in the valence band of Ge nanoislands with the energy distance between them about 0.32 and 0.34 eV. Moreover, the lateral photocurrent is determined by features of transport of nonequilibrium carriers, therefore the supply function properties. Intraband transitions between the localized states of the valence band of Ge nanoislands, in our opinion, are responsible for the observed photosensitivity of lateral photoconductivity and photofield emission in Si/Ge heterostructures with Ge quantum dots in the midinfrared range.

Acknowledgements

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Квантові ефекти в багатошарових Si-Ge наногетероструктурах

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Методами фотопольової емісії та повздовжньої фотопровідності було досліджено багатошарові наногетероструктури з квантовими точками германію. Попередні дослідження показали тісний зв'язок пружних напружень, що виникають при епітаксійному рості матеріалів з різними постійними граток, з виникненням додаткових рівнів в зоні такого гетеропереходу. Таким чином, відносно простими але інформативними методами спостерігалися міжзонні переходи, які відсутні в двовимірних гетероструктурах Si-Ge. Якщо вплив кількості шарів на фотопольову емісію несуттєвий, то при дослідженні фотопровідності виявлено прямий зв'язок зсуву чутливості повздовжньої фотопровідності в середній інфрачервоний діапазон від кількості шарів з квантовими точками германію. Показано, що розмірні характеристики та мольний склад квантових точок відповідають енергетичним рівням з енергетичною відстанню між ними приблизно 0,32 та 0,34 еВ з дуже високою точністю. Результати досліджень дозволяють сподіватися на їх використання в перспективних приладах нано- та оптоелектроніки.