OPTICS, LASERS, AND QUANTUM ELECTRONICS

	PHOTOCONDUCTIVITY AND FIELD-ASSISTED PHOTOEMISSION IN MULTILAYER Si/Ge HETEROSTRUCTURES WITH QUANTUM DOTS
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Lateral photoconductivity spectra and current-voltage characteristics of field-assisted photoemission from multilayer Ge/Si heterostructures with SiGe quantum dots (QDs) have been studied. Atomic force microscopy images of the top layer showed that nanoislands were tetrahedral pyramids by shape, about 30 nm in base and about 2 nm in height. Their average surface concentration is about 10^{10} cm⁻². The photocurrent spectroscopy studies of the structures are carried out at a temperature of 77 K and in the quantum energy range from 0.29 to 1.0 eV. Two peaks of the lateral photocurrent with the maxima observed at 0.32 and 0.34 eV are explained by intraband transitions between localized states in the valence band of nanoislands. A peak observed in the I-V curve of the field-enhanced photoemission from QDs is associated with the resonant tunneling of electrons from the Si valence band into vacuum via quantized energy levels in QDs. The intraband transitions from localized states in the valence band of Ge nanoislands are found to be responsible for the photocurrent and the field-enhanced photoemission of electrons observed in the Si/Ge heterostructures with QDs under study.

1. Introduction

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Progress in the development of nanotechnologies made possible the creation of nanosized semiconductor objects,

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in particular, Ge–Si heterostructures with QDs. These heterostructures draw attention of researchers throughout the world as systems that allow the band engineering and silicon technologies to be combined. This makes promising the development of a new generation of optoelectronic devices, in particular, photoemission detectors of infrared range and photo diodes with QDs [1–3]. Owing to their geometry, heterostructures with ensembles of nanosized Ge quantum dots can provide high electric fields with a strength of about 10⁷ V/cm near the QD base, even if the moderate applied potential difference of about $10^3 - 10^4$ V stimulating the intense electron emission is applied.

Since 1960, the field emission from semiconductor cathodes has been widely studied by a number of scientific groups [4–8]. To our knowledge, the measurement of the field emission from Ge nanoclusters grown up on a Si substrate has been carried out for the first time by Tondare *et al.* in 2000 [9]. Those researches have not discovered any peculiarities in the current-voltage (I - V) characteristics of the field emission. We also studied such systems [10, 11] and revealed an interesting detail, namely, peaks in the I - V characteristics of the field emission from Ge nanoclusters on a Si(100) substrate. The number of peaks was found to depend on the nanoisland dimensions. The peaks observed were associated with effects of size quantization in Ge nanoislands. Moreover, the field emission current substantially changed at the specimen illumination in the wavelength range from 0.4 to 10 μ m. In the structures concerned, the field emission current at room temperature grew by a factor of from 5 to 3 at illuminating them in the wavelength interval from 2 to 10 μ m, respectively [12]. In addition, the spectra of lateral photoconductivity in the near infrared range, which are associated with optical transitions from localized states in the valence band of nanoislands, were also studied separately [13, 14].

It is also worth noting that the photo-electric and electronic properties of Ge-Si heterostructures with Ge quantum dots should be analyzed taking the morphology of such systems into account. Silicon-germanium low-dimensional heterostructures are classed to type II, in which holes in the valence band of Ge quantum dots are localized, whereas electrons in the conduction band are free. A significant amount of works was devoted to studying the spectrum of electron states in QDs. The positions of energy levels were shown to be governed by the QD dimensions, shape, and component composition [15]. An important parameter that considerably affects the optical and photo-electric properties of Ge/Si heterostructures is the magnitude of valence band discontinuity. The average value of valence band discontinuity in a $Si/Si_{1-x}Ge_x$ heterojunction is adopted to depend linearly on the Ge content x and amounts to 0.54x eV. Such an approximation allows the properties of heterojunctions with various Ge contents to be described rather accurately [16].

This work is devoted to experimental researches of the lateral photoconductivity and the field-assisted electron photoemission from multilayer Ge/Si heterostructures with Ge quantum dots. Such researches brought about the discovery of a relationship between the discrete electron spectrum of charge carriers and the nature of electron transport in such structures. Hence, this work aimed at studying the correlation between peculiarities in the I-V characteristics of the field-assisted photoemission and the lateral photoconductivity in Ge/Si heterostructures with QDs.

2. Experimental Part

Structures to study were grown up on a KATUN installation (Novosibirsk, Russia), using the molecular beam epitaxy (MBE) method. Multilayer Ge/Si heterostructures with QDs were grown up on a Si(100) substrate

The process of crystal growing was controlled in situ with the help of the high-energy electron diffraction (HEED) method, by monitoring the intensity of central reflected beam oscillations. One period of oscillations corresponded to a Ge or Si monolayer about 0.135 nm in thickness. The substrate temperature T_s was varied within a wide interval from 450 to 850 $^{\circ}$ C. The growth rates, which were determined in our experiments from HEED measurements, varied from 0.02 to 0.15 nm/s for Si and from 0.01 to 0.05 nm/s for Ge layers. The molecular Si and Ge beams were created with the use of evaporators heated by the electron bombardment. The evaporators were constructed as automated crucibles. They operated in the mode where the evaporation rate was proportional to the electron-beam power. The chemical composition of molecular beams was monitored with a quadrupole mass-spectrometer. The residual background gas pressure in the MBE installation was less than or equal to 10^{-10} Torr.

In order to create a multilayer system of QDs with a more uniform distribution of nanoislands over the substrate surface, we proposed to use a system of $\text{Si}_{1-x}\text{Ge}_x$ intermediate layers with subcritical thicknesses [11]. To avoid the formation of mismatch dislocations, we proposed that, instead of a thin (5 to 50 monolayers) wetting layer, a series of intermediate layers should be formed, where the Ge content x should be gradually, layer by layer, increased from 0 to 0.6–0.8, with a simultaneous gradual decrease of the substrate temperature down to $T_s = 500$ °C. This allowed us to monitor the level of elastic strains in the intermediate layers, where Ge clusters were actually formed, and simultaneously reduce the influence of the Si substrate, provided that the thicknesses of intermediate layers did not exceed critical values.

The technology used to produce multilayer Ge–Si nanostructures differed from that described above in that the time moment, when the nanoislands achieved the stage of hut clusters by forming intermediate Si layers 5 to 15 nm in thickness, was determined more exactly. This process was mastered well earlier, when fabricating Si–SiGe superlattices, whose periodicity was determined with a high accuracy by the operation time of Si and Ge sources [17] (see also similar results obtained for InAs quantum dots on GaAs substrates [18]). Hence, multilayer structures were fabricated by repeating the procedure used to grow a monolayer of Ge nanoclusters three to eight times. Since the time required for the shutter to switch the beam on or off was about 0.1 s, the relative error for the periodicity in Ge-Si multilayer structures amounted to about 1-2%.

Thus, the intermediate-layer technology allowed the arrays of Ge quantum dots with an optimal size of QDs and a fixed level of residual elastic deformations in the nanoisland film to be fabricated. As was shown by us earlier, the relative elastic deformation in the nanoisland film averaged over the substrate surface, ε , is proportional to the quantity $\sqrt{1/L}$, where L is the lateral size of Ge quantum dots [19]. Hence, the possibility to estimate the residual elastic deformation in a nanoisland film made it possible to obtain ensembles of practically identical Ge nanoclusters in every layer. Therefore, we suppose Ge quantum dots in multilayer structures to be integrated vertically.

To study the surface morphology, atomic force microscopy (AFM) methods were applied. In Fig. 1, an AFM image of the top layer of the specimen with nanoislands is depicted. The figure demonstrates that the nanocluster shapes are tetrahedral pyramids with a base of about 30 nm and a height of about 2 nm. The average concentration of clusters distributed over the surface was about 10^{10} cm⁻². The growing of every intermediate Si layer was continued until there appeared a high-contrast HEED pattern Si(100) 2×1 , which is typical of a clean Si surface. In such a way, we formed multilayer arrays of Ge-Si(100) nanoclusters with three, five, and eight lavers of SiGe quantum dots characterized by a thickness of about 3.5 nm at the temperature $T_s = 500$ °C. The Ge content in the researched structures was estimated using the secondary ion mass-spectroscopy (SIMS) technique. This method was applied for the sequential monitoringin the growth direction with a step of 1 nm-of the Ge content in the Si/Ge layers with QDs. Taking the surface concentration of QDs and their average size into account, the average Ge content in the QDs ranged from 75 to 85%.

The field emission and the field-assisted photoemission of electrons were measured in a diode cell. The distance between electrodes was about 300 μ m (Fig. 2,*a*). To visualize the spatial distribution of emitted electrons, a cathodoluminescent ZnS screen was deposited onto a glass plate covered preliminarily with a SnO₂ conducting layer which served as an anode. The details of experiment were described in work [12].

To measure the lateral photoconductivity of multilayer structures with SiGe quantum dots, two Au ohmic contacts 1 mm in diameter each were fused into the surface with epitaxial layers, at a distance of 10 mm from each other (Fig. 2,b). As a result, the contact was provided between every layer with QDs and the Si substrate. The

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Fig. 1. 1×1 - μ m² AFM image of a structure with QDs at 500 °C. The average height and lateral size of QDs are about 2 and 30 nm, respectively



Fig. 2. (a) Diode cell for measuring the field emission and the field-enhanced photoemission of electrons. (b) Specimen for photoconductivity measurements

photocurrent-voltage characteristics of all stidied structures turned out to be linear in the voltage range from U = -50 V to +50 V and the temperature interval 77– 290 K. The spectral dependences of the lateral photoconductivity were measured on an infrared spectrometer at U = 5 V in the $h\nu$ -range from 0.3 to 1.1 eV. The photocurrent was measured making use of a current amplifier and applying the standard lock-in detection technique. The measured spectral dependences were normalized to a constant number of exciting radiation quanta using a nonselective pyroelectric detector.

3. Results and Their Discussion

The structures concerned demonstrated the stable field emission at T = 290 K, when a macroscopic electric



Fig. 3. (1) Current-voltage characteristic of the field-enhanced photoemission from a Si/Ge heterostructure with 5 layers of QDs about 2 nm in height. (2) Current-voltage characteristic of the dark field emission for the same heterostructure

field of about 10^5 V/cm was applied. It should also be noted that local electric fields, which make the surface potential barrier transparent enough for the electrons to be emitted, are much higher by magnitude (of about 10^7 V/cm). It is difficult to evaluate those fields more accurately, since they are determined by details of geometrical and compositional heterogeneities on the nanometer scale [20]. Curve 1 in Fig. 3 is the I-V characteristic of the field-enhanced photoemission from a heterostructure with SiGe quantum dots about 2 nm in height. The specimen was irradiated making use of a 10-W halogen lamp located at a distance of 20 cm from the specimen. The dark field emission current measured without specimen irradiation was several orders of magnitude lower (Fig. 3, curve 2). At the same time, neither the field emission nor the field-enhanced photoemission were registered in the voltage range 2.0–4.3 kV for Si specimens free of QDs. We suppose that the observed field-assisted photocurrent can be generated by the electron emission from QDs.

A specimen, whose I-V curves are exhibited in Fig. 3, revealed practically no dark current (to an accuracy of less than 0.01 μ A) in the voltage range under investigation. However, in our previous works [11], we reported results measured for other Si/Ge heterostructures with QDs, which testified to rather high dark field emission currents (of about $10^{-7}-10^{-5}$ A at anode voltages ranging from 10^2 to 10^4 V and the total specimen surface area of about 10^{-1} cm²). It turned out that structures with QDs about 3 to 5 nm in height demonstrate the field emission I-V curves with a number of well-pronounced



Fig. 4. Energy band structure of a Si/SiGe heterojunction (a) without and (b) with the applied electric field $F = 10^6$ V/cm

maxima (peaks) [11]. The number of those peaks in the field emission I-V curves decreases together with the QD size growth, so that, for QDs higher than 10 nm, the clear current peaks can hardly be observed. We attributed this effect to resonant electron tunneling via quantized energy levels in QDs.

We also suppose that the current peaks in the I-Vcurve of the field-enhanced photoemission (Fig. 3, curve 1) can emerge owing to the energy quantization, in particular, due to the presence of discrete energy levels in the valence band of QDs. If the external electric field is absent, Si/Ge heterojunctions are classed to heterostructures of the second type, in which the potential well for holes is formed in QDs. To elucidate the origin of the emission current peak appearance, we numerically solved the Schrödinger equation for a multilayer Si/Ge heterostructure, by assuming a linear drop of the potential in it. It is worth noting that a real distribution of the potential near the surface is highly asymmetric in any case. In Figs. 4, a and b, the calculated energy-band diagrams for a Si/SiGe heterojunction without and with the applied electric field $F = 10^6$ V/cm, respectively, are depicted. The quantum-mechanical motion of holes in the growth direction is responsible for the appearance of two localized states in the valence band of QDs. It should be noted that the positions of energy levels in the upper QD layer differ from those in the lower layers of

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QDs surrounded by silicon. An electric field applied to the surface changes the positions of energy levels in the valence band of QDs, which corresponds to the so-called quantum-confined Stark shift. The Stark shift of energy levels in a quantum well is larger, if the latter is nonsymmetric. Since the energy barrier of a quantum well is nonsymmetric, and there exists a nonuniform potential gradient in the growth direction near the upper layer, the energy levels in the latter become shifted most considerably. The shape and the transparence of a potential barrier in multilayer Si/Ge heterostructures depend substantially on the applied electric field strength.

In particular, the shape and the width of a potential well for holes in the valence band of SiGe nanoislands changes substantially, when an electric field is applied to the surface. As a result, the discrete energy values E_n change as well. At a certain value of the applied electric field, the position of a quantization energy level coincides with the top of the Si valence band, so that the resonant tunneling of electrons from this band via the quantization energy levels in the SiGe quantum dots into vacuum becomes possible. As the electric field grows further, the number of levels in the well decreases by one, and the resonant tunneling occurs via the next (n-1)-th energy level. The current peak, which is observed for a structure with QDs about 2 nm in height, may testify that there are at least two energy levels in the valence band of a SiGe nanoisland without the electric field. Starting from a certain value of electric field strength, a QD in the upper layer contains a single level only (the ground level, n = 1), which corresponds to the presence of only one peak in the I-V curve.

The model proposed is rather simplified, and it can describe main peculiarities of obtained experimental data at a qualitative level only. It should be noted that a real potential profile near the QD surface still remains indefinite owing to its complicated dependence on a number of factors that are difficult to evaluate, such as an exact geometry and a composition of QD and the strain distribution in the QD nanocluster. Should the real shapes of nanoislands, field penetration, the nature and the properties of surface states, and the Ge distribution in the clusters be determined, it would allow a more precise evaluation for the positions of energy levels in the valence band to be done.

A considerable increase of the field emission current observed, when the structures with Ge quantum dots were irradiated, could be induced by optical transitions in QDs via the quantized energy levels. In lowdimensional Si/Ge heterostructures with Ge quantum dots, there exist several types of electron transitions that



Fig. 5. Spectral dependences of the lateral photoconductivity of a Ge/Si heterostructure with Ge quantum dots measured at the lateral excitation (1) and the normal incidence of light (2) at a temperature of 77 K. The inset details the low-energy section of the spectra

are responsible for the photoresponse in the near infrared range: (a) transitions from the quantized energy levels in the valence band of Ge quantum dots into the conduction band of the Si environment, (b) transitions between localized states in the valence band, and (c) bound-tocontinuum transitions [3, 21].

The spectral dependences of the lateral photoconductivity in Ge/Si heterostructures with QDs were measured at 77 K, at the normal light incidence and at the lateral excitation (Fig. 5). The observed photocurrent might be stimulated by an intraband transition between localized states in the valence band of nanoislands.

In the case of the lateral excitation, when nonpolarized light propagates along the base of Ge nanoislands, there exists a component of the vector \mathbf{E} which oscillates along the growth direction (the z-component) and the quantum-mechanical confinement for which is most pronounced. Intraband transitions in potential wells are known to be induced only by the z-component of the vector \mathbf{E} .

The optical absorption coefficient for the transitions from the i-th onto the f-th level in QDs is proportional to the matrix element

$$\alpha \propto |\langle \psi_i \, | \mathbf{p} | \, \psi_f \rangle|^2 \,, \tag{1}$$

where \mathbf{p} is the momentum operator for the corresponding transition. An integral that determines interlevel transitions looks like

$$P_{mn} = -i\hbar \int \int \int \psi_i^* \frac{\partial}{\partial x_k} \psi_f dx dy dz, \qquad (2)$$

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where $x_k = x, y, z$. Polarization selection rules correspond to nonzero conditions. Transitions with $\Delta n = 1$, i.e. between the first and the second level, dominate [22]. The transitions $E_{111} \rightarrow E_{112}$, $(E_{121}, E_{211}) \rightarrow (E_{122}, E_{212})$, and $E_{221} \rightarrow E_{222}$ are allowed, if the polarization is directed along the growth axis.

The selection rules can change, if the potential well depth is finite and the effective masses of charge carriers are different in the potential well and in the barrier. As a consequence, the electron transitions induced by irradiation with light polarized along the structure (under the action of the x- and y-components of the vector \mathbf{E}) become possible. As concerning GaAs/AlGaAs quantum wells, the absorption coefficient of light polarized along the layers with the quantum size effect is several orders of magnitude lower than that for light with the z-polarization [23]. However, the selection rules change in QDs, which makes it possible to observe intraband transitions within a single band at the normal incidence of exciting irradiation as well. As a result, the photocurrent in our photoconductivity experiments substantially depended on the way of heterostructure irradiation. In the range 0.3 - 1.0 eV and at the normal incidence of exciting irradiation, the photocurrent turned out much lower (Fig. 5, curves 2).

In the case of the lateral excitation in the spectral interval 0.3 - 0.37 eV, two current peaks were observed at 0.32 and 0.34 eV, and the photocurrent grew monotonically as the quantum energy $h\nu$ increased above 0.38 eV. The current peaks observed in the spectral interval from 0.3 to 0.37 eV can be attributed to hole transitions between the levels with n = 1 and 2 in the valence band of nanoislands. It is easy to see that these values fall, by their order of magnitude, within the range of excitation energies that correspond to the level positions evaluated above for SiGe quantum dots. The broadening of those peaks can be associated with a non-uniform distribution of nanoislands over their dimensions (with a dispersion of 10%) and with an interference between the states in two neighbor vertically integrated QDs. The interaction between dots eliminates the degeneracy of energy states and gives rise to the broadening of absorption (photocurrent) spectra.

The photoresponse in the quantum-energy range $h\nu > 0.38$ eV may be connected with hole transitions from the ground state in the valence band of SiGe nanoislands into two-dimensional continuum states in the valence band of either the wetting layer or intermediate layers of the Si environment, i.e. the so-called bound-to-continuum transitions [20, 24, 25]. Nonequilibrium charge carriers excited in the course of such transitions can contribute

to the observed lateral photoconductivity, since there are no potential barriers for the electron transport in the longitudinal direction. However, under the conditions of our experiment, it is impossible to distinguish between the contributions made by bound-to-continuum transitions into the states in the wetting layer and in the Si intermediate layers.

4. Conclusions

The field emission electron current density J depends on the product of the supply function S(E) and the potential barrier transparency T(E). The observation of peaks on the I-V curves of the field-assisted photoemission evidences the existence of quantized energy levels in QDs, which causes the transparency of the Si/QD/vacuum barrier to depend on the applied voltage. This can explain the resonant tunneling of electrons via the energy levels in the QD potential wells. The study of the lateral photoconductivity in Si/Ge heterostructures with SiGe nanoislands revealed localized states in the valence band of Ge nanoislands, with the interlevel distances of about 0.32 and 0.34 eV. Moreover, the lateral photocurrent is governed by the character of nonequilibrium carrier transport and, therefore, by the supply function properties. In our opinion, the intraband transitions between localized states in the valence band of Ge nanoislands are responsible for the observed lateral photoconductivity of and the field-enhanced photoemission from Si/Ge heterostructures with Ge quantum dots in the middle infrared range.

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ФОТОПРОВІДНІСТЬ ТА ФОТОПОЛЬОВА ЕМІСІЯ В БАГАТОШАРОВИХ Si/Ge ГЕТЕРОСТРУКТУРАХ ІЗ КВАНТОВИМИ ТОЧКАМИ

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Резюме

Досліджено спектри поздовжньої фотопровідності та вольтамперні характеристики фотопольової електронної емісії багатошарових гетероструктур Ge/Si з квантовими точками SiGe. Зображення верхнього шару, зроблені за допомогою атомносилової мікроскопії, показали, що наноострівці мали форму тетраедральних пірамідок з розмірами 30 нм у підвалині та 2 нм висотою. Середня густина їх розподілу по поверхні підкладки складала близько 10¹² см⁻². Структури досліджено за допомогою спектроскопії фотоструму при 77 К в діапазоні hvвід 0.29 eB до 1.0 eB. Спостерігали два піки поздовжнього фотоструму з максимумами за 0,32 eB та 0,34 eB, які пояснено внутрішньозонними переходами між локалізованими станами у валентній зоні наноострівців. Спостерігався пік струму на кривій І-V фотопольової електронної емісії з квантових точок, який пов'язуємо з резонансним тунелюванням електронів із валентної зони Si у вакуум через рівні квантування у квантових точках. Внутрішньозонні переходи з локалізованих станів у валентній зоні наноострівців Ge зумовлюють фотострум та фотопольову емісію електронів, спостережувані в гетероструктурах Si/Ge із квантовими точками.