Influence of the Preliminary Annealing Conditions on Step Motion at the Homoepitaxy on the Si(100) Surface

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***Abstract* – In this paper, the motion of steps S_A and S_B on the Si(100) surface in the process of Si Molecular beam epitaxy (MBE) is explored. The study was carried out by means of the reflection intensity dependence behavior analysis of reflection high-energy electron diffraction (RHEED) corresponding to the (2×1) and (1×2) reconstructions. Superstructural rearrangement from a two-domain to a single-domain surface is associated with the bilayer step formation, which occurs due to the different motion rates of the steps S_A and S_B. Based on the research conducted, the conditions under which the step doubling occurs were determined. A behavior analysis of the diffraction reflection intensity dependences showed that an increasing of preliminary annealing time and temperature facilitates to the faster convergence of the steps S_A and S_B, but to the slower recovery of the initial surface. The presented experimental results indicate that step movement rate difference depends on the step A edge kink density.**

***Index Terms* – Reflection high-energy electron diffraction, molecular beam epitaxy, Si(100) surface, reconstruction, elementary steps.**

I. INTRODUCTION

THE MODERN CRYSTALLINE GROWTH techniques, such as Molecular beam epitaxy and Chemical vapor deposition (CVD), provide the ability of creating the nanostructures for solid-state devices, revolutionized electronics [1]. The electronic circuit element base development of the new generation is constantly maintained by efforts, whose goal is introducing the innovative high technologies in the semiconductor device creation [1]. A fundamental understanding of the energetic and atomic process rates of epitaxial growth involved in the device creating technology constitutes an important research area [2, 3]. One of the main and primary tasks of such studies is the control accuracy increasing of the growth mode under different physical conditions. In view of the sharply growing interest in the low-dimensional systems, the need in the new experiment setting and more profound theory development of the crystal growth appeared [4-8]. The previously unresolvable epitaxy tasks become realizable due to the analytical and technological component improvement of MBE installation.

There are two step types of monatomic height. One is perpendicular to the upper terrace dimer rows and is called the S_B step, and the other is parallel to the upper terrace dimer rows and is called the S_A step [9]. As a consequence of the various speeds of these step movement, both monatomic steps (the two-domain surface) and double steps (the single-domain surface) can be present on the Si(100) surface [10-13]. The influence of the misorientation angle and growth conditions on the difference between the step rates is the nontrivial task, which is solved by considering the step thermodynamics and kinetics [14-17]. The kinetic criterion for the single-layer step stability with a slightly misoriented Si(100) surface is derived basing on quantitative considering the combination of the surface diffusion and growth kinetics anisotropy processes [2]. The diatomic steps are formed at a temperature below the critical temperature depending on the misorientation degree [2]. It was previously shown [10-13, 18, 19] that a single-domain surface is formed in a relatively low temperature region and a two-domain surface – in the high temperature region. At high temperatures, the homoepitaxial growth occurs by the mechanism due to the step movement: the adatoms condensed on the terrace surface diffuse before the collision with the step edge and at the step edge they are easier embedded into the crystal. The monatomic steps on the crystalline surface serve as sinks for the adatoms [16, 17]. However, the early results demonstrate that some adatoms can jump over the step edge [20]. In this connection, the problem of vicinal surface instability was raised. The growth at the step edge is determined by the rate at which the adatoms can reach the step edge from the upper or lower terrace. In turn, these transition rates are governed by different energy barriers for the atom motion on the surface [3, 21]. By analyzing the step movement rates, one can understand the character of the adatom incorporation into the steps and determine the energy barrier magnitudes, as well as the step edge structure [3, 21-24].

The investigations of transition dynamics were carried out earlier by the authors of [25, 26], where the main attention was paid to the Si(100) plate misorientation. In our work, the investigations of the influence of preliminary

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annealing conditions on the transition dynamics were carried out.

II. PROBLEM STATEMENT

The fundamental scientific task is to establish the physical mechanisms of the epitaxy proccess kinetics of Si and Ge at elementary steps and their edges on a Si(100) substrate, inclined less than 0.5°. The epitaxy process represents complex elementary acts regularity, performed by adatoms on the surface. The difference in the step movement rate is due to the potential energy surface relief, as well as the step edge structure. By analyzing the step movement rates, one can obtain information about the character of the adatom incorporation into the steps.

III. EXPERIMENTAL TECHNIQUE

The silicon layer growth was carried out in the Katun-S MBE installation equipped with the electron beam evaporators for Si and Ge. The analytical chamber part consists of the quadrupole mass spectrometer, quartz thickness measurement and the RHEED at the energy of 20 keV. The investigations were carried out on a Si(100) substrate, which was preliminarily cleaned to obtain a clean and smooth surface. The film surface structure and morphology control were performed with the help of RHEED. The details of the experiment technique are mainly described in [18]. The diffraction conditions for each dependence had slight differences in the electron beam incidence angles and azimuthal angles. The RHEED patterns were recorded in the azimuthal [100] direction. The reflex intensities located on the fractional order Laue zone were analyzed. To achieve steady-state growth conditions, the diffraction pattern video recording was made in two to three minutes after changing the silicon atom deposition conditions. Fig. 1 shows the RHEED pattern and change in the reflex intensity over time.

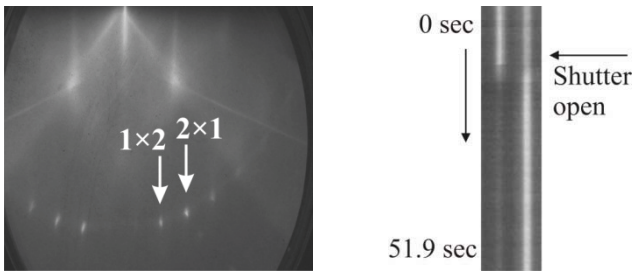

Fig. 1. RHEED pattern and change in the reflex intensity over time.

IV. EXPERIMENTAL RESULTS

The change in the diffraction pattern at opening and closing (in the Si atom flux presence and in its absence) the shutter was investigated. The experimental conditions were as follows: the Si(100) substrate was annealed at 900°C for 5, 20, and 40 minutes without the Si atom flux, then cooled to the substrate temperature of 600°C. The profiles across the (2×1) and (1×2) reflexes were recorded when opening and closing the shutter at the substrate temperature of 600°C and the growth rate of 0.3 Å/s. The dependences of $I_{2\times1}$ and $I_{1\times2}$ intensity maxima on the number of monolayers obtained at 600°C after opening and closing the shutter are shown in Fig. 2. The $I_{2\times1}/I_{1\times2}$ intensity ratio dependences on the number of monolayers obtained at 600°C after opening the shutter at the preliminary annealing during 5, 20, and 40 minutes are shown in Fig. 3.

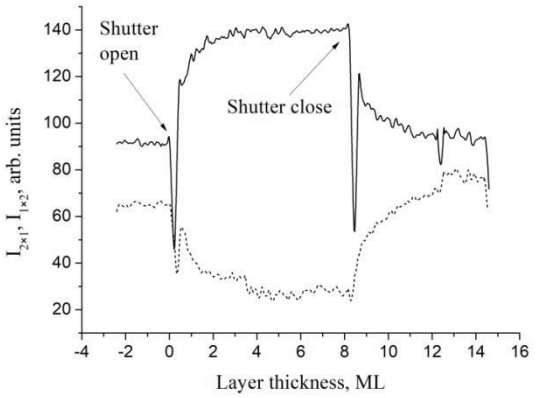

Fig. 2. The dependences of $I_{2\times1}$ (the solid line) and $I_{1\times2}$ (the dotted line) intensity maxima on the number of deposited monolayers. The preliminary annealing is 40 min.

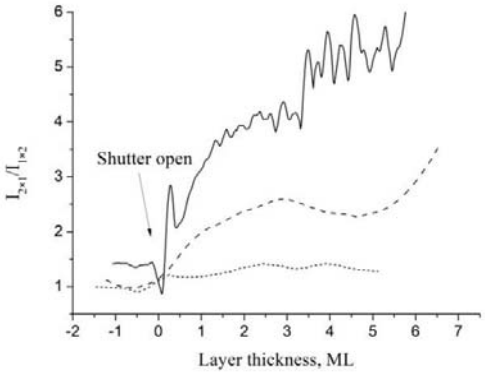

Fig. 3. The $I_{2\times1}$ and $I_{1\times2}$ intensity ratio dependences on the number of monolayers at different annealing times. The solid line is 40 min., the dotted line is 20 min. and the dotted line (the lowest) is 5 min.

The change in the reflection high-energy electron diffraction after closing the shutter (after the Si atom flux interruption) was investigated. The experimental conditions were as follows: the Si(100) substrate was annealed at the temperature of 900°C for 5 and 40 minutes without the Si atom flux (shutter closed), then it was uniformly cooled to the substrate temperature of 600°C and at this temperature, the Si atoms were deposited for 2-3 minutes (growth rate 0.3 Å/s before closing the shutter). The $I_{2\times1}/I_{1\times2}$ intensity

ratio dependences on time were obtained at 600°C after closing the shutter at different preliminary annealing times (Fig. 4). Similarly, the experiments at the annealing temperatures of 900°C and 1000°C for 5 minutes were carried out (Fig. 5).

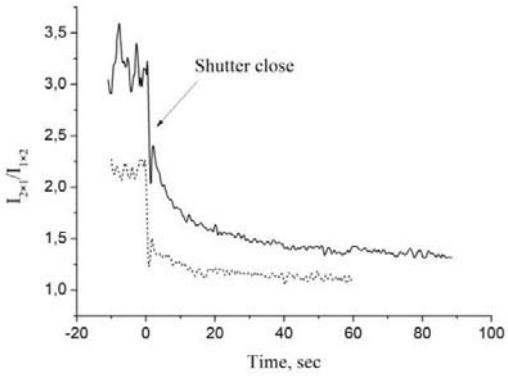

Fig. 4. The $I_{2\times1}/I_{1\times2}$ reflex intensity ratio dependences on time. The solid line represents the dependence obtained at the annealing temperature of 900ºC for 40 minutes, the dotted line represents the dependence obtained at the annealing temperature of 900ºC for 5 minutes.

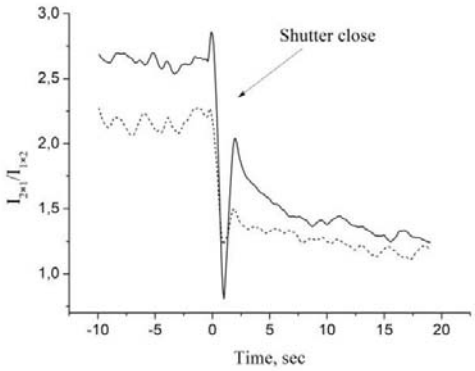

Fig. 5. The $I_{2\times1}/I_{1\times2}$ reflex intensity ratio dependence on time. The solid line represents the dependence obtained at the annealing temperature of 1000ºC for 5 minutes, the dotted line represents the dependence obtained at the annealing temperature of 900ºC for 5 minutes.

V. DISCUSSION OF RESULTS

Because of the inhomogeneous potential energy surface relief [22, 23], which influences the adatom elementary act regularities, the epitaxy process is of complex character. The Ehrlich-Schwoebel energy barrier [27, 28] plays an important role in the surface morphology evolution. The barrier magnitude at the adatom jump from the upper to the lower terrace and vice versa influences the propagation rate of steps A and B. The step motion rate also depends on the features of adatom incorporation into the step edges [20]. Namely, the adatom movement along the step edge with its further embedding into the kink depends on the kink density.

A schematic adatom concentration dependence on the terrace surface at the distance (above) and the terraces with the adatoms on it (below) is shown in Fig. 6. The adatom motion on terrace B surface occurs in the fast diffusion direction, while leaving some depleted adatom zone. The adatom motion on terrace A surface occurs in the slow diffusion direction, forming the depleted zone of a smaller size than that on terrace B [9]. The overlap of diffusion field leads to the adatom concentration decrease in the terrace center. As a result, the adatom concentration in the A terrace center is higher than that in the B terrace center. Thus, it can be noted that the atom fluxes to steps A and B from terrace A are smaller than those from terrace B. The step A and B different energy barrier existence cannot be ruled out [3]. The reason for the equidistant step system instability may be higher barriers for the adatom embedding from the upper and lower terraces in the A step, than in the B step. In the presence of the significant Ehrlich-Schwoebel barrier, the adatom arrival from the upper terrace to the lower terrace will be difficult and, together with it, the step motion rate will decrease [3, 29].

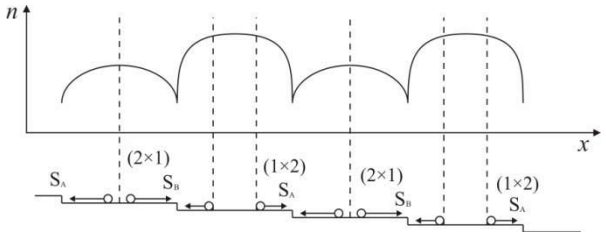

Fig. 6. Schematic representation of the atom distribution on the terrace surface. The schematic atom concentration dependence on the distance is shown above. The terrace image and the atoms on the terraces are shown at the bottom.

The adatom embedding process in the step A and B edges is schematically shown in Fig. 7. It is known that step B moves faster than step A. The different step rate of types A and B may be due to the presence of the long region of type A step edge. If the atom migration length along the step edge is less than the distance between the kinks at the step edge, then the atoms will be reflected or jump over the type A step. Thus, the atoms will not be held on the type A step edge, and the step will grow at a slower rate relative to the type B step.

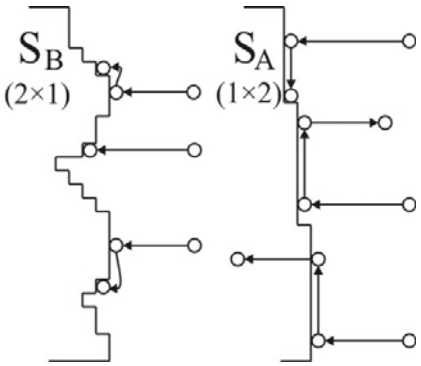

Fig. 7. Schematic representation of the adatom embedding process in the steps.

With the annealing time increase, the transition from the two-domain to the single-domain surface occurs faster, but the recovery to the two-domain surface is slower. This may mean that annealing influences the step edge structure that leads to the increase in the difference between the step rates. The step A smooth edge formation process at the annealing is schematically shown in Fig. 8. The atoms are detached from the kinks, moving along the step edge, followed by the embedding into the kink or detaching from the step edge. Thereby, the step region is left atomically smooth.

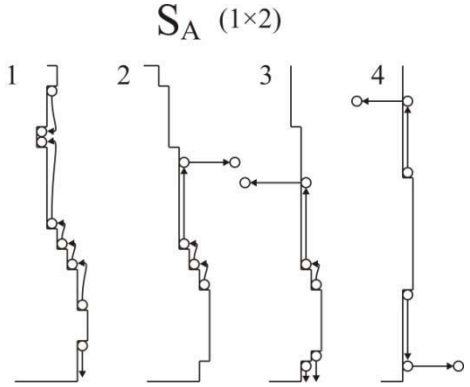

Fig. 8. Schematic image of the step A smooth edge formation process at the annealing.

The change in the rate of surface domain structure depends on the preliminary annealing conditions. At annealing, the step A and B edge structure tends to the equilibrium structure, determined by the specific energy value of this or that step. Namely, the A-step edges are becoming smoother, and the B-step edges are rough. The adatom embedding in the A-step occurs as a result of the elementary acts of attachment, separation and migration along the smooth (without the kinks) step edge region. If the length of atom migration along the step edge is shorter than the distance between the kinks, then the adatoms will detach from the step edge before their embedding into the kink. Thus, the A-step movement rate depends on the distance between the kinks. The longer the distance between the kinks relative to the adatom migration length along the step is, the lower the step rate is. In the case of the rough B-step, this effect is insignificant. The adatom is likely to fall into the kink either directly or as a result of a short migration along the step edge. Therefore, the kink concentration decrease on the A-step after the annealing leads to the slower movement of this step relative to the B-step with a subsequent growth and, consequently, to the more rapid A and B-step convergence, and that contributes to the transition from the two-domain (2×1)+(1×2) to the single-domain (2×1) surface.

The reverse transition to the two-domain surface in the absence of the silicon flux (with the shutter closed) is associated with the atom detachment from the B-step edges (from the kinks), followed by the diffusion on the terraces and embedding in the A-step edges (in the kinks). The atom detachment from the kink requires overcoming a substantially larger energy barrier than the attachment to the kink. Therefore, independently of the kink concentration on the A-step, the reverse transition occurs more slowly than the direct transition. The reverse transition rate decreases with the kink concentration decrease on the A-step (with the average distance increase between the kinks), since the time, during which the adatom "finds" the kink, increases. This explains the decrease of reverse transition rate with the increasing time or annealing temperature.

The experimental results presented indicate that the step motion rate difference depends on the step A edge kink density. Consequently, the Ehrlich-Schwoebel barrier influence on the adatom arrival from the upper to the lower terrace is not significant related to the influence of the presence of a smooth region on the step A edge. This may mean that the direct Ehrlich-Schwoebel barrier with the adatom arrival from the upper to the lower terrace does not have a large value and does not have a strong effect in the step movement.

VI. CONCLUSION

As a result of the research conducted, the conditions under which the step doubling occurs were determined. A behavior analysis of the diffraction reflection intensity dependences showed that an increasing of preliminary annealing time and temperature facilitates to the faster convergence of the steps S_A and S_B, but to the slower recovery of the initial surface. The presented experimental results indicate that step movement rate difference depends on the step A edge kink density.

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He has extensive experience of work with analytical and technological equipment for Molecular beam epitaxy. For over 35 years he has been engaged in research and technology for the creation of semiconductor nanostructures. Currently, he is a leading researcher and head of laboratory of Molecular beam epitaxy of elementary semiconductors and A_3B_5 compounds. Research interests are in Molecular beam epitaxy of semiconductor nanostructures based on GeSiSn and GeSi materials.

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Vyacheslav Timofeev defended his Ph.D. in 2014. His main investigation area is the search of Ge, GeSi, GeSiSn and SnO(x) film growth mechanisms by molecular beam epitaxy (MBE). According to the results of the studies performed by V.A. Timofeev, 41 articles were published in peer-reviewed journals.

He personally participates in the setting the tasks, carrying out the experiments on the ultrahigh-vacuum MBE installation, obtaining the experimental samples for their further structural and electrophysical studies, processing the experimental data, discussing the results, writing articles and grants. He was awarded the scholarship of the President of the Russian Federation to young scientists and graduate students in 2013 and 2018. He has the experience in managing the grants from the Russian Foundation for Basic Research and Russian Science Foundation. Vyacheslav Timofeev is involved in scientific and pedagogical activities in supervising students.