Nonlinear evolution of the step meandering instability of a growing crystal surface

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The growth of crystal surfaces, under non-equilibrium conditions, involves the displacement of mono-atomic steps by atom diffusion and atom incorporations into steps. The time-evolution of the growing crystal surface is thus governed by a free boundary value problem [known as the Burton–Cabrera–Franck model]. In the presence of an asymmetry of the kinetic coefficients [Erlich–Schwoebel barriers], ruling the rates of incorporation of atoms at each step, it has been shown that a train of straight steps is unstable to two dimensional transverse perturbations. This instability is now known as the Bales-Zangwill instability (meandering instability). We study the non-linear evolution of the step meandering instability that occurs on a crystalline vicinal surface under growth, in the absence of evaporation, in the limit of a weak asymmetry of atom incorporation at the steps. We derive a nonlinear amplitude equation displaying spatiotemporal coarsening. We characterize the self-similar solutions of this equation.

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Molecular beam epitaxy (MBE) is often used to grow nano-structures on vicinal surfaces of semiconductor and metallic crystals. The ability to grow smooth crystals with a sharp interface is of considerable importance when manufacturing electronic and opto-electronic devices. One possibility for achieving this goal is to use the step-flow mode, where deposited adatoms diffuse and attach directly to preexisting steps on a vicinal surface. Ideally the surface grows without changing its shape by advancement of a uniform train of step traveling at constant velocity. In experiments, a wide number of additional effects may modify this scenario. Fluctuations in the beam intensity and island growth may lead to kinetic roughening of the surface. The presence of impurities can also pin the steps at random positions. More importantly, intrinsic morphological instabilities such as step bunching and step meandering can take place on a vicinal surface and lead to the destruction of a stable train of equidistant straight steps. Thus, under standard MBE growth conditions (one monolayer per minute) a rich variety of crystal surface morphologies is experimentally observed, resulting from the nonlinear evolution of step bunching and meandering instabilities. Moreover, the self-organization arising from these instabilities has been proposed as a natural candidate for the development of technological applications such as quantum dots and quantum wells.

The step meandering instability was originally predicted theoretically by Bales and Zangwill for a vicinal surface under growth. Its origin comes from the asymmetry between the descending and ascending currents of adatoms. As shown by Bales and Zangwill, a straight train of steps during MBE growth may become morphologically unstable in the presence of a kinetic attachment asymmetry at the step: the Erlich-Schwoebel effect (ES). The physical mechanisms taking place during this instability are the destabilizing ES effect combined with the stabilizing effect provided by the step rigidity. It was shown that the most dangerous mode is the synchronized mode for which all the steps have the same phase. Nonlinear extensions of this work have shown that the meander evolution can be described by amplitude equations showing various behaviors. Close to the instability threshold, starting from the Burton-Cabrera-Frank (BCF) model, it was shown that the step position in the presence of desorption (evaporation) obeys the Kuramoto-Sivashinsky equation:

\[
\partial_t h = -\partial_y^2 h - \partial_y^4 h + (\partial_y h)^2, \quad (1)
\]

where \( y \) is the coordinate along the step and \( x = h(y, t) \) is the meander amplitude which describes the step shape in the \((x, y)\) plane. The ultimate stage of this dynamics is thus found to be spatiotemporal chaos. In the case of negligible desorption with strong or moderate ES effect, it was shown that the step shape obeys a highly nonlinear equation

\[
\partial_t h = -\partial_y \left[ \frac{1}{1 + (\partial_y h)^2} \left( \partial_y h + \partial_y^3 \left( \frac{\partial_y^2 h}{(1 + (\partial_y h)^2)^{3/2}} \right) \right) \right]. \quad (2)
\]

This equation cannot be derived from a weakly nonlinear analysis but is based on the assumption that the slope of the steps is order unity. Instead of spatiotemporal chaos, a regular pattern is revealed, the lateral modulation wavelength is fixed while the amplitude of the step deformation (transverse meandering amplitude) increases like \( t^{1/2} \). Later, it was shown that the inclusion of the elastic step interactions affects the step dynamics in the sense that they induced a lateral coarsening. Finally, it was recently shown that interrupted coarsening occurs when two-dimensional diffusion anisotropy is included.

In this paper, we show using standard weakly nonlinear analysis, that under the assumption of negligible desorption and weak Erlich-Schwoebel effect (ES), the time-evolution of the Bales-Zangwill instability is governed by...
the following equation:
\[ \partial_t h = - \partial_y^2 h - \partial_y^3 h + \partial_y^2 (\partial_y h)^2 . \]  
(3)

This equation was already mentioned in Ref.18 on the basis of symmetry arguments as a possible candidates for the time evolution of the meandering amplitude but was not explicitly obtained due to a different choice of the physical parameters (large and order one ES effects). Our results are illustrated by numerical simulations. We show using a simple similarity argument, that the characteristic transverse meandering amplitude (step width) grows like \( t \) and that the characteristic lateral coarsening exponent grows like \( t^{1/2} \). Our result are in agreement with the numerical simulations performed in Ref.24.

Let us denote by \( x_n(y,t) \) the positions at time \( t \) of the \( n \) step (see Fig. 1). For simplicity we neglect elastic interactions between steps and assume that the desorption of adatoms and transparency of the steps are negligible. During growth, the adatom surface concentration on each terrace \( C_n(x,y,t) \), obeys the following diffusion equation,18-24,
\[ D \nabla^2 C_n(x,y) + F = 0 , \]  
(4)

where \( D \) is the adatom diffusion coefficient, and \( F \) the deposition flux. This equation for \( C_n \) is supplemented by the following boundary conditions:
\[ D\hat{n} \cdot \nabla C_n = \nu_+(C_n - C_{eq,n-1}) , \quad x = x_{n-1}(y,t) , \]  
(5)
\[ D\hat{n} \cdot \nabla C_n = -\nu_-(C_n - C_{eq,n}) , \quad x = x_n(y,t) . \]  
(6)

Here \( \nu_+ \) and \( \nu_- \) are the ES coefficients which are proportional to the rate of attachment of adatoms on the steps from the terrace; \( \hat{n} \) is the external normal to the step :
\[ \hat{n} = (1, -\partial_y x_n)/\sqrt{1 + (\partial_y x_n)^2} , \]  
(7)

and \( \nabla \) is the two dimensional gradient operator (\( \partial_x, \partial_y \)). The adatom equilibrium concentrations \( C_{eq,n} \) depend on the step curvatures \( \kappa_n \),
\[ C_{eq,n} = C_{(0)}(1 + \Gamma \kappa_n) , \]  
(8)

where \( \Gamma = \Omega \gamma/k_BT \), with \( \Omega \) the unit atomic surface, \( T \) the temperature, \( k_B \) the Boltzmann constant, \( \gamma \) the step rigidity and \( C_{(0)} \) the reference adatom concentration. The step curvature is given by
\[ \kappa_n = - \frac{\partial_x^2 x_n}{(1 + (\partial_y x_n)^2)^{3/2}} . \]  
(9)

The normal velocity of the \( n \) step is given by
\[ v_n = D\Omega(\hat{n} \cdot \nabla C_{n+1} - \hat{n} \cdot \nabla C_n) , \quad x = x_n(y,t) . \]  
(10)

The \( x \) component of the normal velocities reads:
\[ \dot{x}_n = v_n(1 + (\partial_y x_n)^2)^{1/2} . \]  
(11)

where the dot represents the time derivative.

In order to get a non-dimensional version of equations (14-15-18-19), we set the unit of length to be the initial size of the terrace \( l_0 \) (initial distance between steps) and the unit of time to be \( l_0^2/(C_{(0)}\Gamma D) \). Specifically, one sets
\[ \hat{x} = \frac{x}{l_0} , \quad \hat{y} = \frac{y}{l_0} , \quad \hat{C}_n = \frac{l_0[C_n - C_{(0)}]}{C_{(0)} \Gamma} , \quad \hat{t} = \frac{t \Omega C_{(0)} D \Gamma}{l_0^2} \]  
(12)

in equations (14-15-18-19). We obtain after omitting the tildes on the variables the following equations for the dimensionless variables
\[ \nabla^2 C_n = -f , \]  
(13)
\[ \hat{n} \cdot \nabla C_n = \alpha_+(C_n - \kappa_{n-1}) , \quad x = x_{n-1}(y,t) \]  
(14)
\[ \hat{n} \cdot \nabla C_n = -\alpha_-(C_n - \kappa_{n}) , \quad x = x_n(y,t) \]  
(15)
\[ v_n = \hat{n} \cdot \nabla C_{n+1} - \hat{n} \cdot \nabla C_n , \quad x = x_n(y,t) \]  
(16)

The system is thus controlled by three independent positive nondimensional parameters:
\[ f = \frac{F l_0^3}{C_{(0)} \Gamma D} , \quad \alpha_+ = \frac{\nu_+ l_0}{D} , \quad \alpha_- = \frac{\nu_- l_0}{D} , \]  
(17)
respectively related to the flux and attachment lengths. Let us investigate the linear stability of a train of equidistant steps traveling at a constant velocity \( f \) when perturbed transversally. The shape of the steps can be decomposed in Fourier modes of the form
\[ x_n(y,t) = n + f t + \xi_n(y,t) \]  
(18)
where
\[ \xi_n(t,y) = e^{\sigma(q,\phi)t+iqq+in\phi} , \]  
(19)
where \( q \) and \( \phi \) are respectively the wavenumber and the phase of the perturbation (see Fig. 1). Inserting these
expressions into equations [13-15,16], we obtain the general dispersion relation \( \sigma = \sigma(q, \phi) \) which possesses, for each \( \phi \), one maximum [13,14]. The maximum growth rate is reached for the in-phase perturbation \( \phi = 0 \). Although the full expression of the dispersion relation is cumbersome, near the instability threshold (\( f = 0 \)), we introduce a small parameter \( \epsilon \) measuring the distance to the threshold and we also assume that the E-S effect is small. This latter assumption was not used in previous works [10,18]. The small parameter \( \epsilon \) arises naturally when considering the long wavelength limit, in which \( q \rightarrow \epsilon q \). We therefore set

\[ \tilde{f} = \epsilon f, \quad \tilde{C} = \epsilon^2 C, \quad \alpha_+ = \alpha_- + \epsilon^3 \delta, \quad \tilde{t} = \frac{t}{\epsilon^2} \] (20)

in equations [13-15,16]. This scaling will lead to the following equations after omitting the tilde

\[ \nabla^2 C_n = -f \epsilon \] (21)
\[ \hat{n} \cdot \nabla C_n = \alpha_+(C_n - \epsilon^2 \kappa_{n-1}), \quad x = x_{n-1}(y, t) \] (22)
\[ \hat{n} \cdot \nabla C_n = -\alpha_-(C_n - \epsilon^2 \kappa_n), \quad x = x_n(y, t) \] (23)
\[ v_n = \hat{n} \cdot \nabla C_{n+1} - \hat{n} \cdot \nabla C_n, \quad x = x_n(y, t). \] (24)

To lowest order in \( \epsilon \) we find that the growth rate \( \sigma \equiv \sigma(q, 0) \) is,

\[ \sigma = \epsilon^6 \left( \delta f \alpha_- (\alpha_- + 2) \frac{\epsilon^2}{2} - \epsilon^4 \right), \] (25)

The growth rate \( \sigma \) is maximum for the wavenumber \( q_{\text{max}} = \sqrt{\delta f \alpha_- (2 + \alpha_-)/2} \), so that the most unstable wavelength \( \lambda \) should scales \( \sim 1/\sqrt{f} \). The meandering instability originates from the asymmetry between the descending and ascending currents of adatoms; the instability occurs when the rate of attachment of adatoms from the terrace lower \( \alpha_+ \) is greater than the rate of attachment of adatoms from the upper terrace. In the opposite case, the meandering instability is not present but a step bunching instability develops. However, at this moment due to the technical difficulties of the experiments there has not yet been a clear demonstration of the Bales-Zangwill instability.

We study now the nonlinear evolution of the meandering instability, in the limit of weak amplitudes and long wavelengths. In order to obtain the relevant nonlinear dynamics we use a standard multi-scale method. Adatoms concentrations and step shapes are expanded in powers of \( \epsilon \). The adatoms concentrations depends on the slowly varying space and time variables: \( Y = \epsilon y \) and \( T = \epsilon^2 t \). The details of the calculations are referred to Appendix A. We thus write

\[ C_n(x, y) = \sum_{i=1}^{7} \epsilon^i C_n^{(i)}(x, Y), \quad \xi_n(y, t) = \epsilon u(Y, T). \] (26)

Solving diffusion equation (21) and boundary conditions [22,23] up to order \( \epsilon^3 \), and inserting the results into the step velocity equations (24), we find the equation for the slowly varying amplitude at order \( \epsilon^2 \). It may be written as:

\[ \partial_t u = -\partial_y^2 \left[ \frac{\delta f}{2} \alpha (\alpha + 2) u + \partial_y^2 u + \frac{f [\alpha (\alpha + 6) + 6]}{6\alpha (\alpha + 2)} (\partial_y u)^2 \right], \] (27)

where \( \alpha = \alpha_- \). We have renamed the capital letters \( T \) and \( Y \) as \( t \) and \( y \) for simplicity. It is necessary to go to the seventh order to balance the nonlinear term with the linear ones. This equation was recently presented in the context of surface growth on a vicinal surface of silicon characterized by different types of steps and terraces [25]. In this previous work, it was shown that the anisotropy of diffusion induces a meandering instability and simple similarity and matching arguments lead to a complete picture for the long time behavior of the solution of equation (27). It is worth noting that Eq. (27) admits an exact particular solution in the form of a stationary parabola

\[ u(y, t) = -\frac{3\alpha^2 (2 + \alpha)^2 \delta}{4(6 + 6\alpha + \alpha^2)} y^2. \] (28)

Trying a similarity solution \( u(y, t) = t^a \varphi(y/t^b) \) of equation (27), we obtained the exponents \( b = 1/2 \) and \( a = 1 \), which agree with our numerical results [25]. The general, asymptotic solution of equation (27) can be thought of a superposition of parabolas as shown on the Figure 3 of Ref. [25]. The joining regions between parabolas can be matched using a solution of the Burgers equation obtained by a Hopf-Cole transformation [25]. A typical spatiotemporal evolution from a random initial condition is shown in Fig. 2. The step width (transverse meandering amplitude) of the steps increase linearly in time [25] and this results is in agreement with the results obtained by Kinetic Monte-Carlo methods in the Fig. 7 of Ref. [22].

In this article, we have shown that the amplitude of the meanders is governed by Eq. (27) which displays non-interrupted coarsening. It would be interesting in a further study to investigate the effect of phase freedom of the steps and to derive a continuous coarse-
FIG. 2: Spacetime plot of \( u(y, t) \) with nondimensional \( y \) and \( t \) axes, given by the numerical solution of equation (27) with \( \alpha = 1, \delta = 1 \) and \( f = 1 \). The coarse-graining of structures leads to a superposition of parabolas, with a size \( \langle u^2 \rangle^{1/2} \sim t \). In the long-time state all the parabolas tend to have unity curvature at their maximum, and width increasing as \( \sqrt{t} \).

This paper and the strongly nonlinear regime described in Refs. 16, 17, 18.

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**APPENDIX A: DETAILS OF THE EXPANSION**

From equations (21-22-23-26), we obtain the following relations:

\[
\partial_x^2 C_n^{(1)} = -f, \quad (A1)
\]

\[
C_n^{(1)}(x, Y) = \frac{f - fx(x + 1)\alpha}{2\alpha},
\]

\[
\partial_x^2 C_n^{(2)} = 0, \quad (A2)
\]

\[
C_n^{(2)}(x, Y) = \frac{1}{2}fu(Y)(2x + 1),
\]

\[
\partial_x^2 C_n^{(3)} + \partial_Y^2 C_n^{(1)} = 0, \quad (A3)
\]

\[
C_n^{(3)}(x, Y) = -\frac{fu^2(Y)}{2},
\]

\[
\partial_x^2 C_n^{(4)} + \partial_Y^2 C_n^{(2)} = 0, \quad (A4)
\]

\[
C_n^{(4)}(x, Y) = \frac{6f(x\alpha - 1)\delta - f(2x + 1)\alpha^2 [x(x + 1)(\alpha + 2) - 1] u''(Y)}{12\alpha^2(\alpha + 2)},
\]

\[
\partial_x^2 C_n^{(5)} + \partial_Y^2 C_n^{(3)} = 0, \quad (A5)
\]

\[
C_n^{(5)}(x, Y) = \frac{3(\alpha + 2) [f(2x(x + 1)\alpha - 1)u'(Y)^2 - 4\alpha u''(Y)] + fu[\alpha + 6x(x + 1)(\alpha + 2)]u''(Y) - 6\delta}{12\alpha(\alpha + 2)},
\]

\[
\partial_x^2 C_n^{(6)} + \partial_Y^2 C_n^{(4)} = 0, \quad (A6)
\]
\[ C_n^{(6)}(x, Y) = \frac{-f(2x + 1)\left(360u(Y)u^2(Y)(\alpha + 2)^2 + 180u(Y)u''(Y)(\alpha + 2)^2\right)}{720(\alpha + 2)^2} \]

\[ - \frac{f(2x + 1)\left(-\alpha + x(x + 1)(\alpha + 2)(-\alpha + 3x(x + 1)(\alpha + 2) - 12) + 12)u'''(Y)\right)}{720(\alpha + 2)^2}, \]

\[ \partial_x^2 C_n^{(7)} + \partial_Y^2 C_n^{(5)} = 0, \quad (A7) \]

\[ C_n^{(7)}(x, Y) = \frac{1}{(720\alpha^3(\alpha + 2)^2)^\gamma}. \quad (A8) \]

\[ \gamma = 360f(\alpha + 2)^2u^2(Y)u^2(Y)\alpha^3 + 120f(\alpha + 2)^2u^3(Y)u''(Y)\alpha^3 \]

\[ - f(30(\alpha + 2)^2x^4 + 60(\alpha + 2)x^3 + 30(\alpha + 2)x^2 - 60(\alpha + 2)x - \alpha(\alpha + 12))u(Y)u'''(Y)\alpha^3 \]

\[ - 30(f\alpha(\alpha + 2)(\alpha(2 + \alpha) + x(x + 1))u(3x(x + 1)(\alpha + 2) - 2(\alpha + 6)) - 12)u''(Y)) \]

\[ - 30(-2f\alpha(3(\alpha + 2)x^2 + 2(\alpha + 3)x - 2) - 6)\delta u''(Y) \]

\[ - 30(2(6f(\alpha + 1)(x\alpha - 1)\delta^2) \]

\[ - 30(\alpha(\alpha + 2)(f\alpha(\alpha + 2)(x(x + 1)u(-\alpha + 2x(x + 1)(\alpha + 2) - 7) - 6) + 6)) \]

\[ - 30(-6\alpha(\alpha + 2)(x(x + 1)\alpha - 1)u'''(Y)), \]

Here the symbol \( \prime \) denotes the derivative with respect to the variable \( Y \).
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