Control of the formation of a stepped-terrace Surface on $6H$-SiC(0001) substrates by *in situ* reflection electron microscopy

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The formation of stepped-terraced morphology on the vicinal surface of $6H$-SiC(0001) during high-temperature annealing was studied using *in situ* ultrahigh-vacuum reflection electron microscopy in combination with *ex situ* atomic force microscopy. It was established that under ultrahigh-vacuum annealing conditions at temperatures of 1160−1200 °C, step bunching processes are activated, leading to the formation of macrosteps consisting of three SiC bilayers with a height of 0.75 nm without altering the stoichiometric composition. Annealing at 1250 °C for 10 min resulted in step bunching with the formation of the smoothest terraces, accompanied by a carbon buffer layer on the surface resulting from silicon sublimation.

Keywords: graphene, SiC, AFM, REM.

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1. Introduction

Silicon carbide (SiC) continues to draw the significant interest as a promising material for electronics of high power and high frequency, and also as a unique substrate for the growth of broadband semiconductors and 2D materials, such as graphene [1]. The key aspect that determines the quality of the subsequent epitaxial layers, is the state of the initial surface of the substrate. In particular, the presence of wide, atomically smooth terraces separated with ordered steps is critically important for the implementation of the controllable layerwise growth [2]. Formation of wide terraces on vicinal surfaces of SiC in process of high-temperature annealing is the result of complex interaction of thermodynamic and kinetic factors. The most important observed phenomenon in this context is the grouping of steps or stepping up — the process, when the initially evenly distributed atomic steps are combined to form higher macrosteps [3,4] and, as a result, expanding the terraces between them.

For deeper study of the dynamics of processes causing formation of wide terraces, in this paper, apart from *ex situ* methods for the study of the morphology and structure of the sample surface, methods of *in situ* diagnostics are used, which make it possible to tract the evolution of surface morphology in real time. Earlier we demonstrated the possibility to control graphitization of the $6H$-SiC(0001) surface in the process of high-temperature annealing, and the conditions were defined for the formation of a buffer carbon layer and a single layer of graphene by method of *in situ* reflected high energy electron diffraction (RHED) [5]. Graphitization may be controlled by this method through the change in the superlattice structure of the initial substrate on $(6\sqrt{3} \times 6\sqrt{3})$R30° in process of the buffer layer formation, and when one layer of graphene is formed, the superlattice structure of the buffer layer $(6\sqrt{3} \times 6\sqrt{3})$R30° changes to the superlattice structure of graphene (1×1). But stepping up of the steps changes the surface morphology, and the superlattice structure remains unchanged, therefore stepping up may not be controlled by *in situ* observations of the RHED pattern. In its turn the mirror electron microscopy (MEM) is used to track the changes of surface morphology and atomic structure directly in the process of high temperature annealing, which is suitable for tracking the stepping up process in real time mode. This method also makes it possible to directly relate the kinetics of the observed processes, such as grouping of steps and smoothening of step terraces and the specific thermodynamic conditions [6].

The study of the dependence between the process of steps stepping up and the annealing time is the central objective, since these parameters may be the ones to be used in the development of technological modes of industrial epitaxial growth plants and make it possible to quantitatively determine the energy barriers of key processes, such as the surface diffusion and mass transfer between the terraces. Therefore, the determination of the annealing parameters makes it possible to optimize the parameters of pre-epitaxial preparation of substrates for the achievement of the required quality of the surface under the conditions of large scale production and is necessary to build a complete physical pattern of terrace formation on the SiC surface.

2. Method for the experiment and measurements

The studies were conducted on the samples with size of $8.8 \times 1.5 \times 0.33$ mm cut from commercial plates of silicon carbide of $6H$ polytype, oriented in direction (0001) with a deviation of 0.5° and doped with nitrogen that acts as a donor impurity. The prepared samples were exposed to chemical treatment with petroleum ether and placed into a chamber of ultrahigh vacuum mirror electron microscope (UHV MEM), where they were annealed by resistive heating method by sending direct electric current through them. The UHV MEM method made it possible in the *in situ* mode to observe the formation of the stepwise-terraced morphology on the surface of $6H$-SiC(0001) in process of annealing. The MEM operation principle is based on the fact that the beam of high energy electrons with energy of 100 keV falls on the crystalline surface of the sample at Bragg angles of around several degrees. Since the angle of electron incidence on the surface is small, the electrons penetrate the sample volume into the depth not exceeding the units of nanometers, accordingly, the elastically scattered electrons carry information only on near-surface layers. The intensity of the diffracted beams depends on the structure of several atomic planes near the crystal surface, which provides for the high sensitivity of this method to the surface layer structure. Therefore, this method makes it posssible in the *in situ* mode to visualize the morphology of the crystal surface at the level of individual atomic steps, and also makes it possible to observe morphological transformations related to the superstructural transitions, epitaxial growth and sublimation [7–9]. The feature of the UHV MEM is the compression of the image in the direction of electron beam incidence ~ 50 times. Besides, this method makes it possible to switch directly in process of the experiment from the mode of MEM-image to the mode of microdiffraction (reflected high energy electron diffraction, RHED). The temperature of samples was monitored using an optical pyrometer.

Upon completion of the experiments on high temperature annealing and further cooling. the samples were withdrawn from the UHV chamber to atmosphere for detailed *ex situ* analysis of the surface morphology by the method of atomic force microscropy (AFM, Bruker Multimode 8). The processing and quantitative analysis of the AFM data were carried out using Gwyddion software.

3. Results and discussion

For the first time in the real time mode the UHV MEM method was used to obtain the image of the initial surface of the substrate $6H$-SiC(0001) prior to the start of the thermal annealing (Figure 1, *a*), which shows a stepped-terraced surface with nanoscratches and contamination, which corresponds to the AFM image of the sample surface prior to annealing shown in Figure 1, *b*. The light contrast on the MEM-image corresponds to the regions of the surface, which in all points meet the Bragg diffraction equation, in other words, the light contrast corresponds to terraces (indicated with number *1* in Figure 1, *a*), matching the crystallographic planes (0001). In its turn the dark contrast on the MEM-image corresponds to the regions of the surface and the objects on it, where the diffraction equation is not met. Thus, the atomic steps separating the neighboring crystallographic terraces, differing in height by one interplane distance, are those objects on the surface, near which the diffraction equation is not met, and from which the dark contrast is formed on the MEM-image. Note that on the MEM-image the steps do not keep straight, contrary to the steps on the AFM image, in connection with the fact that the MEM-image of the surface is compressed in ~ 50 times in the direction of electron beam incidence. Scratches and contamination indicated with numbers *2* and *3*, accordingly, also have the dark contrast, at the same time the deeper the scratch, the darker contrast it has.

The crystalline lattice of polytype $6H$-SiC is characterized by laying six Si-C bilayers along the axis [0001] in the ABCACB sequence with the lattice constant of $d = 1.5$ nm [10,11]. Designations A, B and C in SiC crystallography are related to three possible lateral positions of dense-packed bilateral Si-C layers, which differ from each other by a shift in the basis plane and, in case of the C-layer position, by an additional rotation of the lattice by 60° relative to the previous layer, which in a combination determines various sequences of laying for different polytypes. The height of one step is $d/6 = 0.25$ nm, which corresponds to the distance between the neighboring bilayers in a $6H$-SiC crystal. Every three bilayers form structural ABC and ACB blocks [3,11], which is clearly seen on the MEM-image (Figure 1, *a*) in the form of a darker contrast of the step.

Further annealing at $T = 1000\,^{\circ}$C improves the surface structure and morphology. The surface becomes cleaner, since contamination flies away, and scratches become blurred and disappear. When the annealing temperature increases up to $\approx 1120\,^{\circ}$C, no other changes in the morphology and structure of the surface of the studied samples — MEM-images of the SiC surface in process of annealing at this temperature in general do not undergo significant changes relative to the MEM-image, presented in Figure 1, *a*.

The change in the surface structure may also be tracked by the change in the diffraction patterns (see inserts in Figure 1, *b* and *c*). A typical diffraction pattern (DP), obtained by RHED method from the initial substrate (see insert in Figure 1, *b*) shows the vertical rod reflexes in the Laue zero zone, specific for the SiC substrates with a hexagonal polytype. The central reflex has the maximum intensity, which indicates high perfection of the volume crystalline structure of the substrate. Side reflexes are characterized by blurring and low intensity, which in a combination with the undeveloped system of Kikuchi lines indicated the presence of the structural imperfections of the initial surface of the substrate, which correlates with the AFM-image in Figure 1, *b*. After annealing at $T \approx 1120\,^{\circ}$C

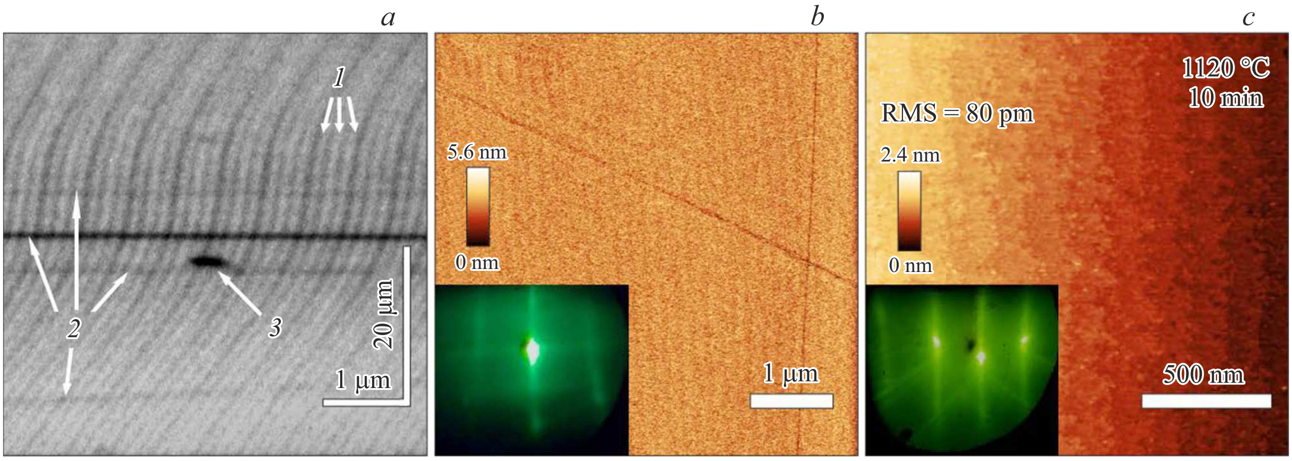

Figure 1. *a* — MEM-image of the surface in process of annealing at $T < 1120\,^\circ$C, arrows *1, 2, 3* indicate terraces, scratches and contaminations, accordingly; *b* — AFM-image of the sample surface prior to annealing; *c* — AFM-image of sample surface after annealing at $T = 1120\,^\circ$C for 10 min. The inserts show DPs along the direction [1100], corresponding to AFM-images.

the side reflexes on DPs (see insert in Figure 1, *c*) became more intensive and narrower compared to the DPs prior to annealing, the system of the Kikuchi lines became visible, which means the improvement of the crystalline structure of the surface layer. Nevertheless, at annealing temperature of up to 1120 °C the DP will not change in principle.

The AFM-image of the sample surface prior to annealing (Figure 1, *b*) is a stepped-terraced rough surface (with mean square roughness of the surface RMS ≈ 500 pm) with soft edges of the steps, where the nanoscratches are present with the depth of up to 5 nm, which appeared in process of plate polishing, agreeing with the interpretation of MEM-images and RHED patterns. The width of the terraces is within 150−200 nm at the angle of surface misorientation in 0.5° for the studied samples. It should be noted that this angle may slightly vary within a single substrate, which in its turn causes local fluctuations of terrace widths. Under annealing at $T = 1120\,^\circ$C for 10 min the edges of the steps become sharper, and the terraces between them — smoother: mean square roughness of the terrace measured by AFM-images obtained after annealing (Figure 1, *b*), is 80 pm in average.

Under further increase of the sample annealing temperature, the considerable change int he surface morphology occurs, which consists in the clustering of the neighboring steps in a macrostep. In the literature this process is known as „separation of steps“ [3], which directly during annealing may be observed by *in situ* MEM method in Figure 2, *a*. The white arrow *1* shows terraces, which, as in Figure 1, *a*, have the white contrast, and the edge of the step is in the form of the dark contrast. The MEM-image in Figure 1, *a* has wider terraces compared to the MEM-image in Figure 2, *a*, which means that on the surface the step separation process takes place. As it has already been mentioned, 6*H*-SiC crystal contains six Si−C layers oriented in the direction (0001) and laid in the A-B-C-A-C-B sequence. Due to high annealing temperature, the atoms gain sufficient energy to exit the steps and become adatoms. Some of these adatoms may be desorbed from the surface, whereas other diffuse in the terraces and are either built into the edges of the below steps or return to the initial steps. For the adatom moving on the 6*H*-SiC surface, the landscape depends not only on the surface layer, but on the entire sequence of the below layers in the 6*H*-SiC polytype, therefore, each bilayer has crystallographically non-equivalent terraces. That is why the adatoms, in order to move in the surface regions corresponding to different layers must overcome energy barriers of different values to make a diffusion jump between the neighboring positions of adsorption on the layer surface. The atom diffusion rate is higher (diffusion barriers are lower) in B and C layers compared to the A layer [12]. The flows of adatom integration and exit for different steps turn out to be unbalanced. As a result, some steps lose more atoms than they gain, and others — on the contrary, which causes their displacement with different speed. Faster steps catch up with the slower ones every three layers, merging into a single high macrostep.

The described phenomenon has a threshold behavior by temperature, since the process of surface regrouping requires overcoming energy barriers for atom migration on the surface. The step unification process activation temperature may be determined by observing the sample surface using MEM *in situ* method. The structural blocks of three steps, which are seen on the corresponding MEM-image (Figure 1, *a*) start unifying at annealing temperature $> 1160\,^\circ$C, as a result of which the width of terraces between such macrosteps becomes equal to 450−500 nm, which you can see on the AFM-images (Figure 2, *b*−*e*), and the measured height of steps of the studied samples (see insert in Figure 2, *d*) in 0.75 nm is equal to half of the 6*H*-SiC lattice constant in direction [0001]. Therefore, the height of macrosteps and the width of terraces correspond to the tripled height of single steps and trippled width of terraces observed previously on the sample surface.

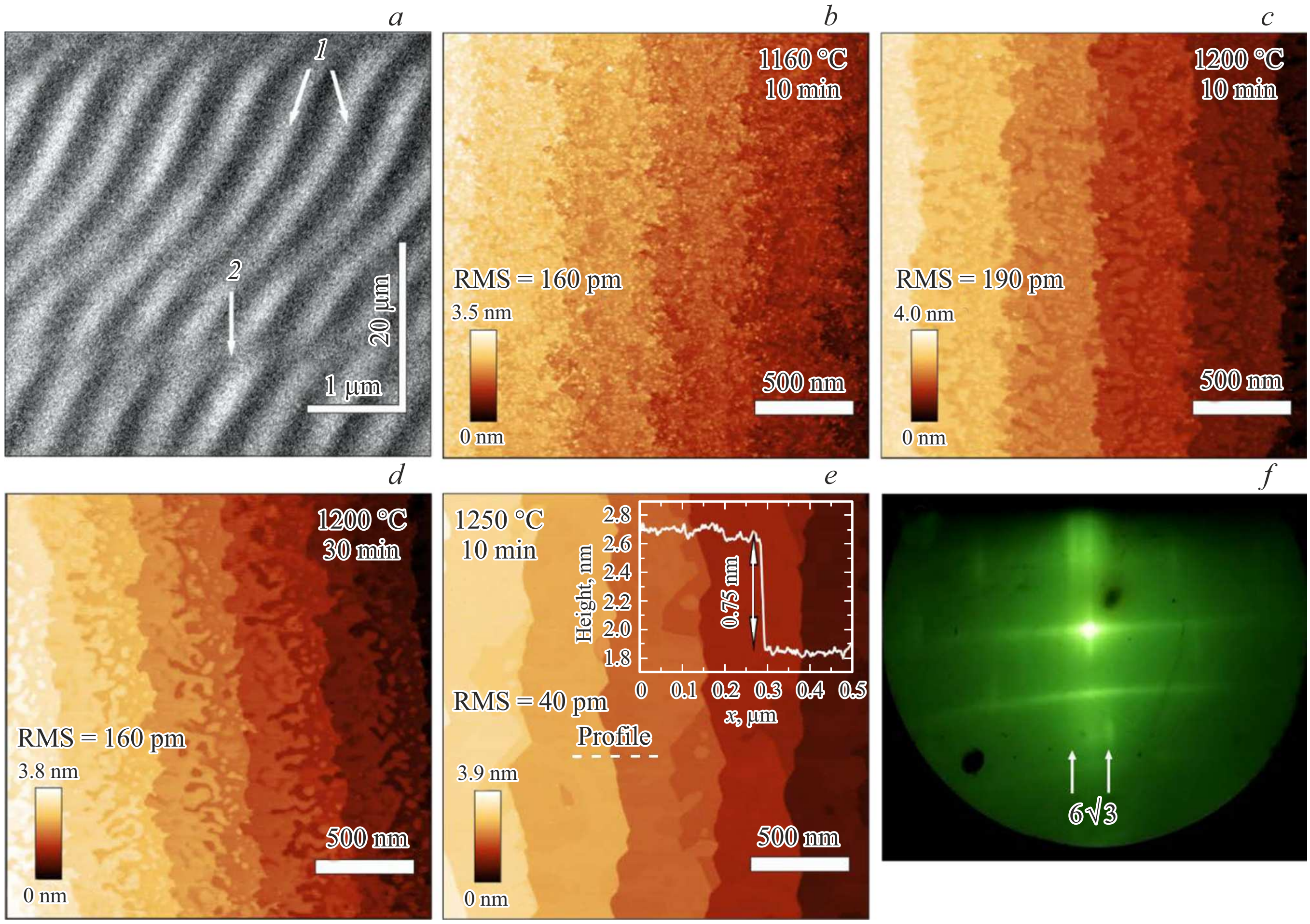

Figure 2. *a* — MEM-image of the SiC surface after 10-minute annealing at $T = 1200\,^\circ$C, arrows *1* and *2* indicate terraces and the scratch, accordingly; AFM-images of the SiC surface after annealing at $T = 1160\,^\circ$C for 10 min (*b*), $T = 1200\,^\circ$C for 10 min (*c*), $T = 1200\,^\circ$C for 30 min (*d*), $T = 1250\,^\circ$C for 10 min (*e*) (the insert shows the profile of the surface measured along the dotted line); *f* — DP, observed after the 10-minute annealing at $T = 1250\,^\circ$C, arrows indicate reflexes corresponding to the superstructure $(6\sqrt{3} \times 6\sqrt{3})$R30°.

You can also observe how small nanoscratches produced by polishing of the substrate surface, are filled with atoms and disappear, and deep scratches do not disappear even at surface transformation, but become blurred and wider, which you can see on the MEM-image (Figure 2, *a*) and as indicated with the arrow *2*. This happens due to the fact that the atom diffusions on the surface at these temperatures are smaller than the step terrace width. Such scratches locally influence the surface morphology, since the step moves and changes the direction near the scratch due to the scratch filling with atoms from the steps and a small length of atom diffusion on the surface.

In order to study the dependence of the surface morphology on the annealing temperature, the experiments of substrate annealing were conducted at different temperatures for 10 min, and AFM-images were taken from each image. At $1160\,^\circ$C the process of step combination is activated, as a result of which the stepped-terraced surface is observed with higher roughness of terraces (RMS = 180 pm) and blured boundaries of the steps (Figure 2, *b*). When temperature increases to $1200\,^\circ$C, the boundaries of the steps become clearer, and you can see formation of islands on the terrace already, which form a new layer on the terraces, i. e. the surface becomes more ordered, which you can see in the AFM-image in Figure 2, *c*. Nevertheless, the roughness of the terrace remains high RMS= 180 pm. Since the RMS value measured across the terraces is half of the average step height, this correlates with the height of the elementary bilayer $c/6 = 0.25$ nm and is specific for the situation, when adatoms migrate in the partially formed terraces, corresponding to the ABC or ACB arrangement, forming partially only the initial stages of the new layer — 2D islands, and not a solid surface, which depends on the diffusion kinetics.

Since the diffusion of atoms on the surface depends on temperature, therefore, formation of smooth terraces on the silicon carbide surface also directly depends on the annealing temperature. To form the terraces fully filled with SiC bilayers, it is necessary to increase the annealing temperature more, in order to increase the length of the adatom diffusion. But as the annealing temperature increases, the silicon atom sublimation speed increases, the surface becomes enriched with carbon, i. e. the surface graphitization happens, which also influences the formation of macrosteps. Further the sample was annealed at temperature 1250 °C for 10 min. In the AFM-image obtained after surface annealing (Figure 2, *e*), you can see that SiC bilayers cover the most fo the terraces, whereas the surface is characterized with the least roughness of RMS = 50 pm relative to all studied samples. Besides, there are no small islands on the terraces, as in previous samples, but on the contrary, pits are observed, which indicates sublimation of silicon atoms from the surface. This is confirmed by the fact that at the end of annealing the *in situ* RHED method found that in the DPs the rod-like reflexes corresponding to the structure of silicon carbide, began to be added with two reflexes at both sides from the main reflex [13], which corresponds to the reconstruction of $(6\sqrt{3}\times 6\sqrt{3})$R30° carbon buffer layer (Figure 2, *f*) [13–15]. Therefore, to obtain atomically smooth surface without a change in the stoichiometric composition, it is necessary to either reduce the annealing time with the annealing temperature unchanged, or to reduce the annealing temperature with the annealing time unchanged.

4. Conclusion

For the first time the 6*H*-SiC(0001) step separation was conducted and visualized in the real time mode by *in situ* MEM method. It is shown that the combination of *in situ* MEM/RHED methods and *ex situ* AFM makes it possible to directly relate the kinetics of the surface rearrangement with the technological parameters of annealing. It was found that under UHV conditions the annealing at $T < 1160$ °C causes smoothening of the step edges, and activation of the step separation process with formation of macrosteps from three SiC bilayers with the height of 0.75 nm happens at 1160−1250 °C. Under further temperature increase the formation of macrosteps is accompanied by graphitization, which was established by the *in situ* RHED method. Therefore, in this paper for the first time it was possible to separate the diffusion and sublimation mechanisms of step separation.

It was found experimentally that the degree of terrace filling with SiC bilayers depends not only on temperature, but on time of annealing and sample geometry, at the same time the increase in the duration above 30 min at 1200 °C does not result in substantial improvement of morphology under UHV conditions. Therefore, the optimal mode providing for the formation of atomically smooth stoichiometric surface 6*H*-SiC(0001) consists in the combination of correctly selected temperature and time of annealing.

The obtained data may be used to develop standardized methos of pre-epitaxy treatment of silicon carbide substrates, which are critical for the subsequrnt growth of high quality epitaxial layers of graphene and other 2D materials.

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Conflict of interest

Authors declare no conflict of interest.

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