PROPERTIES AND INTERCONVERSION OF SELF-INDUCED SiGe NANOISLANDS OF DIFFERENT SHAPES

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In this work, SiGe nanoislands formed on Si and $Si_{0.9}Ge_{0.1}$ buffer layers are studied with the use of the atomic force microscopy and Raman spectroscopy techniques. To study the islands of a certain shape, the monomodal arrays of the islands of this shape (hut-clusters, pyramids, domes) were formed by means of a variation of both the thickness of a deposited Ge layer and the growth temperature. It is established that, among the uncaped islands of different shapes, the greatest values of elastic deformation are characteristic of the pyramids, while the smallest ones — of the domes. A degree of the stress relaxation due to a geometric factor is greatest in the dome-like islands, minimal in size. The utilization of a $Si_{0.9}Ge_{0.1}$ buffer layer for the subsequent growth of islands on it favors their lateral ordering.

1. Introduction

Self-induced SiGe nanoislands, which are formed in the process of epitaxial growth of Ge on Si, have intensively been studied in recent years. This is stimulated by two factors. First, the nanoislands have the dimensions, for which quantum effects begin to manifest themselves. This makes it possible to experimentally verify the correctness of the utilization of existing theoretical approximations for the description of phonons and charge carriers confined in space. Secondly, it is possible, by means of a change in dimensions, a shape, the density, and the component composition of nanoislands, to "construct" structures with the parameters required for opto- and nanoelectronic devices. It is the latter argument that makes the researchers to fabricate an ordered ensemble of islands, which, being of the same shape, have the small size dispersion and the maximally high spatial density. It is shown in works [1,2] that, by means of a variation of such parameters of molecular beam epitaxy (MBE) as the substrate temperature, nominal thickness of a deposited Ge layer, and velocity of a molecular flux, it is possible to change, within wide limits, the island parameters, including their shape. The islands of different shapes are characterized by different values of stresses and the degree of their relaxation at the expense of a geometric factor. The stresses in the islands define the interdiffusion processes, which occur with a participation of the substrate atoms and influence their component composition. However, the investigations of islands of a certain shape are complicated by the fact that the structures fabricated contain the islands of at least two shapes (pyramidal and dome-like), and it is difficult to separate their contributions to experimental spectra. The aim of this work is the fabrication of the nanostructures containing nanoislands of a homogeneous shape and the determination of the component composition and elastic deformations peculiar to each of the possible shapes.

2. Experimental Procedure

Nanostructures, which were formed in the process of MBE of Ge on the Si(100) substrate with a preliminary grown either Si or $Si_{0.9}Ge_{0.1}$ buffer layer (200 and 10 nm in thickness, respectively), were used as the objects of investigation. For each of the structures grown, the homogeneity of an island shape was achieved by means of a variation of the set of parameters, including the substrate temperature in the process of growth, the deposition rate of Ge atoms, and their amount. The islands of a hut-cluster shape were formed at a temperature of 500 °C by means of the deposition of 8 monolayers (MLs) of Ge on the Si buffer. To form the pyramidal islands, 4.2 MLs of Ge were deposited on the $Si_{0.9}Ge_{0.1}$ buffer layer at 700 °C. The dome-like islands were formed at a temperature of 700 °C by means of the deposition of 11 MLs of Ge on the silicon buffer layer or 13 MLs of Ge on the $Si_{0.9}Ge_{0.1}$ one. To prevent the oxidation of the structures with hut-clusters, which were subsequently used for the measurements of Raman

scattering (RS) spectra, they were covered with a Si layer of 50 nm in thickness.

The RS spectra were registered at room temperature with the use of a DFS-24 diffraction spectrometer. For the excitation of spectra, an Ar^+ laser with a wavelength of 487.9 nm was used. The signal was registered with a cooled photomultiplier operating in the photon counting mode. The experiments were carried out in the back-scattering geometry. For a more precise determination of the RS band location, the known values of the plasma discharge frequencies for an Ar^+ laser were used.

The surface morphology of the structures with nanoislands was studied with the use of a NanoScope-IIIa atomic force microscope (the Digital Instruments firm) in the tapping mode. Before and after the measurements, the test of a probe was carried out to check a tip shape. Highly symmetric probes with a section radius of less than 6 nm, when measured at a distance of 10 nm from the tip beginning, were used for the nanostructure investigation. This made it possible to neglect the effect of the convolution of a probe shape and the surface under investigation on the results of the analysis of the nanoisland shape and dimensions.

3. Results and Discussion

Fig. 1 shows the AFM images of self-induced SiGe nanoislands of various shapes. According to our previous AFM studies [3], as well as to other results [4], nanoislands in the form of pyramids with a rectangular base (the so-called hut-clusters) are formed (Fig. 1,*a*) at temperatures of epitaxy $T \leqslant 550$ °C, when the thickness of a germanium layer is small enough ($d_{\mathrm{Ge}} \leqslant 8$ MLs). For such islands, the (105) planes are side faces and an angle of their slope to the base is 11°. An increase in the nominal thickness of the deposited germanium layer ($d_{\mathrm{Ge}} > 8$ MLs) gives rise to a transformation of the hut-clusters into dome-shaped islands. For these islands, the (113) planes are side faces and an angle of their slope to the base is $\sim 25°$. The annealing of the structures with hut-clusters at $T = 550$ °C gives rise to the same effect [5]. As the temperatures of epitaxy $T \geq 550$ °C and the thickness of a Ge layer corresponds to a 2D—3D transition ($d_{\mathrm{Ge}} \approx 4$ MLs), the energetically most favorable shape of islands is a pyramid with a square base (Fig. 1,*b*). As a nominal thickness of the deposited Ge layer increases to $d_{\mathrm{Ge}} \approx 5$ MLs, the volume of the pyramids grows and, for a certain fraction of the pyramids, it reaches the critical value V_{cr}, at which the chemical potential of the pyramids gets equal to that of

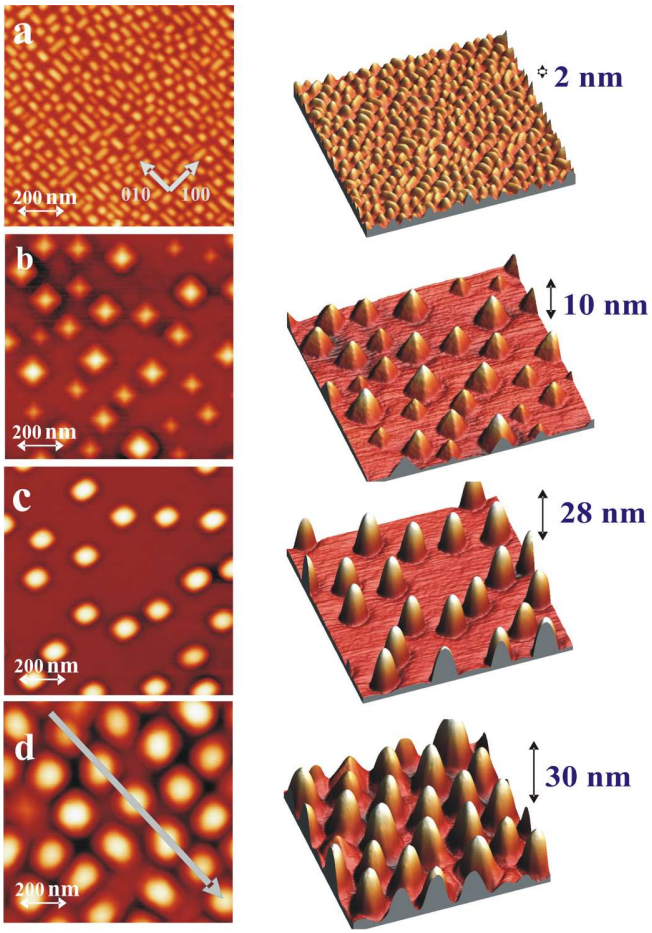

Fig. 1. AFM images (top and three-dimensional views) of the self-induced nanoislands formed at 500 °C by means of the deposition of 8 MLs of Ge on Si (*a*) and at 700 °C by means of the deposition of 4.2 MLs of Ge on $Si_{0.9}Ge_{0.1}$ (*b*), 11 MLs of Ge on Si (*c*) and 13 MLs of Ge on $Si_{0.9}Ge_{0.1}$ (*d*) buffer layers. The arrow shows the direction, along which the islands are aligned

the domes. As a result, a transformation of the island shape from one to another begins [6]. The transitions reflecting this transformation are shown by the arrows *1* and *2* in Fig. 2, where the dependences of the island height on its lateral dimension are depicted for various substrate temperatures in the process of MBE. Due to the non-simultaneous nucleation of pyramidal islands on the sample surface, a scattering in their dimensions is expected. In fact, as is evident from the AFM studies, as the thickness of the Ge layer deposited on the Si substrate at T=700 °C varies from 5 to 11 MLs, the ensembles of islands are formed, which have the shape of both pyramids and domes. For the same temperature

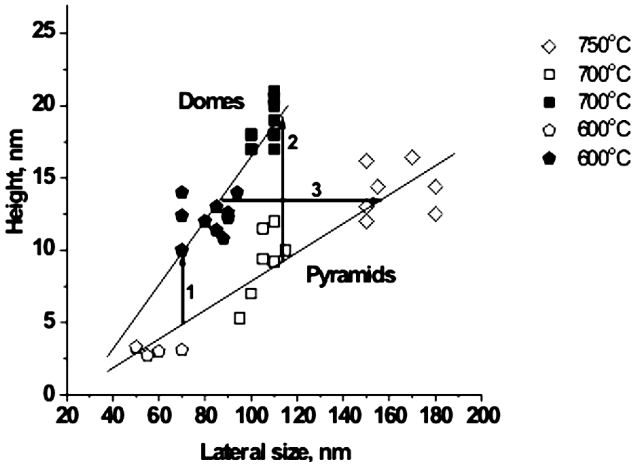

Fig. 2. Dependences of the island heights on their lateral dimensions for the dome-like and pyramidal islands. The arrows show possible transitions from the pyramidal to dome-like shapes in the process of growth of nanoislands (*1,2*) and the inverse transitions at annealings (*3*)

of epitaxy and the nominal thickness of the deposited germanium layer $d_{\mathrm{Ge}} \approx 11$ MLs, all the islands are solely of the dome-like shape (Fig. 1,*c*).

Among others, the energy of interaction of islands between one another also affects a value of the critical volume, at which the transition from one shape to another occurs. As the surface density of islands is sufficiently high, the island free energy increases by a value of the energy of interaction of the given island with all the rest. In this case, the chemical potential of pyramids becomes equal to that of domes at smaller values of the pyramid volume.

When the dome-like nanoislands, which are already formed, are subjected to the additional annealing at the growth temperature or greater, the inverse transformation of domes to pyramids is also possible. This occurs due to a growth of the Si content in domes owing to the considerable temperature-induced interdiffusion enhanced by inhomogeneous stresses within islands and around them. As a result of the substantial Si—Ge intermixing, there is a decrease in a mismatch of the lattice constants of the Si buffer layer and SiGe islands, which leads, in turn, to a decrease in the elastic energy of domes. For this reason, at a given island volume, the chemical potential of pyramids becomes smaller compared to that domes, and the inverse transformation begins. In Fig. 2, this inverse transition is shown by arrow *3*. The transitions of such a kind were also observed when dome-like islands were covered with Si [7], as well as at long-term annealings [8]. We showed in [1] that, when a temperature of epitaxy rises, the SiGe intermixing in pyramids increases, which gives rise to a growth in the critical volume V_{cr}, at which the pyramid—dome transition occurs. As a result, the pyramids can dominate again at growth temperatures of about 750 °C.

Contrary to the case of the Si buffer, the process of formation of islands on the stressed $Si_{0.9}Ge_{0.1}$ buffer 10 nm in thickness somewhat differs from that described above. When, in the MBE process, Ge gets deposited on such $Si_{0.9}Ge_{0.1}$ layer, the total elastic energy of the system rises and, thus, the 2D—3D transition occurrs at a lower thickness of the deposited layer [9]. As is shown in [9], the epitaxial layer thickness, at which the 2D—3D transition occurs, decreases with a growth in the Ge content in a $Si_{1-x}Ge_x$ buffer. The "earlier" transition leads to a reduction in the thickness of the wetting layer (WL), and, thus, the amount of deposited Ge, from which the nanoislands are formed, increases. Furthermore, during the formation of islands, the removal of atoms of the WL from around the islands and the beginning of the diffusion of Si atoms from the buffer layer occur earlier. It is in the initial stages that a rise in the content of Si in the islands affects their subsequent growth, since this reduces the value of their elastic energy, and, correspondingly, brings about an increase in the critical volume V_{cr}, at which the transition to the dome-like shape occurs. That is why, for the $Si_{0.9}Ge_{0.1}$ buffer, to obtain the ensemble of nanoislands, exclusively dome-like in shape, the nominal thickness of the deposited Ge layer should be increased to 13 MLs, whereas it equals 11 MLs for the Si buffer.

It is seen from Fig. 1,*a—c* that, depending on the MBE parameters and a kind of the buffer layer, the formed islands are different in shapes, average heights, and surface densities. The values of these quantities, determined from the AFM images, are shown in Table 1.

T a b l e 1. MBE conditions and parameters of SiGe nanoislands determined from the AFM studies

Temperature of epitaxy, °C	Number of Ge monolayers	Buffer layer	Surface density, cm^{-2}	Shape of islands	Mean island height, nm
500	8	Si	3.0×10^{10}	hut-clusters	2
700	4.2	$Si_{0.9}Ge_{0.1}$	3.1×10^{9}	pyramids	10
700	11	Si	2.1×10^{9}	domes	28
700	13	$Si_{0.9}Ge_{0.1}$	2.6×10^{9}	domes	30

As is seen from Table 1, it is the hut-clusters that have the least heights (∼2 nm) and the greatest surface density (3×10^{10} cm^{-2}). These values equal 8 nm and 3×10^{9} cm^{-2} for pyramids, and 28 nm and 2.5×10^{9} cm^{-2} for domes, respectively. The analysis of the AFM images obtained by us has shown that the total volume of the islands formed at the temperatures of 600 °C and higher exceeds far the nominal volume of deposited germanium. This fact is reasonable, since we showed earlier [1, 10] that, at a temperature of epitaxy 700 °C, the content of Si in islands is more than 40%, which is brought about by a high degree of the component intermixing resulting from the intense Si diffusion from the buffer layer. Since the maximal stress gradient is realized along the island perimeter, i.e. near the boundary with the WL [11], the maximal diffusion flow goes just from this area of the substrate. In this case, the atoms from the WL diffuse at first, while those from the buffer layer, either Si or $Si_{0.9}Ge_{0.1}$ one diffuse later on. The cavities around an island that divide the formed islands and penetrate by 2—3 nm into Si or the $Si_{0.9}Ge_{0.1}$ buffer layer (see Fig. 1) serve as a confirmation of this fact.

Fig. 3 shows the RS spectra for the structures with nanoislands of various shapes. To quantitatively interpret the experimental spectra, the spectrum of the Si or $Si_{0.9}Ge_{0.1}$ substrate was subtracted from each of them. As was shown in works [1, 12], the contribution to the RS spectra from the Ge WL which is sufficiently thin is marginal and can be neglected. It should be noted that, with the aim to prevent the oxidation of small hut-clusters ($h \approx 2$ nm) in air, they were covered with a silicon layer 50 nm in thickness. Of course, this results in some change of the island characteristics, which will be discussed below. The effect of the SiGe component intermixing becomes apparent from the presence of the bands that correspond to both Ge—Ge, Si—Ge, and Si—Si oscillations in the islands. It is known that, for a $Si_{1-x}Ge_x$ solid solution, the frequency of each mode depends on both the component composition x and the elastic deformation ε according to the relations [3, 13]:

$$\omega_{\mathrm{SiSi}} = 520.5 - 62x - 815\varepsilon, \qquad (1)$$

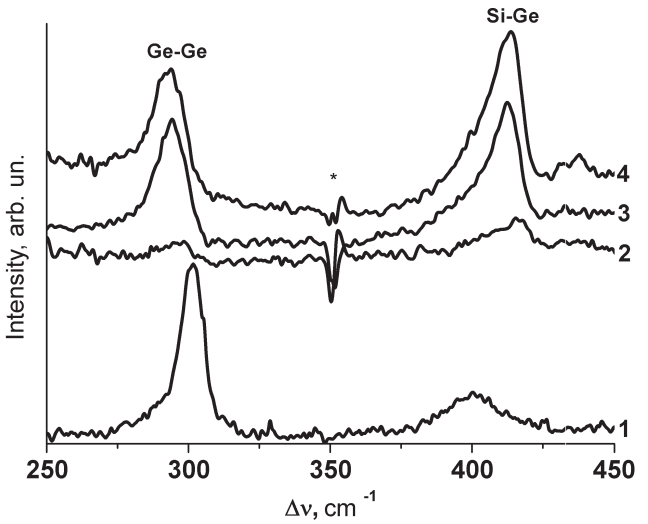

Fig. 3. RS spectra for the structures formed at 500 °C by means of the deposition of 8 MLs of Ge on Si (*1*) and at 700 °C by means of the deposition of 4.2 MLs of Ge on $Si_{0.9}Ge_{0.1}$ (*2*), 11 MLs of Ge on Si (*3*) and 13MLs of Ge on $Si_{0.9}Ge_{0.1}$ (*4*) buffer layers

$$\omega_{\mathrm{GeSi}} = 387 + 81(1 - x) - 78(1 - x)^2 - 575\varepsilon, \qquad (2)$$

$$\omega_{\mathrm{GeGe}} = 282.5 + 16x - 385\varepsilon. \qquad (3)$$

As a result of the solution of the system of equations (1)—(3), the values of both the component composition and elastic deformations were obtained for the islands formed at various technological parameters of MBE (Table 2). As is seen from Table 2, there is a noticeable amount of Si in the islands, although, according to the classical Stranski—Krastanow mechanism of growth [14], the formation of purely germanium islands should occur. The intensity of the diffusion caused by inhomogeneous stresses within the islands and around them decreases with the stresses and the Si concentration gradient. For this reason, there is no linear dependence between the duration of the Ge deposition process and the content of Si in islands.

As regards the elastic deformations in islands, they depend on both the component composition and the island shape.

T a b l e 2. MBE conditions and parameters of SiGe nanoislands determined from the RS spectra

Temperature of epitaxy, °C	Number of Ge monolayers	Buffer layer	Covering Si layer	Elastic deformation $\varepsilon_{\parallel}$, %	Content of Ge in $Si_{1-x}Ge_x$ islands	Degree of stress relaxation δ, %
500	8	Si	+	−3.9±0.1	0.88±0.02	—
700	4.2	$Si_{0.9}Ge_{0.1}$	−	−1.5±0.1	0.54±0.03	30
700	11	Si	−	−0.8±0.05	0.52±0.02	60
700	13	$Si_{0.9}Ge_{0.1}$	−	−0.9±0.05	0.39±0.02	40

Estimate the contribution of each of these factors to the island relaxation. For the dome-like islands formed by means of the deposition of 11 MLs of Ge on the silicon buffer (Fig. 1,*c*), the values of the elastic deformation and component composition, determined from the RS spectra, equal $\varepsilon_{\text{exp}} = -0.008$ and $x = 0.52$, respectively. For a completely strained layer of $Si_{1-x}Ge_x$ with the same x value, which is deposited on Si, the elastic deformation is $\varepsilon_x = (a_{\text{Si}} - a_{\text{SiGe}})/a_{\text{Si}} = -0.019$. Since the islands of such dimensions still do not contain dislocations [15], the additional relaxation occurs in them owing to an increase in the lattice constant with the nanoisland height, i.e. owing to the geometric factor. Let us introduce a quantity which characterizes the degree of the stress relaxation originating from the three-dimensionality of the shape: $\delta = (\varepsilon_x - \varepsilon_{\text{exp}})/\varepsilon_x$, where ε_{exp} is the elastic deformation determined from experimental RS spectra, ε_x is the elastic deformation for the completely stressed layer of $Si_{1-x}Ge_x$ grown on the Si substrate. For the islands formed by means of the deposition of Ge layers various in thickness, the degree of the relaxation δ caused by the geometric factor is shown in Table 2. Since the contribution originating from the geometric factor is negligible for the relaxation of the small islands covered with silicon, the corresponding value is not displayed in Table 2.

An analysis of the stresses in the islands not covered with silicon indicates that it is domes that have the smallest value of elastic deformation and the greatest degree of relaxation, as concerns the contributions caused by the geometric factor (see Table 2). This result is reasonable, since the ratio of a height to a lateral dimension is greatest for domes (1/6), intermediate for pyramids (1/10), and smallest for hut-clusters (1/10 in one direction and varies from 1/10 to 1/40 in the other one). For the islands, which are the same in shape but different in dimensions, a greater degree of relaxation will be in the islands which are smaller in size. This is brought about by the fact that the lattice structure on the island surface and within the near-surface layers corresponds to the undistorted lattice. Actually, as is seen from Table 2, the domes formed on the Si buffer are smaller in size and have a greater degree of stress relaxation, as compared to the domes formed on the $Si_{0.9}Ge_{0.1}$ buffer layer. It is also evident from Table 2 that the hut-clusters are characterized by high values of the elastic deformation. This is caused by a marginal content of Si in them, since they were formed at 500 °C and additionally subjected to a hydrostatic pressure from the side of the covering silicon layer.

The island parameters determined from the RS spectra, namely the elastic deformation, degree of a stress relaxation, and component composition, bring about a modification of the band structure of the island. This leads, in turn, to a change in the emitting characteristics of the structures with nanoislands.

Consider one more important characteristic of nanoislands, namely their space ordering. It is known that the nucleation of islands on a Ge WL is a spontaneous process which results in their disordered formation on the specimen surface [14]. On the other hand, the production of opto- and nanoelectronic devices with controlled parameters, based on the structures with nanoislands, requires the achievement of the ordering of nanoislans both in size and shape, as well as their ordered arrangement on a specimen surface. As follows from investigations, it is easier to realize a sufficiently narrow distribution in sizes, than to solve the problem of space ordering. In particular, for the dome-like islands formed at T=700 °C, if the thickness of the deposited layer equals 11 MLs, the scatter in size does not exceed 10 %. For the domes, the narrow distribution in size is explained by the fact that, as a dome grows, the increase in the elastic deformation in both the substrate and the dome base reduces its growth rate, the effect being opposite to the Ostwald ripening [16]. However, for these islands, a relatively small scatter in size does not affect their lateral ordering (Fig. 1,*c*).

A general idea of the approaches recently used for the formation of an ordered ensemble of nanoislands consists in the utilization of the idea of the creation of a regular direction on the growth surface, along which the local barrier for the island nucleation is lower compared to those in other directions. Such easy direction, which corresponds to the smallest activation energy for the island nucleation, can be set by the elastic crystal anisotropy or a local decrease in the mismatch of the lattice constants of the WL and islands.

It was calculated in work [17] that a considerable interaction between neighboring small islands occurs by means of stress fields in a substrate. This can lead to the ordering of islands in two directions. In particular, due to the elastic anisotropy characteristic of the crystals with cubic symmetry, a minimum of the full energy for an island ensemble is achieved when they are formed in such a manner that a side of the pyramid base is parallel either (100) or (010) directions (see Fig. 1,*a*). For the islands which are greater in size and more distant from one another, the elastic interaction through the substrate becomes negligible and does not affect their ordering any more (see Fig. 1,*b*). However, the AFM studies show

that, in this case, some island chains are formed in parallel to the elastically pliable directions.

The space ordering of islands becomes enhanced as the degree of surface covering (the ratio of the total area of islands to that of the substrate) increases. This effect is caused by the minimization of the repulsive forces of the elastic interaction between neighboring islands. Thus, the most ordered island arrays occupy the greater part of the buffer layer area, and the islands practically touch one another (see Fig. 1,*d*). Such island ensemble can be formed on stressed $Si_{1-x}Ge_x$ buffer layers. In fact, it is with the use of the MBE of Ge (d_{Ge} = 13 MLs) on the $Si_{0.9}Ge_{0.1}$ buffer layer at 700 °C that a partially ordered ensemble of dome-like islands was obtained, the parameters of which are presented in Tables 1 and 2.

For the multilayered structures with islands, the vertical ordering is achieved due to the influence of the elastic fields of islands already formed on the structure of the next WL, on which the islands are nucleated. We have studied such multilayered structure fabricated by means of the fivefold repetition of the procedure, which includes the deposition of 7.5 MLs of germanium at 600 °C and the subsequent covering of the formed islands with a silicon layer 26 nm in thickness. The image of this structure obtained with the use of transmission electron microscopy gives a clear evidence for the vertical correlation of the island growth (Fig. 4). As was shown in work [18], the characteristic of a multilayered structure is that each WL, beginning from the second one, is smaller in thickness than the first one. This is brought about by the influence of the mechanical stresses created in a silicon spacer by the islands of a previous layer, which are already formed. The stresses within the spacer are distributed in such a way that it becomes stretched over the islands and compressed between them. That is why, the critical thickness of the 2D—3D transition will be observed at a smaller thickness for the germanium layer deposited on such silicon spacer, than for that on unstressed silicon. Germanium atoms will be assembled into islands in those places, where a lattice is stretched, i.e. over the islands already formed within a previous layer. A decrease in the thickness of the WL results in that germanium forming the islands remains in the amount approximately by 20 % more than that in the first layer. As a result, beginning from the second layer, the growth in the size of islands is observed in the multilayered structure. To obtain islands of the same dimension in each of the layers, it is necessary to reduce the amount of deposited Ge, beginning from the second layer.

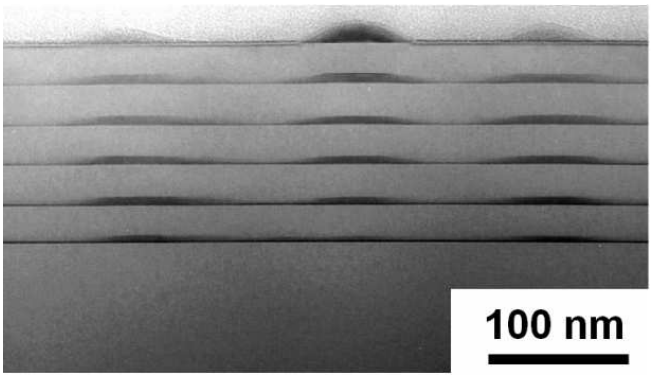

Fig. 4. Image of a multilayered structure with nanoislands obtained with the use of transmission electron microscopy

4. Conclusions

In this work, by means of a proper choice of both the thickness of a deposited Ge layer and the temperature of epitaxy, three types of the ensembles of nanoislands of a certain shape are formed. It is established that the degree of stress relaxation due to the geometric factor is greater for the non-covered dome-like islands, than for the pyramidal ones. For the nanoislands of the same shape, the relaxation is the greater, the smaller their dimensions are. The smallest scatter in dimensions not exceeding 10 % is observed in the dome-like islands, and this is explained by the self-restricting growth of the islands, which are greater in size. It is also shown that the utilization of a $Si_{0.9}Ge_{0.1}$ buffer layer favors the lateral ordering of islands.

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ВЛАСТИВОСТІ ТА ВЗАЄМОПЕРЕТВОРЕННЯ САМОІНДУКОВАНИХ SiGe-НАНООСТРІВЦІВ РІЗНОЇ ФОРМИ

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Р е з ю м е

Методами атомної силової мікроскопії та комбінаційного розсіяння світла досліджено SiGe-нанооcтрівці, сформовані на кремнієвому та $Si_{0,9}Ge_{0,1}$ буферних шарах. З метою дослідження острівців певної форми (hut-кластерів, пірамід, куполів) було отримано одномодові масиви острівців цієї форми шляхом варіювання товщини осадженого шару Ge та температури епітаксії. Встановлено, що серед незарощених острівців найбільші пружні деформації виникають у пірамідальних, а найменші — у куполоподібних острівцях. Ступінь релаксації напружень за рахунок геометричного фактора найбільший у мінімальних за розмірами куполоподібних острівцях. Показано, що використання $Si_{0,9}Ge_{0,1}$ буферного шару для вирощування на ньому острівців сприяє їх латеральному впорядкуванню.