RHEED Oscillations during the MBE Process under the Coexistence of Step Flow and Two-Dimensional Nucleation Growth Modes

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The evolution of different types of surface relief: vicinal surface with equidistant steps of monoatomic height and echelons of steps; singular surface with steps of opposite directions, and the surface with previously formed islands of monoatomic height during MBE growth was investigated by Monte Carlo simulation. Parameters of the model were chosen for the simultaneous appearance of two-dimensional islands creation and step flow during the growth process. The coexistence of these two modes leads to a decrease in RHEED oscillations period, self-organization of specific asynchronous structure of islands, fast damping of RHEED oscillations on the vicinal surface, and to initial relief smoothening and beat-shape RHEED oscillations on the singular surface. Reasons of asynchronous structures formation and conditions for beats and other distortions in RHEED oscillations were investigated.

1. Introduction

Molecular beam epitaxy (MBE) is one of the basic methods for production of low-dimensional structures. RHEED intensity oscillations widely practiced as the method of growing surface control allow to determine thickness of the obtained film with an accuracy of one monolayer. However, oscillations damping with time and distortions of their shapes decrease the accuracy of thickness determination. Surface relief has a great impact on electronic properties of grown structures. But it is a cumbersome task to combine the MBE technology with the direct observation of the surface during the growth process. Complicated shape of oscillations contains information not only on the number of growing monolayers but on the initial microrelief, evolution of this relief through the surface processes, and the final morphology of the grown layer [1–4]. Relationship between the shape of RHEED oscillations and microrelief of growing film under the coexistence of the step flow and two-dimensional nucleation growth modes is demonstrated. Simulations of MBE growth processes on the vicinal surfaces with equidistant steps and echelons of steps as well as on the surfaces with initially formed two-dimensional islands at different effective surface temperatures were carried out. Surface step density oscillations of islands and steps in the simulation process were compared with RHEED oscillations obtained during MBE growth of Ge on Ge(111).

2. Simulation. Model

Simulation of the MBE growth process was carried out by the Monte Carlo method under the assumption of the solid on solid (SOS) deposition on the Kossel crystal (100) surface. Simulations were performed on the lattice with 160×160 atomic sites for cyclic boundary conditions [5–6]. The RHEED oscillations in real MBE technology were compared with the calculated values of the perimeter of the islands and steps, the so-called step density. Surface step density oscillations along with a computer film demonstrating the surface relief evolution during the growth process were obtained by simulation.

Step density dependence on time during the growth process was analyzed. The role of one or another mechanism of growth was determined by the ratio of such parameters as the migration length λ and terrace width W and was practically independent of bond energy of neighboring atoms for temperatures lower than the roughening temperature. Migration length was the measure of the effective surface temperature under constant intensity flux conditions. In our model migration length is defined by the number of diffusion steps per unit time. Elevation of the surface temperature is equivalent to a rise in diffusion steps number, i.e. an increase in diffusion length. The maximum migration length used in simulations corresponds to the pure step flow mode and the minimum one to the two-dimensional island growth mode. Three-dimensional growth mode was not considered in this work. The main features of surface relief evolution considered in this work did not depend on the presence of active adatom exchange between steps, islands, and lattice gas.

Fig.1 demonstrates the main processes that are taken into consideration in our model. Depletion region near steps is also indicated in this figure because of its essential influence on RHEED oscillations in passing from the step flow to the two-dimensional nucleation mode.

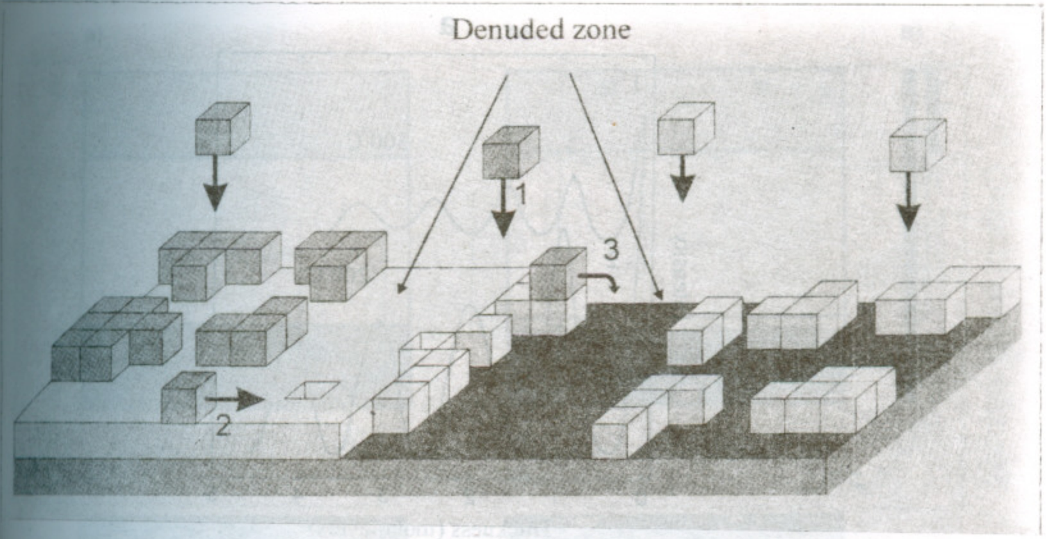

Figure 1. Schematic view of the modelling surface. The main processes considered during simulation: 1 – deposition, 2 – diffusion and 3 – incorporation of adatoms at the step edges. Denuded zones created near step edges are shown.

3. Influence of the epitaxial growth temperature on the period of RHEED oscillations

The RHEED oscillation dependence on the substrate temperature during the growth was detected for the first time in experiments for MBE of GaAs [7]. More recently a decrease in the oscillation period was shown experimentally for Ge epitaxy on the Ge(111) surface [8,9]. Monte Carlo simulations enabled to clarify the phenomenon of such reduction of the oscillation period with the increase in temperature. Intensity of RHEED oscillation during Ge growth on the vicinal surface at various temperatures could be seen in Fig.2a, whereas results of simulation for the same process are presented in Fig.2b. To find the relationship between the oscillation period and Ge thickness, the film thickness was measured independently by a quartz monitor during RHEED intensity registration. The oscillation period decreased with the temperature increase. Variations in the oscillation period are observed at a constant deposition rate, while the temperature changes in the range corresponding to transformation from the 2D growth to the step-flow mode. The oscillation period is close to the time of growth of one monolayer in the case of 2D nuclei growth mode. In this case two-dimensional nuclei are formed at a terrace completing the whole terrace during the deposition process, while the steps move only slightly after one

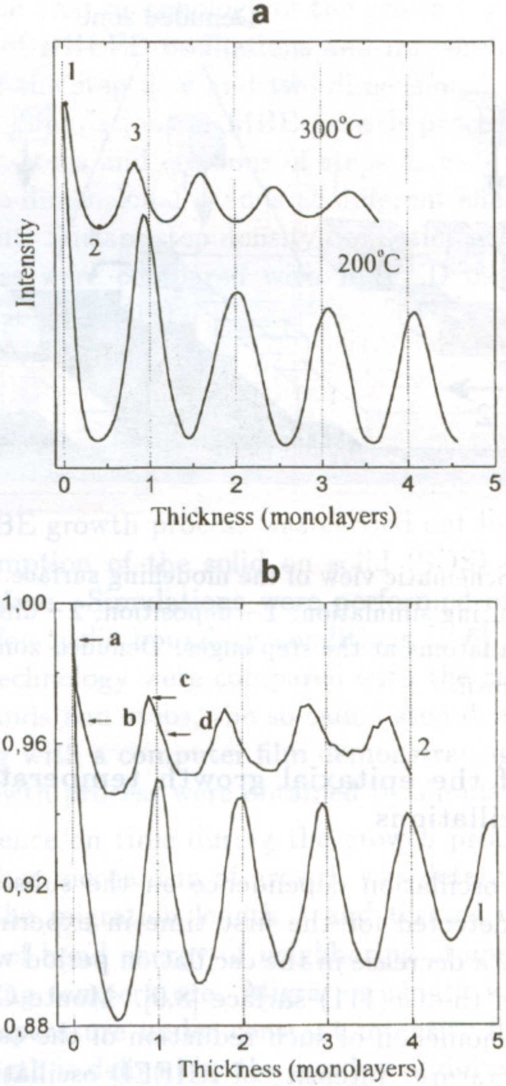

Figure 2. (a) RHEED oscillations during Ge growth at various temperatures. (b) Step density oscillations on a vicinal surface for two values of migration length: 1 – $\lambda = 7$ atomic units (a.u.), 2 – $\lambda = 15$ a.u.; terrace width $L = 80$ a.u.. Intensity of the deposition flux during the growth process is constant.

monolayer deposition. Dependence of the oscillation period on the substrate temperature is observed for temperatures when expansion of two-dimensional nuclei is associated with a noticeable step propagation. The maximal decrease in the oscillation period occurs under the conditions close to the transition from

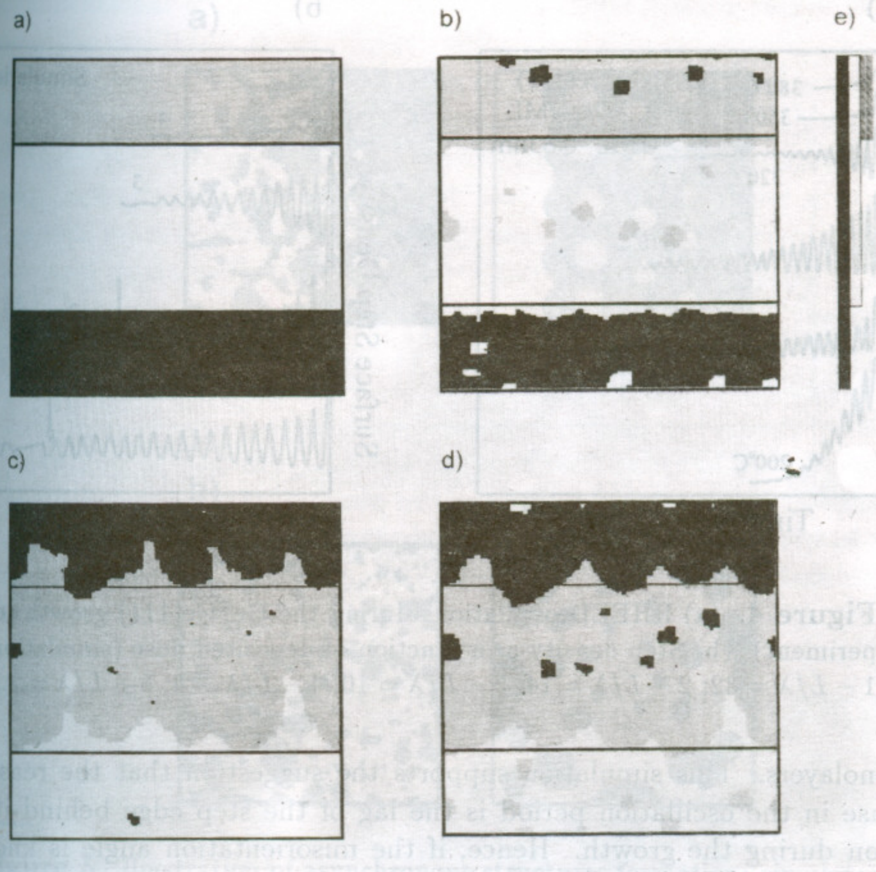

Figure 3. The simulated surfaces at the instants of time indicated by arrows in curve 2 in Fig.2b: a – initial surface; b – after 0.15 monolayer deposition; c – 0.9 monolayer deposition (first maximum); d – 1 monolayer deposition; e – profile of the initial surface relief.

the 2D to step-flow mode. Further increase in temperature leads to the total disappearance of the oscillations because of the absence of two-dimensional islands at the surface. Fig.3 represents the simulated surfaces corresponding to different deposition doses (indicated by arrows in Fig.2b). Depletion areas at the terraces nearby the steps, as well as the co-existence of 2D and step-flow mechanisms are seen in Fig.3b. Fig.3c corresponds to the maximum of the oscillations, this is seen to be the instant of time corresponding to the maximal smoothness of the terraces and steps. There are no islands of a new generation. The average step lag behind the initial position is close to the migration length. A further deposition leads to the nucleation of islands of a new generation; this is just the reason that decreases the step density after the deposition of an integer number

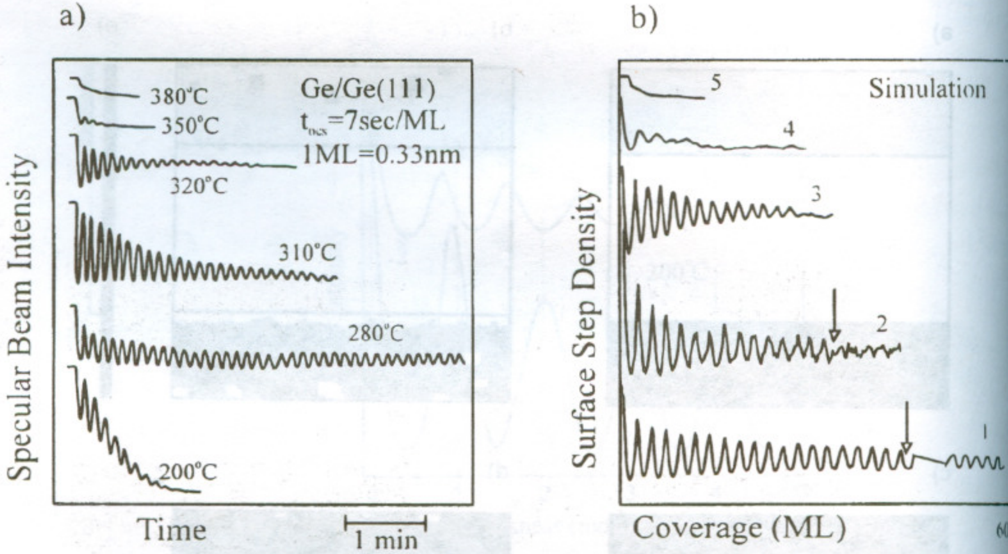

Figure 4. (a) RHEED oscillations during the Ge/Ge(111) growth (experiment): (b) Step density as a function of deposited dose (simulation): 1 - $L/\lambda = 32$; 2 - $L/\lambda = 14$; 3 - $L/\lambda = 10$; 4 - $L/\lambda = 4$; 5 - $L/\lambda = 2$.

of monolayers. This simulation supports the suggestion that the reason for decrease in the oscillation period is the lag of the step edge behind its initi position during the growth. Hence, if the misorientation angle is known, th magnitude of λ can be determined from the variation in the oscillation perio [10].

4. Influence of the epitaxial growth temperature on surface reli evolution

Although dependence of RHEED oscillations shape on the growth temper ture is well known [9,11–13], its interpretation is ambiguous. We try to obta information from these dependences on the initial morphology of the surface, evolution during growth process and the final microrelief of the grown layer. this section simulation results of MBE growth processes on the vicinal surfac with equidistant steps and echelons of steps are presented.

Fig.4a demonstrates RHEED oscillations obtained during homoepitaxy Ge on the surface with (111) orientation at various surface temperatures a the deposition rate equal to 2.5 nm/min. Step density as a function of deposi dose is represented in Fig.4b.

All results presented in Fig.4b were obtained by simulation on the stepp

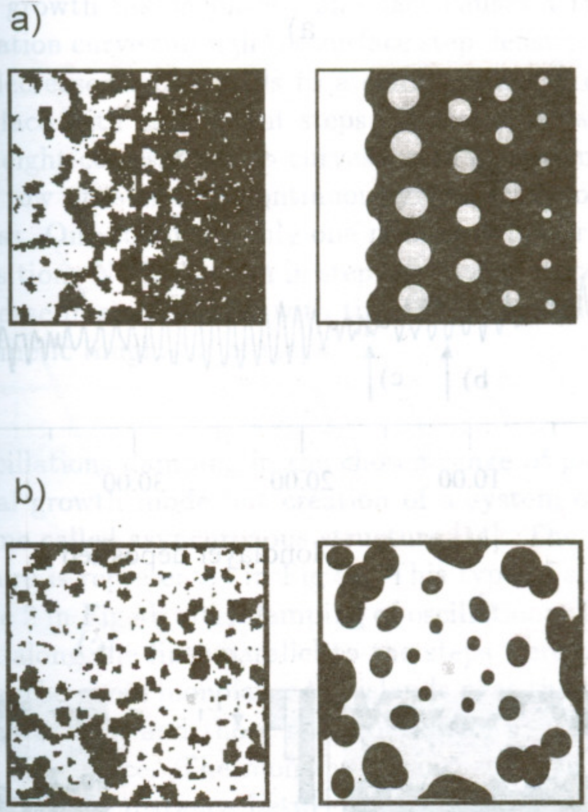

Figure 5. Illustration of asynchronous structures formation: simulated (left) and schematic (right) surfaces. (a) – linear asynchronous structure after 15 monolayers deposition ($L/\lambda = 14$) (upper terrace on the left); (b) – circular asynchronous structure after 19 monolayers deposition ($L/\lambda = 32$).

surfaces. Maximum terrace width L was 160 atomic units (a.u.). Terraces could be separated by monoatomic steps, or by steps echelons of four monolayers height. Oscillations damping shapes depended on the parameter L/λ. Three bottom curves in Fig.4b correspond to the echelon type initial relief. For curves 1 and 2 the parameter $L/\lambda \gg 1$, so there is no step flow and no decay of echelons. Increasing number of the simulated curve corresponds to reduction of L/λ. For the curve number 1 L/λ is so high that the oscillations behavior is similar to their behavior on the flat surface. For the curve 2 echelons are practically not decomposed during the growth process but one could observe a shift of the lower step simultaneously with creation behind it of the region depleted with islands. This causes rapid damping of oscillations through the

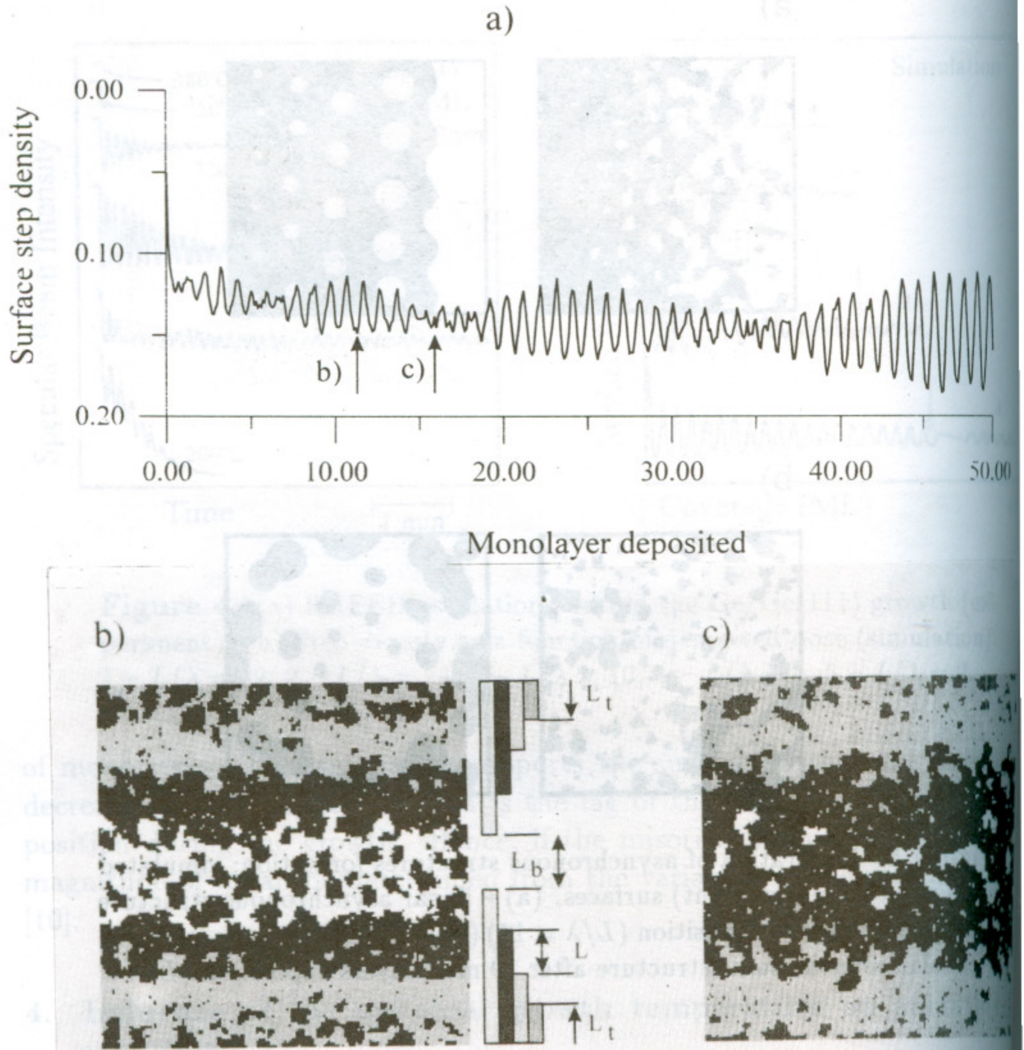

Figure 6. Simulation of the step density and surface morphology evolution during the deposition on the surface with the steps of opposite directions ($\lambda = 8$ a.u.): (a) – step density; (b), (c) – top view of the simulated surfaces for deposition doses indicated by arrows in Fig.8a; L_t – width of the top terrace, L_b – width of the bottom terrace, L – width of the intermediate terraces.

formation of a specific structure of islands with asynchronous nucleation [... Reduction of L/λ results in the echelons decay: appearance of narrow terra... free of islands with a simultaneous decrease in the initial terrace width ...

the two-dimensional growth taking place. This fact causes a rise of the lower envelope of the oscillation curve through the surface step density decrease (curve 3 Fig.4b). Further decrease in L/λ leads to a complete decay of echelons and formation of the surface with equidistant steps. Initial relief with equidistant steps of monolayer height corresponds to curves 4 and 5. For migration length λ of about $L/4$ one row of islands is continuously formed before moving step during growth process. One could see only one minimum in curve 4 during the first monolayer deposition. No oscillation in step density curve could be seen for $L/\lambda < 2$. Some decrease in step density with time appreciable in curve 5 could be associated with kinetic roughness of steps.

The reason of oscillations damping in the chosen range of parameters is not the three-dimensional growth mode but creation of a system of islands reproducing itself with time called asynchronous structure [14]. The view of a linear asynchronous structure is represented in Fig.5a. This type of surface relief corresponds to the curve 2 in Fig.4b after damping of oscillations. In this structure the islands arranged along the lines parallel to the steps were nucleated simultaneously; however in the given monolayer the islands near the top terrace were nucleated earlier than others, and those near the low one – later. Thus asynchronous nucleation took place. The moment of nucleation as well as the size of the island depend on the distance between the nucleus and the step. Linear asynchronous structure reproducing its form apparently moves during the deposition process. So its perimeter on the average remains constant causing damping of oscillations.

The top view of the surface corresponding to the curve 1 in Fig.4b is represented in Fig.5b. Nearly completed atomic layer contains vacancy-islands. The system of islands with increasing average radius as they move away from some center with vacancy-island could be seen in Fig.5b. Since the islands near the center with vacancy-island nucleated later than at the periphery due to the outflow of adatoms in vacancies, such a system of islands was called a "circular asynchronous structure". Circular asynchronous structure could be formed on the singular surfaces as well as on the vicinal ones with wide terraces without step flow. Oscillations damping in the initial part of the curve 1 in Fig.4b is accounted for by the formation stage of the circular asynchronous structure. Slightly damped oscillations observed at a later time are characteristic of the asynchronous structure of the circular form. Arrangement of such circular structures is maintained in every following monolayer.

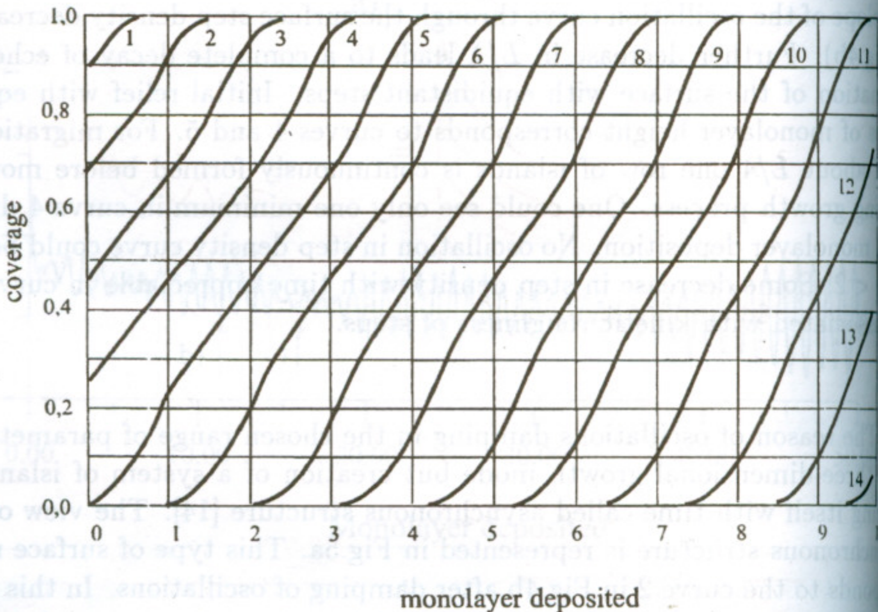

Figure 7. Coverage of individual atomic layers during the growth process. Figures in the curves indicate the number of the individual growing layer.

5. Nonmonotonous modulation of the oscillations amplitude on th surfaces with the steps of opposite directions

Quite different character of oscillation shapes one could observe during t growth process on the surfaces with the initial relief with the steps (or grou of steps) of opposite directions [15]. Fig.6a demonstrates the step density o cillations during the growth process for described initial morphology. Fig (right) represents such a profile of initial surface relief with two echelons o steps of opposite directions. The initial relief consist of five terraces and fo uncompleted atomic layers: L_t is the width of the top terrace, L_b is width of t bottom terrace (valley), and L is the width of the intermediate terraces. Chos relationship between the migration length and terraces width in this simulati leads to 2D island nucleation only on the top terrace at the initial phase of t growth process ($L_t > 2\lambda$). Only few islands are created on the intermedi terraces ($L \leq 2\lambda$), for this terraces growth process is determined by the st flow mode. Coverage kinetics of 14 atomic layers on the surface outlined abo after the deposition of 10 monolayer dose is illustrated in Fig.7. At the init stage of the deposition five atomic layers are growing on 5 terraces simulta

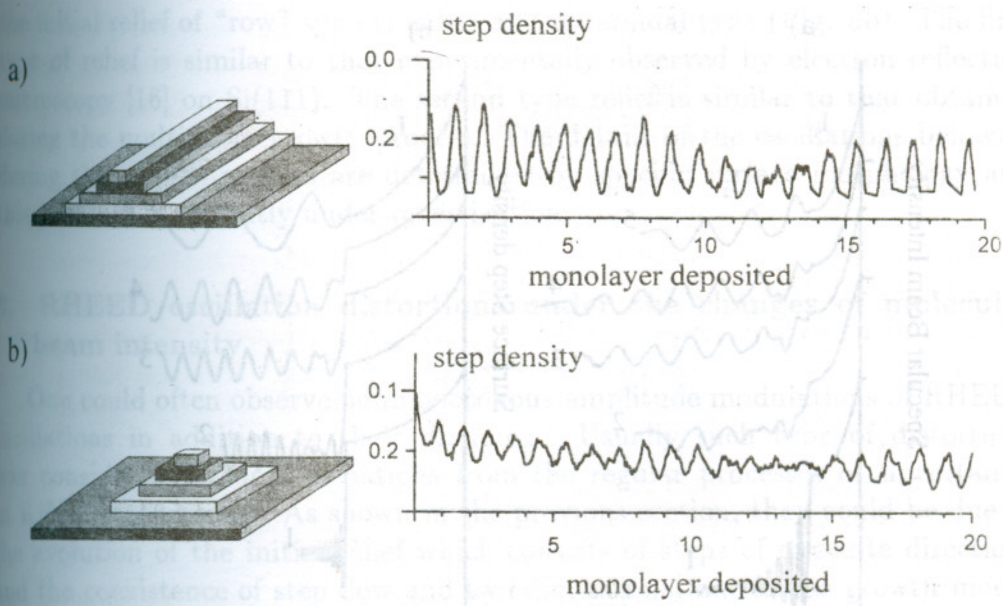

Figure 8. Step density during the growth process for two types of the initial surface morphology: a –"row"-type relief; b – pyramidal type relief.

ously: the first one corresponds to the bottom terrace and the fifth to the top one. Using the definition of the growth front as the number of simultaneously growing layers one could say that at the initial stage of the deposition process the growth front is equal to five. Analyzing the curves in Fig.7 one could notice one or two breaks of the curves slope and see five growing layers at the initial stage of the deposition process and only three growing layers at the final stage. These facts indicate the surface smoothing and reduction of the growth front and demonstrate the growth rate dependence of the layer on its position among other uncompleted layers.

Analysis of the simulated surface shows that the growth rate of the layer on the top is lower than the rate in the valley through the coexistence of two-dimensional and step-flow modes of growth. The faster growth rate in the valley is due to the fact that two processes are responsible for the growth in this region: the atom flux impinging on the surface and "downslope" flow from the upper steps followed by incorporation of these additional atoms into the step edges. There is only one growth mode for uppermost layer — the islands growth mode, and at the same time this layer is the source of atoms for lower steps. That is the reason for reduction of the growth rate of the top layer.

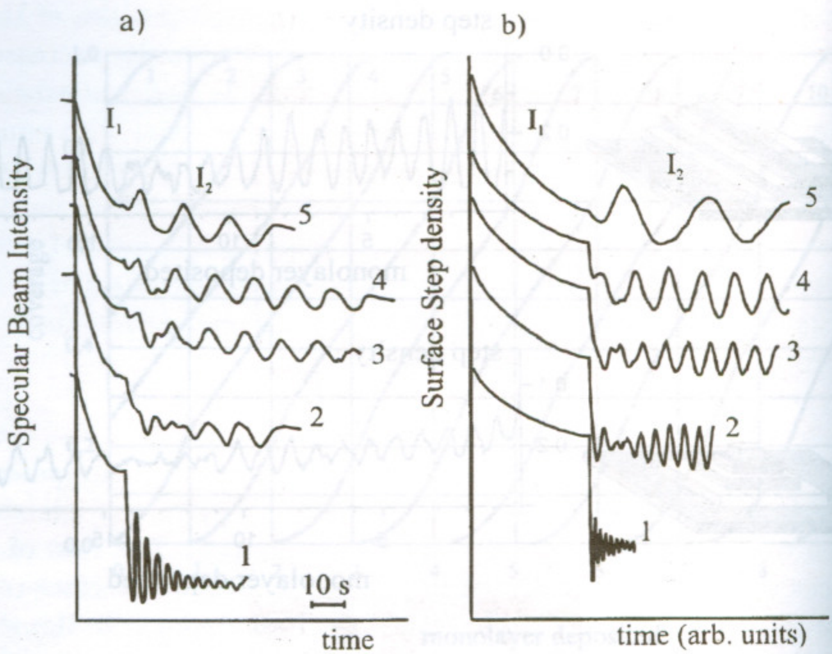

Figure 9. RHEED oscillations during homoepitaxy of Ge on the vicinal surface under changing flux intensity after 0.5 ML deposition. I_2/I_1 for curves 1-5: 1 – 16; 2 – 10.9; 3 – 7.7; 4 – 5.6; 5 – 4.2 (I_1 – initial, I_2 – final flux). (a) – experiment: Ge/Ge(111), T = 300°C; (b) – simulation.

For the types of surfaces shown in Fig.6b the step density oscillations determined only by the lowest and top terraces, as on the intermediate terra independently of whether islands or asynchronous structures with permane constant step density are created. So there are only two regions with d growth rates that are responsible for step density oscillations. Addition of the two rates leads to the beat-shaped oscillations.

The simulated surfaces after deposition of different numbers of monola (indicated in Fig.6.a by arrows) are presented in Figs.6(b-c). Different cover rates for the lowest and top terraces causes its "in phase" filling correspondi to the maximum oscillation amplitude (moment b) and its antiphase filling w the minimum amplitude (moment c). Watching surface relief evolution one co see that after passing each beat the growth front decreases by one.

We have observed the onset of amplitude modulation of the beating typ all cases when the initial surface is defined as the terraces enclosed by the steps opposite directions and two growth modes coexist: step-flow and island grow Fig.8 presents the simulation results for epitaxial growth on the surfaces w the initial relief of "row" type (Fig.8a) and pyramidal type (Fig. 8b). The first type of relief is similar to that experimentally observed by electron reflection microscopy [16] on Si(111). The second type relief is similar to that obtained during the multi-layer growth process. The details of the oscillations behavior during the growth process are determined by specific surface morphology and this problem is currently under investigation.

6. RHEED oscillation distortions under the changes of molecular beam intensity

One could often observe nonmonotonous amplitude modulations of RHEED oscillations in addition to their damping. Usually such type of distortions was considered as some deviations from the regular processes or uncertainty in RHEED conditions. As shown in the previous section, they could be due to the evolution of the initial relief which consists of steps of opposite directions and the coexistence of step flow and two-dimensional nucleation growth modes during the growth process. In this section we analyze the shapes of RHEED oscillations for the growth process with changing flux intensity.

Simulation of the MBE growth under the changes of molecular beam intensity during the deposition process demonstrates nonmonotonous amplitude distortions of the step density oscillations mentioned above. This type of simulations was performed to explain available experimental RHEED oscillations obtained during the Ge/Ge(111) deposition using two sources of different intensities. The experiments were carried out with a home-made MBE system described in [9]. Equivalent to the variation of the flux intensity in the experiments is the migration length variation according to the ratio $\lambda \sim (D/I)^{1/4}$ [14] in simulations.

RHEED oscillations and step density oscillations under changing flux intensity during growth process are represented in Fig.9. (I_1 – initial flux, I_2 – final flux, $I_1 < I_2$). Changing I_1/I_2 ratio one could obtain a set of simulated curves with different shapes of step density oscillation similar to experimental ones. After half monolayer deposition in low intensity flux large islands are formed on the terraces of simulated surface along with the creation of depletion regions free of islands behind moving steps. Further deposition in higher flux results in simultaneous islands nucleation on the flat parts of the terraces and on the large islands grown earlier. Presented in Fig.10 are the simulated vicinal surfaces under changing flux intensity during the growth process after the deposition of 0.5 monolayer in the flux I_1 (Fig.10a) and after additional deposition of 0.20 ML in the greater fluxes I_2 (Fig.10.b).

Oscillation frequency of the step density for these two systems of islands

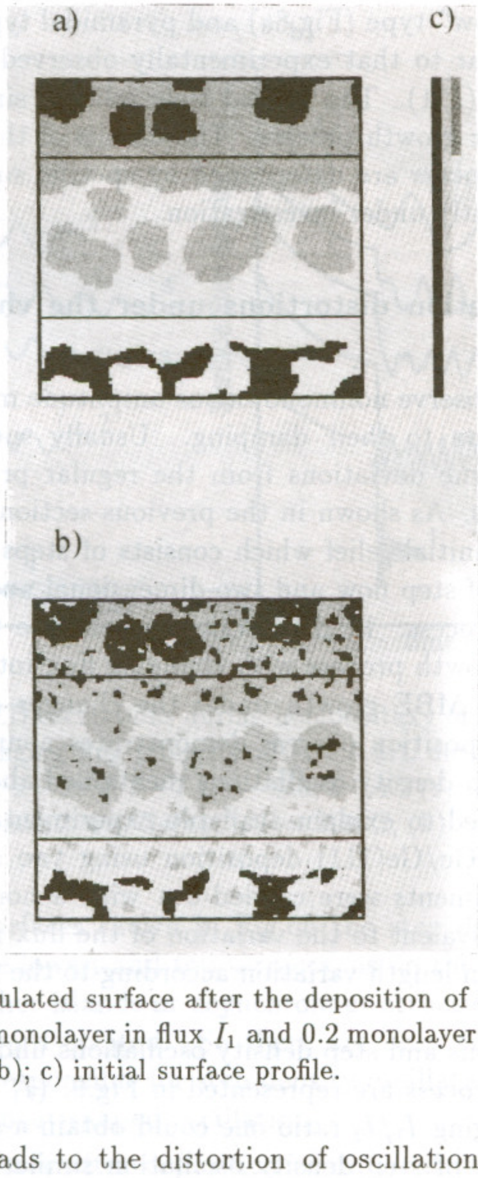

Figure 10. Simulated surface after the deposition of a) 0.5 monolayer in flux I_1; b) 0.5 monolayer in flux I_1 and 0.2 monolayer in flux $I_2 = 16I_1$ (for curve 1 Fig.6b); c) initial surface profile.

are different which leads to the distortion of oscillations until the flux w intensity I_2 determines the growth process. Analysis of the simulation resu indicates a simultaneous growth of two layers in the higher flux with subseque smoothening of this surface relief. The oscillations shape is actually sensiti to the relationship between the terrace width and migration length (or fl intensity), i.e. the relation between 2D and step flow modes.

7. Conclusion

Peculiarities of intensity oscillations damping at various temperatures a the evidence of a rather complicated mechanism of this process. Simulati of the MBE process shows that the shape of RHEED oscillations is directly determined by the features of the surface morphology. Initial surface relief as well as its transformation during the growth process influence the oscillations shape. In some cases this allows just in the MBE process to draw conclusions not only about the thickness of the layer but about its surface relief as well from the shapes of oscillations.

It is shown that the presence of the steps on the surface of the growing crystal introduces an additional contribution to the oscillation damping when the migration length becomes comparable with the terrace width. Damping process is connected not only with the formation of a new layer on the uncompleted previous one but with the desynchronization of the islands nucleation as well. The number of oscillations observed in this case is approximately equal to the ratio of terrace width to migration length of adatoms. An obligatory condition of self-organization of linear asynchronous structures on the vicinal surfaces is the coexistence of two modes: step flow and two-dimensional nucleation growth.

The Monte Carlo simulation was used to predict a decrease in the RHEED oscillation period with the increase in temperature. This phenomenon is observed at the range of transition from the 2D growth mode to the step flow one. The influence of growth temperature on the oscillation period should be taken into account when the registration of oscillations is used to control the growth parameters and to study the processes occurring on the surface of growth.

Simulation of MBE process with a complex initial relief shows: first – smoothening of the initial relief (that may be surface refaceting) as well as reduction of the growth front take place during the MBE process; second – the smoothening process could be a cause of the distortion of RHEED oscillations, such as nonmonotonous amplitude modulation. So not only the incident flux and temperature non-uniformity or the diffraction condition changes during the growth process could distort oscillations but these distortions could reflect peculiarities of the initial surface morphology.

Acknowledgements

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