Emergence of regular meandered step structure in simulated growth of GaN(0001) surface

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Abstract

Step meandering during the growth of gallium nitride crystal is studied on using kinetic Monte Carlo method. Cause of instability is identified to be the particle advection caused by the step flow. Growth process is conducted in N-rich conditions and GaN(0001) surface kinetics is modeled by setting jump probabilities for Ga atoms adsorbed at the surface. We show that at low enough temperatures and relatively high external particle fluxes periodic regular pattern of meanders is created with its wavelength inversely proportional to the particle flux. An increase of the meander amplitude saturates after some period and further crystal grow is stationary, creating "finger-like" structure. For medium fluxes regular structure of meanders builds up for low or zero value of Schwobele barrier. For higher fluxes wavelengths of meanders become shorter than the terrace width and they start to grow independently and finally transfer the surface to a rough structure. For very low fluxes or at relatively high temperatures steps move steadily remaining their initial shapes of straight, parallel lines.

1. Introduction

Crystal growth dynamics and its relation to the formation of various geometric patterns remains a subject of continuous interests of many researchers. Simple picture of straight parallel steps flow during growth process was enriched by additional phenomena either observed in the real growth experiments or emerging during Monte Carlo simulations. It was noticed that parallel steps have tendency to create step trains, in accordance to the prediction of kinematic step train theory, proposed first by Frank [1] and later discussed by Vekilov et al. [2]. This tendency could lead to a creation of train of steps, double steps, macrosteps of even the formation of new crystallographic face during growth [3–7]. Steps can form bunches, but also bend creating characteristic meanders. Instability phenomena, denoted as step meandering, were observed during the growth of different crystal types [8–15]. The experimental data include step meandering in various systems, both metallic and semiconductor. Step meandering was observed during the growth on vicinal surfaces, such as Cu(1 1 1 7) or Cu(0 2 2 4) [9]. Similarly, step meandering was identified during growth on densely packed Si(111) surface [10–13]. Such phenomenon was discerned on vicinal Si(100) surface [16].

It was also shown that electromigration of adsorbed atoms considerably enhances the strength of the process [17]. Similar meandered structures have been lately observed at GaN(0001)surface [14,15]. In addition, in Monte Carlo simulations such behavior was observed [18]. Step meandering was recognized as important phenomenon which enhanced activity in this domain. Several analytical approximate approaches were proposed, including Kardar, Parisi, Zhang (KPZ) equation, describing kinetic roughening of the step during the growth of epitaxial layers [19]. The KPZ equation, derived using symmetry arguments, is frequently used to analyze scaling properties of the Eden model describing the growth of epitaxial layers in lower temperature Molecular Beam Epitaxy (MBE) processes. Another approach, based on more physical arguments, was undertaken by Bales and Zangwill who considered linear stability analysis of single step with respect to meandering [20]. They showed that the instability is related to different incorporation rates from the lower and upper terraces and is diffusional in nature. Their analysis was extended to the nonlinear case by Bena et al. who derived equation governing the temporal evolution of the system [21]. The equation was in fact published earlier by several authors [22–24], and is referred as Kuramoto–Shivashinsky equation [25]. Bena et al. showed that the instability is followed by chaotic behavior of the system [21].

An addition of elastic interaction to the equations changes the character of the instability development [26,27]. Below we report the results of MC simulations of the GaN(0001) surface model with isotropic diffusion constant and uniform flux of Ga atoms. Low or
none Schwoebel barrier is assumed in the basic version of the model. We study the emergence of regular meandered structure of steps at the misoriented surface.

We use recently developed lattice gas model, which was able to recover basic features of the growth of GaN(0001) layers by metalaorganic vapor phase epitaxy (MOVPE) [28, 29]. It was shown that the characteristic anisotropy of step dynamics can be modeled by using four-body interaction. It was also shown that in addition to step pairing of [10T0] steps, step meandering can be obtained for large system sizes, which is observed in real crystal growth experiments [14,15]. As it was lately discussed in Ref. [30] the study of GaN(1T00) surface stability of step flow strongly depends on the surface orientation, the miscut orientation and the temperature. In this work we study GaN(0001) surface with steps along [1T210] direction. Every step of this orientation has the same adsorption energies and is mirror image of the neighboring steps. We study meandering in the systems of equivalent steps [1T210] with small or zero Schwoebel barrier. Linear stability analysis approach does not predict such behavior. Such analysis usually does not take into account step movement. In fact step movement is a source of particle advection, i.e. particle flow towards moving steps. Our results suggest that the step movement can be a source of step meandering. Meandering often evolves into a regular finger-like structures at the surface. Such regular structures are quite often observed GaN surface morphologies [14,15]. Similar structures are also created at Si(111) surface [17]. Steps can both meander and bunch at the same time [13,31]. In general meandering instability can lead to the further bunching phenomenon [31]. In simulations of our GaN(0001) model, we have not found any bunching. Presumably some additional model parameters are necessary to reproduce bunching step instability during crystal growth. However, meandering instability is evident in studied system and leads to the stable meandered structures similar to that which materialize at the surface.

In the next section we shortly describe model. Section 3 is devoted to the presentation of the simulation results of the step meandering process which builds regular, “finger-like” structure. They are compared with meandering for higher particle fluxes what ends up as a rough surface. Section 4 contains analysis of the observed meander evolution.

2. The model

We simulate growth of GaN(0001) layer in the N rich conditions. The model we use here has been developed in [34], and used to study the dynamics of steps in [10T0] direction. Here we concentrate on general meandering phenomenon and study the evolution of steps in [1T210] direction. Crystal growth is controlled by Ga atoms accumulated and diffused on the top of growing crystal. We model the wurtzite lattice of the GaN crystal out of tetrahedrons of Ga atoms centered on N atom as illustrated in Fig. 1. Three bonds connecting Ga atom with the nearest N atoms within one layer are rotated by 60° with respect to the bonds in the consecutive Ga layer. We have shown in the previous work [29] that the simplest way to account for the layer difference is to introduce four-body interaction between Ga atoms. Accordingly, our system is described by modeling the energy and dynamics of Ga atoms only. We assume that the energy affecting jump probability of each Ga from the lattice site depends on the number of Ga neighbors and on the N position as a tetrahedron center. It is determined in the following way:

\[
\eta_i = \begin{cases} 
1 & \text{when tetrahedron has all atoms;} \\
\frac{1}{4} \eta & \text{when tetrahedron has empty sites}, 
\end{cases}
\]

where \(\eta\) is a number of occupied neighboring sites, belonging to a selected tetrahedron and \(r\) describes the relative strength of the four-body and the two-body interactions in the system. When \(r = 1\) two body Ga–Ga interactions sum up to the value characteristic for fully occupied tetrahedron, i.e. no additional four-body Ga interactions are present in the system. When \(r < 1\) three pair bonds to the nearest neighbors of a given particle in tetrahedron do not sum up to a value of one multiparticle bond. In such case tetrahedron energy is not a simple sum of two-body interactions. This is the case we study below, the value \(r = 0.4\) is used throughout the remainder of this work. It should be noted that the main results recounting step meandering process do not depend on the parameter \(r\) significantly.

At GaN(0001) surface, each Ga surface atom belongs potentially to four tetrahedrons, three in the present layer and the one above. Its total energy could be expressed as

\[
\alpha(j) = \sum_{i=1}^{4} n_i,
\]

where the parameter \(J\) scales bonding energy and the sum runs over four tetrahedrons surrounding every atom.

It is assumed that Ga atoms are adsorbed at the surface uniformly. Thus, the Ga adsorption is accounted for by a creation probability of an adatom at any empty adsorption site, at each MC step. This probability is given by the expression defining the external particle flux

\[
F = v_2 e^{-\beta \mu},
\]

where \(\mu\) is a chemical potential, \(v_2 = 2\) sets the timescale of simulation and \(\beta = 1/k_B T\) with \(T\) as the temperature and \(k_B\) Boltzmann constant. Each adsorbed particle diffuses over the terrace until it is attached at the steps. Thus, the possibility of reevaporation is neglected. Probability of a jump from the initial to the final site, in the diffusional movement, is given by diffusion parameter \(D\), expressed as

\[
D = D_0 e^{-\beta \mu},
\]

where \(D_0 = 1\) is diffusion timescale, and

\[
E = E_B - \alpha(j)
\]

is the difference between the transition state energy \(E_B\) and initial state bonding energy \(\alpha(j)\). Note that the uniform increase of the energy barrier for all jumps by the same value amounts to mere rescaling of the timescale. We construct barrier for the diffusion in such a way that it is lower for jumps along step than for jumps over terrace. The simplest way for the construction of such a barrier is choosing \(E_B = \min(\alpha(j), \alpha_{i}(j))\) where \(\alpha_{i}\) is bonding energy of the final state of diffusing atom. As a result we get the...
which ensures lower energy barrier for jumps to the step and along step. We checked also system dynamics with additional barrier, which increased the difference between diffusion along step and over the terrace, but final structure of meanders did not change much. Adsorption rate at the step is for some of the step and over the terrace, but final structure of meanders did not along step. We checked also system dynamics with additional which ensures lower energy barrier for jumps to the step and

\[ D_B = e^{-\beta B} D. \]  

Thus, the height of the barrier \( B \) modifies jump rate given by Eq. (4). As typically assumed, we impose Schwoebel barrier for the jumps from upper terrace. One barrier height \( B \) is used for each jump which crosses step.

Crystal surface microstate is modeled by setting two uppermost layers of atoms. Every second layer of Ga atoms has different bond orientation. In all simulations the surface is misoriented along one direction. When step moves forward the upper layer is converted into the lower one, and a new layer is built on top of the terrace. In such a way a continuity of particle–particle interaction at the step is guaranteed. In all systems we study below steps were initially oriented along [1\( \bar{7} \)10]. According to (1) at steps of this orientation the same number of particles attaches step boundary strongly and weekly. As seen in Fig. 1 one step has weakly bonding sites at the left kink side and strongly bonding sites at its right side and for the next step the situation is inverted. For the same density of adatoms, the step velocity depends on the particle interaction and therefore steps of [1\( \bar{7} \)10] orientation move with the same rate. Our simulations start with an even number \( n \) of equally spaced by \( d \) lattice constants, straight steps. Heights of the neighboring steps differ by one Ga atomic layer. Periodic boundary conditions are applied in the lateral direction and in the direction in which the crystal grows they are helical, i.e. they are corrected by constant height difference between both ends of the system.

The simulation schema outline is as follows: new Ga atoms materialize at any of the lattice site with probability (3). If new adatom appears it diffuses by jumps to the nearest neighboring sites with interaction dependent speed in accordance to (4) and then the diffusive step attachment jump is modified by Schwoebel barrier (7). In such a way crystal growth is realized. Apart from the single particle events described above no other actions are realized during the simulation.

3. Creation of meandered structure

On studying surface evolution for various temperatures, fluxes and crystal miscuts we found set of parameters for which system builds up regular meander structure. This structure results in characteristic, stable during growth pattern of finger-like lines oriented perpendicularly to the initial step orientation. We obtain such regular structures from the simulations of microscopic model. To obtain this result we do not need any additional contribution from the Schwoebel barrier asymmetry. Steps destabilize from the initial straight forms. There seem to be no other factor inducing such behavior than the existence of net flow of diffusing particle relative to moving steps. Such flow can be understood as particle advection induced by the step motion. The dependence of the step meandering on Schwoebel barrier is illustrated in Fig. 2. Top and middle rows of this figure compare step meandering with Schwoebel barrier absent in the model with the situation of relatively low barrier equal to the half of the interaction strength. In both cases the same external flux was applied. It can be seen, that in the case of zero Schwoebel barrier we have longer meanders and it takes twice as time to build them. The step movement is neglected in most of analytical approaches [20,21,25], whereas as was shown in Ref. [33] step advection in fact cannot be ignored. This agrees with our observations. Up to some point step-flow advection during crystal growth affects the system in similar way as usually Schwoebel
barrier does [25]. Such correspondence, however, stops working when Schwoebel barrier is too high. As we can see in Fig. 2, in the bottom row of pictures when Schwoebel barrier is too high, particles stick together breaking step continuity and disordered surface builds up. We conclude that the regular step structures can be build at the surface only when steps have possibility of exchanging particles. Regular stable in time meandered step structures build up only for low fluxes. As we see below for higher fluxes step meandering process has completely different character. Described above main features of meandering stay the same for \( r = 0.4 \) and \( r = 1 \). The only difference is in the relative width of the even and odd terraces at the left and right sides of meanders.

During the process of step meandering steps bend into the wavelike pattern, with the wavelength shorter for lower surface temperature and for smaller terrace width. Meanders which are long in comparison with the mean distance between steps create regular pattern of wavy like structure. Amplitude of these waves increases slowly during growth. Meanders of smaller wavelengths evolve in different way, becoming more and more irregular and finally form rough surface structure. We can compare an example of regular pattern obtained for lower external flux, plotted in Fig. 3 and the rough surface, obtained for higher flux, shown in Fig. 4. The step height correlation function

\[
f(x_1 - x_2) = \langle [h(x_1) - h(x_2)]^2 \rangle, \tag{8}\]

calculated for distances measured along vertical, parallel to step direction, where \( h(x) \) is a step height at point \( x \). Average \( \langle \rangle \) is taken over all system. It can be seen that the correlation function looks differently for these two surface patterns. When structure is well ordered, height correlation (8) oscillates from zero to zero value. Such structure is shown in Fig. 3 for surface patterns after two different numbers of simulation steps. In both cases the height difference falls down to zero, repeating such behavior regularly at the distance equal to the meander wavelength. This does not occur for the case shown in Fig. 4. The correlation function calculated for the system, not so well ordered, is small, with minute amplitude of its oscillations. Such structure of \( f \) shows that some characteristic length exists in the system only locally, as can be seen from the top panel of Fig. 4, but it vanishes at larger distances. For longer times such structure is concealed by the global step disorientation as shown in the middle panel of Fig. 4.

The characteristic wavelength of the step meandering can be measured as the position of the first minimum of function (8). The length defined in such a way can be found in the regular, ordered system, and also in the not so well ordered one, as these in Figs. 3 and 4. Starting from the initial configuration of straight steps, the characteristic length of the step meanders grows rapidly up to its final value, for all studied cases. The wavelength increase follows an universal function of the grown layer number \( n \) and it can be written as \( \lambda \sim n^{0.2} \). Such exponent is characteristic for early stages of pattern formation of the driven system [34]. It describes time growth of the characteristic length of domains in the direction perpendicular to drive. In our situation the system is driven by step movement and \( \lambda \) is measured along direction perpendicular to the velocity. This exponent is also in between exponent 1/6 and 1/4, which characterize terrace and line diffusion dominated crystal growth [27].

In our simulations \( \lambda \) for each system reaches plateau, stabilizing at one chosen value, which builds characteristic finger-like pattern. The plateau is reached at different values of \( n \). The final wavelength is proportional to characteristic power 0.5 of the flux. The lower flux and the shorter inter-step distances, the meander wavelength is longer, thus, we get \( \lambda \sim (F \sigma)^{-0.5} \). This dependence is illustrated in Fig. 5. This, quite general relation, comes out from various types of calculations [25] and appears to change when the
diffusion along the steps is seriously distorted [35]. In our case jumps are thermally activated with barrier given by site bonding energy calculated as inter-particle interaction. As long as the particle is attached to the step it wanders along it quickly and unhindered. It is stopped for some time at kinks, where its energy is lower. Time needed for particle to jump out of a kink changes with the temperature and that is why meanders have shorter wavelengths at lower temperatures.

Generally, the wavelength of meanders is of the order of a distance passed by a particle along the step during the time separating two subsequent particles attaching to the same site at the step. Assuming that particle realizes free random walk along the step and account that mean time between particles striking the step. Assuming that particle realizes free random walk along the step during the time evolution steps, initially ideally smooth, becomes rough. This behavior for wider terraces. We have shown that there is a range of parameters, where steps build regular, meandered structure. Below, we will analyze meander width evolution during crystal growth process.

4. Time dependence of meander amplitude

We measure the development of the meander by determining the amplitude of step meander waves. The amplitude is calculated as a difference between the most advanced and the most retarded points of given step, measured along direction perpendicular to the step initial orientation. We average the amplitude value over all steps in the system and plot it as a function of the simulation time. For all studied cases the time evolution of step meander amplitude is very similar. In Fig. 6, we show data for terraces twice or changes smoothly with the flux. In Fig. 5 we show data for one temperature and most of them were obtained for single terrace width $d=10$. We show, however, that the points for terraces twice wider or narrower follow the same line. Transition from one line to another depends on the ratio $\lambda/d$ and happens when it is close to $2\pi$, hence for each surface miscut it occurs for different $Fd$ values. The same wavelength that results in smooth meandering for small inter-step distance becomes too short for longer inter-step distance which leads to the rough surface pattern. Hence both points plotted for $d=5$ lay at upper curve and two different points for $d=20$ are located at two different lines.

Most examples we analyze here are for much narrower terraces than those observed experimentally, however, proper rescaling of the flux values is sufficient to obtain the same behavior for wider terraces. We have shown that there is a range of parameters, where steps build regular, meandered structure.

Fig. 6. Time evolution of the mean width of step meanders for three different surface miscuts. All data are plotted for systems at temperature given by $\beta J=5.5$, and for several terrace widths: for $d=5$—full black circles and for $d=10$—purple diamonds. All data are located along the lines, fulfilling two different $\lambda \sim (Fd)^{-0.5}$ relations. The lower line describes rough systems and the upper one describes the systems which form regular patterns. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The upper line is plotted for the smooth meandering process, obtained for lower fluxes. These points also follow the $\lambda \sim (Fd)^{-0.5}$ dependence. For these points, however, there is much more dispersion. They all are obtained for meanders of the length comparable or longer than the system size, hence boundary conditions become important. For wavelengths close to the system size one cannot be sure if this wave should not be slightly shorter or longer. Therefore it is very difficult to determine whether the increase of the meander length breaks down rapidly or changes smoothly with the flux. In Fig. 5 we show data for one temperature and most of them were obtained for single terrace width $d=10$. We show, however, that the points for terraces twice wider or narrower follow the same line. Transition from one line to another depends on the ratio $\lambda/d$ and happens when it is close to $2\pi$, hence for each surface miscut it occurs for different $Fd$ values. The same wavelength that results in smooth meandering for small inter-step distance becomes too short for longer inter-step distance which leads to the rough surface pattern. Hence both points plotted for $d=5$ lay at upper curve and two different points for $d=20$ are located at two different lines.

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Fig. 5. Wavelength of the step meanders as a function of average particle flux arriving at single site of the step $Fd$, i.e. particle flux impinging single lattice site at the terrace from the vapor, multiplied by the terrace width. Data are shown for $\beta J=5.5$ and the following terrace widths: for $d=10$ are plotted using open circles, for $d=20$—full black circles and for $d=5$—purple diamonds. All data are located along the lines, fulfilling two different $\lambda \sim (Fd)^{-0.5}$ relations. The lower line describes rough systems and the upper one describes the systems which form regular patterns. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
evolution period and the third is stationary growth of meandered structures. The amplitude value at which meandering saturates depends mainly on the width of terraces $d$ being relatively weekly dependent on the other parameters. Saturation of step meandering is an entirely new phenomenon. It is observed only for regular meander patterns, having wavelengths longer than the terrace width $d$. Such behavior has not been predicted by the equations published in the literature, describing particle flux balance [25,27], nor seen in previously simulated systems [37].

Evolution of systems simulated in Ref. [37] is shown for much shorter times and higher fluxes than in our case, so they could rather classify as systems of shorter wavelengths. More important difference, however, is that the system there was simulated for infinite Schwoebel barrier, but overhangs and voids were excluded in the step configuration, what causes that the simulated systems behave in different way.

Described here step pattern formation happens during crystal growth and several different processes play their role. A constant flux of particles $F$ arrives from above, subsequently distributed over the surface by the diffusion jumps. The particles attach to the steps, causing their movement forward and some of them can detach from the steps, increasing particle density at the terrace. Very important element of the pattern formation is the existence of the diffusion along and across steps, usually faster and slower than the diffusion over the surface. The particle exchange between different terraces can be blocked by high Schwoebel barrier replacing regular meander structure by rough surface of irregular step forms (see Fig. 2). It is evident that the particle flow across steps is a very important component of the pattern formation. Comparing steps obtained for low and for high Schwoebel barrier, shown in Fig. 2, we conclude that the meanders build up and order in regular structure all over whole system for communicating terraces, while the lack of inter-terrace communication destroys meanders and the system becomes rough (chaotic). It is evident that the communication between steps is crucial factor for the analysis of the step dynamics.

Jumps of individual particles depend on the local surrounding at the surface, summing up and averaging into various fluxes, which are described and discussed on using different approaches [18–21]. The phenomena, which should be considered, depend on the analyzed phase of the growth. Below we describe the scenario, according to which the regular meanders are formed and they grow less or more quickly, depending on the difference between fluxes incoming to the step and the step bent downward. Eq. (11) evidently sets the limit of the communication amplitudes of meanders in the simulated systems when constant $\lambda$ is 3 times larger. Step communications one with another by the exchange of particles. As we have discussed previously, their communication appears to be very important for regular meander pattern formation. It is evidently very important mechanism of the particle transport and it has to be proportional to the step length. Step length grows with meander amplitude $W$ as $\sqrt{1+(2W/\lambda)^2}$ multiplied by a constant, dependent on the meander shape. Finally the following expression describes the outgoing particle stream:

$$j_+ \sim F d \frac{d^2 z}{dx^2} \sqrt{1 + \left(\frac{2W}{\lambda}\right)^2}. \quad (10)$$

In addition to these two, specified above, main factors affecting the step dynamics, there are many others [25], which we did not include here. For example when one part of the step grows faster than the other, this causes that particle density becomes locally lower, whereas in the surrounding regions, where step moves slowly, a net particle density is increased by all those, which are not built into the step. When particle density on the top of one terrace surface region is higher than on the other, that difference in particle density induces diffusion flux towards less dense regions which reduces particle stream (10) a little. Such effect can be account for by the correction of the constant in front of this part of stream. In conclusion it seems that in the case of regular and highly curved meander pattern these two expressions above describe qualitatively all main aspects of step kinetics.

When we add both terms, the resulting equation is as follows:

$$dW \frac{dt}{dx} = \alpha F d L \frac{W}{\lambda} - \kappa c W \frac{d^2 W}{dx^2} \frac{1}{\lambda^2} \sqrt{1 + \left(\frac{2W}{\lambda}\right)^2} \quad (11)$$

with the parameters $\alpha$ and $\kappa$ being generally constants, depending on several mechanisms of particle flow between the step bent upward and step bent downward. Eq. (11) evidently sets the limit value of the meander amplitude $W_0$, for which the step becomes stationary. It is given by

$$W_0 = \frac{\sqrt{2}}{Z} \frac{\sqrt{\frac{E_0^2 d^4 a^2}{4 D_c k^2} - 1}}{Z} \quad (12)$$

and is dependent on one constant $Z/\kappa$. Relation (12) fits saturation amplitudes of meanders in the simulated systems when constant $Z/\kappa = 3 \times 10^5$.

Eq. (11) can also be easily solved, describing the time evolution of the step meander amplitude, which has characteristic functional shape and which, for larger $W$ value is given by the hyperbolic tangent. In Fig. 6 we have plotted time dependence of the meander amplitude $W$ for three different meander evolution examples. We also fitted hyperbolic tangent shape to each evolution obtaining very good agreement, independently of the terrace width $d$ or the meander length $\lambda$.

5. Conclusions

Kinetic MC simulations of GaN(0001) surface model were performed at different conditions. Meanders emerge out from initially straight steps of direction [1210] and for wide range of parameters they grow up to given width and then keep stable shape of well ordered waves during further crystal growing.
Meander wavelength depends on the particle flux incoming to the step as \( \left( \frac{F_d}{C_0} \right) \). When wavelength is of the order of terrace width or lower, all meanders grow independently and eventually surface ends up as a rough structure. Larger meander wavelength create regular, ordered structure. The amplitude of meanders of this structure grows up to the limit value and then the step shape does not change during further growth of the crystal. Such surface patterns are seen in the experiment as characteristic regular microscopic structures perpendicular to the initial step direction, “finger-like” structures [14,15,17]. We have shown that the crucial role in the explanation of described phenomena plays particle exchange between terraces via step. Such step to step communication is possible only when the Schwoebel barrier is low enough or zero. Meandering instability in this case most probably is caused by step movement which induces drift relative to steps. We described the mechanism of the step amplitude saturation leading to stationary motion of the meandered step pattern. We have not seen bunching instability of the simulated model at any parameters of study. It seems that the simple surface model we presented here is enough to analyze step meandering evolution, but additional elements have to be added to the model in order to observe both meandering and bunching during the same crystal growth process. We continue work under the model along this direction.

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