Meandering instability of curved step edges on growth of a crystalline cone

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We study the meandering instability during growth of an isolated nanostructure, a crystalline cone, consisting of concentric circular steps. The onset of the instability is studied analytically within the framework of the standard Burton-Cabrera-Frank model, which is applied to describe step flow growth in circular geometry. We derive the correction to the most unstable wavelength and show that in general it depends on the curvature in a complicated way. Only in the asymptotic limit where the curvature approaches zero the results are shown to reduce to the rectangular case. The results obtained here are of importance in estimating growth regimes for stable nanostructures against step meandering.

Keywords: Growth, surface diffusion, models of surface kinetics.

\section*{I. INTRODUCTION}

Film growth by Molecular Beam Epitaxy (MBE) is essentially based on the possibility to control growth on a submonolayer level. Usually one aims at surfaces as smooth as possible with atomistically sharp structures consisting of step edges or nanoscale islands. Growth of these structures in MBE is affected not only by stochastic but also by deterministic instabilities such as step meandering and bunching. Since the advent of modern film growth techniques these surface instabilities have been of theoretical and practical interest [1,2].

To obtain nanostructures in MBE with desired quality one of the fundamental concerns is the stability of step edges. Instabilities lead to nonuniform layers so it is advantageous to suppress them during growth. In step flow growth the basic mechanism and properties of the meandering instability were treated by Bales and Zangwill (BZ)\textsuperscript{3} on the basis of the classic Burton-Cabrera-Frank (BCF) model\textsuperscript{4}. The case of circular nanostructures has so far received much less attention although it is an example for a layered nanostructure\textsuperscript{5}. Decay of mesoscopic circular stepped structures can be described in some special cases of stable step flow with good accuracy using the BCF model\textsuperscript{4,5}. Also, recently the decay and bunching of a circular crystalline cone in the stable growth regime has been studied in detail using a similar approach\textsuperscript{6}. However, to our knowledge there has been no study of the meandering instability in the spirit of BZ during growth of circular nanostructures even though its importance was noted already a decade ago\textsuperscript{7}. It is thus of interest to understand under which conditions circular step edges are stable against meandering. In this report we present our study of the morphological instability of curved step edges. Our viewpoint is based on the BCF model and we generalize the BZ results to the case of a circular geometry. We perform a linear stability analysis and calculate the corrections to the results for the straight steps. As expected, our results reduce to the BZ case in the limit of an infinite step radius.

\section*{II. STEP FLOW GROWTH IN CIRCULAR GEOMETRY}

We study a model of circular steps which are placed concentrically on top of each other. The steps can absorb and emit atoms which diffuse on terraces between the steps. The terrace \(j\) is bounded by the steps at \(r_j\) and \(r_{j+1}\). Assuming the flux of adatoms onto and evaporating from the terraces, the adatom concentration \(c_j\) on the terrace \(j\) obeys the well known form of the BCF equation\textsuperscript{3,4}

\[
\frac{\partial c_j}{\partial t} = D_s \nabla^2 c_j - \frac{c_j}{\tau} + F, \tag{1}
\]

where \(D_s\) is the diffusion coefficient of an adatom on a flat terrace, \(\tau\) is the time scale for evaporation, and \(F\) is the deposition flux. Assuming that the adatom concentration relaxes much faster than the step edge moves we can assume that the terrace is in a quasi-stationary state corresponding to \(\partial c_j/\partial t = 0\). Mass transport through the bulk of the material is ignored.

The model is now fully specified with the choice of the boundary conditions and the requirement of the mass conservation at the step edges. The mass conservation implies that the edge velocity is given by\textsuperscript{6}

\[
V_j = V_{j+} + V_{j-} = \Omega D_s \left( \frac{\partial c_j}{\partial r_j} |_{R_j} - \frac{\partial c_{j-1}}{\partial r_j} |_{R_j} \right), \tag{2}
\]

where \(\Omega\) is the atomic area, \(V_{j+}, V_{j-}\) are the contributions to the step edge velocity due to surface currents from the upper and lower terrace, respectively, and \(R_j\) is the radius of the \(j\)th step edge. We assume that the velocities \(V_{j\mp}\) are related to the deviations of the adatom concentration from the equilibrium value\textsuperscript{6}

\[
V_{j\mp} = \Omega k_{\pm} [c_j - c_j^e], \tag{3}
\]

where \(c_j^e = c_0^e \exp[\tilde{\Gamma}_\kappa(R_j)]\) is the equilibrium adatom concentration at the edge, \(\tilde{\Gamma} = \Omega \gamma/(k_B T)\), \(\gamma\) is the free
energy/(unit length), \(c_0^{eq}\) is the equilibrium adatom concentration at the straight step, \(\kappa(R)\) is the local curvature of the step in the circular geometry (see e.g. Eq. (7) in Ref. [8]), and \(k_+\), \(k_-\) are the attachment coefficients associated with the upper and lower terraces, respectively. From Eqs. (2) and (3) we obtain the mixed boundary conditions at the step edge

\[
D_s \frac{\partial c_j}{\partial r} |_{R_j} = k_+ |c_j| R_j - c_j^{eq},
\]

\[-D_s \frac{\partial c_j}{\partial r} |_{R_{j+1}} = k_- |c_j| R_{j+1} - c_j^{eq} + 1.
\]

Defining a new field \(u_j = c_i - \tau F\) Eq. (1) becomes the Helmholtz equation in the stationary limit [9]:

\[
\nabla^2 u_j - \frac{1}{x_s^2} u_j = 0,
\]

where \(x_s = \sqrt{D_s} \tau\) is the diffusion length. The solution of Eq. (1) for the perfectly circular step is given by \(u_0^0(\hat{r}, \theta) = a_0^0 I_0(\hat{r}/x_s) + b_0^0 K_0(\hat{r}/x_s)\), where \(I_0(\hat{r})\) and \(K_0(\hat{r})\) are the zeroth order modified Bessel functions and \(a_0^0\) and \(b_0^0\) are coefficients determined by the boundary conditions. In the linear stability analysis a small perturbation is added to the step edge and the equations are solved to first order in the perturbation amplitude. We set \(\hat{r}_j(\theta) = R_j + \epsilon \exp[\imath n \theta + \omega t] + c.c.,\) where \(\epsilon\) is a small parameter, \(\omega\) a growth rate, \(|n| \geq 1\) is an integer (due to periodicity), and c.c. denotes the complex conjugate. The solution to Eq. (1) in \(\epsilon\) is given by

\[
u_j(\hat{r}, \theta) = u_0^0(\hat{r}, \theta) + \epsilon [A_n^0 I_n(\hat{r}/x_s) + B_n^0 K_n(\hat{r}/x_s)] e^{\imath n \theta + \omega t},
\]

where \(I_n(\hat{r})\) and \(K_n(\hat{r})\) are the modified Bessel functions of integer order \(n\), and the coefficients \(A_n^0\) and \(B_n^0\) are determined by the boundary conditions. When the solution is found, the growth rate \(\omega\) can be deduced using Eq. (2) and \(V_j = V_0^j + \omega \hat{r}_j(\theta)\). If \(\omega > 0\) the step edge is linearly unstable.

### III. MORPHOLGICAL INSTABILITY OF A CIRCULAR STEP

Qualitatively, the growth of stepped structures is unstable against step meandering when the flux of adatoms from the upper terrace is reduced, e.g. due to the Ehrlich-Schwoebel barrier. This is basically the origin of the morphological instability on vicinal surfaces (the BZ case) [3]. However, in the case of a circular step, the stabilizing effect of the step curvature is expected to be more pronounced than in the rectangular geometry. Therefore, we expect the possible instability to be weaker than in the rectangular case since the line tension tends to smooth out the steps. Here we consider the cases \(k_+ \to \infty, \ k_- = 0\) (one-sided model), and \(k_+ \neq k_-\) non-zero and finite (asymmetric model).

### A. One-sided model

In the one-sided model \(k_+ \to \infty\) corresponds to instantaneous attachment from the lower terrace and \(k_- = 0\) implies an infinite Ehrlich-Schwoebel barrier. In this limit the velocity of the step with radius \(R\) is given by \(V = V_+ = D \partial u/\partial r\) and the stability function becomes

\[
\omega(n) = \frac{\omega_BZ(q)}{\Omega \Delta F} = \frac{\xi}{\rho} - 1) \frac{b_1^+ + b_0^+ b_0^+}{a_n} + c_n \frac{\xi}{\rho^2} (1 - n^2),
\]

where \(\xi = \tilde{\Gamma}/(x_s \tau \Delta F)\) is the capillary length, \(\Delta F = F - c_0^{eq} \tau\), and \(\rho = R/x_s\). The coefficients are given by

\[
a_n = [I_n \tilde{K}_n - I_n \tilde{K}_n] [I_1 K_0 + I_0 K_1], \quad b_0^+ = [I_n \tilde{K}_n - I_n \tilde{K}_n] [I_1 K_0 - I_n K_1], \quad b_0^+ = [I_n K_n - I_n K_n] [I_1 K_1 - I_n K_1], \quad b_0^+ = [I_n K_n - I_0 K_n] [I_1 K_n - I_0 K_n], \quad c_n = [I_n K_n - I_n K_n] [I_1 K_n - I_0 K_n],
\]

where \(I_n = I_n(\rho), \tilde{I}_n = I_n(\rho + i/\xi, \xi), l\) is the terrace width, and the prime indicates the derivative with respect to the scaled variable \(\rho\). By using the asymptotic formulae of the modified Bessel functions for \(n\), large with \(n/R \equiv q = const.,\) the growth rate [3] can be shown to reduce to the result of Bales and Zangwill [3] in the limit \(R \to \infty\).

The stability function is plotted in Fig. 1 and it approaches a limiting form when the radius of the step increases (curvature decreases). As can be seen from the figure, there exists a limiting value \(R_\ast\) for the radius such that steps with \(R < R_\ast\) are always stable. For radii \(R > R_\ast\) there exists a critical value of the wavevector \(q\), such that for \(q > q_\ast\) the edge is stable. The critical wavevector depends on curvature and with increasing curvature \(q_\ast\) decreases, i.e. the critical wavelength where the instability sets in is shifted to larger wavelengths.

In the limit of large \(n\), \(R\) we obtain the correction term to the results of Bales and Zangwill of the order of \(1/R\) as

\[
\omega(n) = \omega_BZ(q) + \frac{x_s}{2R} \Sigma_q,
\]

where \(\omega_BZ\) is the rectangular result (Eq. (13) in Ref. [3]),

\[
\Sigma_q = \frac{1}{x_s^2} \left[ x_s^2 \lambda_2^2 \lambda_2^{-2} + \lambda_4^{-1} \tanh(\lambda_4 \tilde{r}) (2 \lambda_4^2 + \lambda_4^{-2}) + \tanh(\tilde{r}) - 2 \xi \frac{1}{\cosh(\lambda_4 \tilde{r}) \cosh(\tilde{r})} - \tan(\lambda_4 \tilde{r}) (2 + \lambda_4^2) (2 \lambda_4^2 + \lambda_4^{-2}) \tanh(\lambda_4 \tilde{r}) \tan(\tilde{r}) + 2 \xi (2 \lambda_4^2 - \lambda_4^{-2} \sin(\lambda_4 \tilde{r}) \sin(\tilde{r}) + \lambda_4^2 \sin(\lambda_4 \tilde{r}) \sin(\tilde{r})) \tanh(\lambda_4 \tilde{r}) + \lambda_4 \sqrt{1 + (x_s q)^2} + \lambda_3 \tilde{r} \right),
\]

\(\lambda_4 = \sqrt{1 + (x_s q)^2}\), and \(\tilde{r} = r/x_s\). Using the equation above we can determine analytically the corrections to
the critical wavevector defined as \( \omega(q_c) = 0 \). The results are for \( l \gg x_s \):

\[
x_{s}q_{c} \approx \left\{ \begin{array}{ll}
\frac{\sqrt{\frac{1}{\xi_{s}} + \frac{27}{24}(\frac{1}{\xi_{s}} - 2)}}{\sqrt{\frac{1}{4}(1 - 2\xi_{s}) - \frac{2x_{s}}{\xi_{s}}}}, & x_{s}q_{c} \gg 1;
\sqrt{\frac{1}{2\xi_{s}} - 1 - \frac{l}{R}}, & x_{s}q_{c} \ll 1,
\end{array} \right.
\]

(8)

and for \( l \ll x_s \) (and \( l^2 q_c^2 \ll 1 \)):

\[
x_{s}q_{c} \approx \sqrt{\frac{lx_{s}}{2\xi_{s}} - 1 - \frac{l}{R}}.
\]

(9)

Omitting the \( 1/R \) terms we obtain the BZ results for the rectangular geometry [3]. In Fig. 2 the curves for the critical wavevector \( q_c \) against the capillary length \( \xi_s \) are shown for \( l \gg x_s \), \( x_s q_c \ll 1 \). The corrected result follows the numerically plotted curve whereas the BZ result deviates considerably. For \( \xi_s \) large enough the edge is always stable and \( q_c \) approaches zero. The inset of Fig. 2 shows the case \( l \ll x_s \) which behaves in a similar way.

**B. Asymmetric model**

When the kinetic coefficients at the step edge from the lower and upper terrace are both finite and nonzero, the velocity of the \( j \)th step edge is given by \( V_j = D_j\partial u_j/\partial r - (\partial u_j-1/\partial r)|_{r = \xi_j(\theta)} \) and the growth rate becomes

\[
\omega(n_j) = \frac{\alpha_j}{\Omega \Delta F} \left[ \alpha_{j+1} - \alpha_{j-1} \right] I_0'(\rho_j) + \frac{\beta_j}{\gamma_j} - \frac{\beta_{j-1}}{\gamma_{j-1}} \frac{K''_0(\rho_j)}{K_0(\rho_j)},
\]

(10)

\[
+ \frac{B_j}{D_j} - \frac{B_j}{D_j} = I_n(\rho_j) + \frac{D_j}{D_j} K''(\rho_j),
\]

where \( \alpha_j, \beta_j, \) and \( \gamma_j \) are the coefficients related to unperurbed steps, and \( A_j, B_j, \) and \( D_j \) are obtained from the expansion linear in \( c \). Define \( d_\pm = d_\pm/x_s = (D_j/k_\pm)/x_s \), and \( K_{n,0}^i = K(\rho_j) + d_\pm K''(\rho_j) \) and \( T_{n,0}^j = I_n(\rho_j) + d_\pm T_{n,0}^j(\rho_j) \). Then the coefficients are given by

\[
\alpha_j = (\xi_\rho \rho_j - 1)K_{j,0}^{j+1} = (\xi_\rho \rho_j - 1)K_{j,0}^{j+1},
\]

\[
\beta_j = -(\xi_\rho - 1)\xi_\rho + (\xi_\rho - 1)\xi_\rho K_{j,0}^{j+1},
\]

\[
\gamma_j = T_{j,0}^{j+1} - T_{j,0}^{j+1} K_{j,0}^{j+1},
\]

\[
A_n = \alpha_j T_{n,0}^{j+1} K_{j,0}^{j+1} - \beta_j K_{j,0}^{j+1} + \beta_j K_{j,0}^{j+1} K_{j,0}^{j+1},
\]

\[
B_n = \alpha_j T_{n,0}^{j+1} K_{j,0}^{j+1} - \beta_j K_{j,0}^{j+1} + \beta_j K_{j,0}^{j+1} K_{j,0}^{j+1},
\]

\[
D_n = -\beta_j T_{n,0}^{j+1} K_{j,0}^{j+1} + \beta_j T_{n,0}^{j+1} K_{j,0}^{j+1}.
\]

The resulting expression for the growth rate \( \omega(q) \) is rather complicated and we have not found any essentially simpler form which would be accurate enough for all curvatures. However, the essential qualitative features can be extracted from the special cases, and the
behavior is similar to the one-sided case. The numerically plotted values of $\omega$ indicate that as $k_-$ approaches $k_+$ the growth rate $\omega \leq 0$ at all values of $q$. This is shown in Fig. 3. We can thus conclude that the one-sided model is the most unstable.

So far, we have only considered the case of in-phase step growth. The stability of growth depends also on the phase of the neighboring steps, and the above analysis can be extended to the more general situation with an arbitrary phase between adjacent steps. However, the general results give only little additional insight and only the numerical results are shown in the inset of Fig. 3. Increasing the phase difference between the two adjacent steps makes the steps more stable as in the case of rectangular geometry [11].

IV. DISCUSSION AND CONCLUSIONS

In this work we have generalized the meandering instability to structures made of concentric islands. We have shown that in the case of circular cones growth instability due to terrace diffusion can arise as in the case of rectangular geometry. However, the instability is suppressed by the curvature and structures with smaller sizes than a critical size are stable. The critical size depends on the microscopic parameters of the system. We present here also the asymptotic analytical results for small curvatures. In the limit $R \to \infty$ the growth rate approaches the expression found by Bales and Zangwill [3]. The more general case where attachment from the upper and lower terraces to the step are both finite and non-zero behaves qualitatively similarly to the one-sided model.

The results presented here extend the results of the BZ model for rectangular geometry and they are of interest in modeling e.g. the behaviour of isolated crystalline cones. Results given here can be used to check whether the simplified model which assumes the perfectly circular step edges in decaying cones is valid. If the meandering instability is expected to be present, it can play a significant role in the evolution of nanostructures.

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