Pulses and disorder in a continuum version of step-bunching dynamics

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Starting from the Burton-Cabrera-Frank model in the presence of electromigration, we derive a nonlinear continuum version of step dynamics, where steps undergo a bunching instability. The obtained equation for the steps density is a mixture of the Korteweg–de Vries equation, which leads to solitons, and the Burger equation, which exhibits spatiotemporal chaos. For a small dispersive term, spatiotemporal chaotic pulses prevail. On increasing this term, we observe gradually a transition towards regular pulses. [S1063-651X(96)51705-8]

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One of the most common causes for morphology alteration of an initially vicinal surface is due to step bunching. Step bunching can occur in a variety of situations. For example, during heteroepitaxial growth, the strain energy can cause a step-bunching instability, which may be either of thermodynamical origin (the Asaro-Tiller-Grinfeld effect [1–3]), or may result from a kinetic instability [4]. Similarly, during homoepitaxial growth the impurity pinning on the terrace can lead to step bunching [5,6]. A more familiar example of the step bunching instability arises during sublimation provided that an asymmetry of the sticking between the upper and lower terraces at the steps exists [7] (the Schwoebel effect). On the experimental level, perhaps the most controllable situation corresponds to the case of subliminating silicon (111) by a dc heating current. In such a situation, it has been reported by several groups [8–12] that the surface morphology depends on the direction of the heating current. More precisely, when the current direction coincides with the ascendent one, step bunches appear in the temperature range $\sim 1300-1500$ K, disappear in the range $\sim 1500-1600$ K and again reappear above 1600 K. Reversing the current direction leads to a complementary picture (stable intervals become unstable, and vice versa). A step forward was made by Stoyanov [13], who evoked the electromigration effect on adatoms. This explains nicely the first transition (from stable to unstable), but it gives no hint to the reappearence of step bunches at higher temperature. We have recently shown [14] that the inclusion of advacancies in the step flow model reproduces the high temperature behavior of vicinal surfaces. Another alternative based on the assumption of incomplete melting was suggested which seems to account for the low temperature behavior [15].

All these studies were linear. The main outcome of a linear theory is the determination of the onset of instability, and the range of those perturbations which are likely to grow first. If the new structure is to be determined, and/or the long time evolution to be ascertained, then a nonlinear analysis is necessary. The aim of this Rapid Communication is to deal with the nonlinear behavior above the step-bunching instability. For the sake of simplicity, and without loss of generality, we shall consider the case of relatively low temperatures where advacancies can be neglected, and focus on the case of electromigration-induced step bunching, which seems to us as a canonical experimental example. It must be

kept in mind, however, that the type of equation we shall derive is generic, and it should therefore apply to the above mentioned situations as well.

The step flow model incorporates adatom diffusion, electromigration, evaporation, and sticking at the steps. Let $c(\mathbf{r},t)$ denote the adatom areal density. Mass conservation imposes

$$\frac{\partial c}{\partial t} = D\nabla^2 c - \frac{c}{\tau} - \frac{DF}{k_B T}\frac{\partial c}{\partial x}, \qquad (1)$$

where τ is the desorption time, D the diffusion constant, F the electromigration force, and $k_B T$ the thermal excitation energy. At each step $x=x_m(t)$ (m labels the mth step) the kinetic equation takes the form

$$\pm D\left[\frac{\partial c}{\partial n} - \frac{F}{k_B T}c\right]_{\pm} = \nu_{\pm}\left[c - c_{eq}\left(1 + \frac{\Omega}{k_B T}\frac{\partial \mathcal{E}}{\partial x}\right)\right] \qquad (2)$$

where $\nu_{\pm}$ is a kinetic coefficient (having the dimension of a velocity). The $+$ sign refers to the lower side, and $-$ to the upper one. Our convention is that the descendent direction is along the positive x axis. The quantity $\mathcal{E}$ designates the elastic interaction. For homoepitaxy, and if only first neighbors interaction is taken into account, $\mathcal{E}=\mathcal{A}[(x_m-x_{m-1}+l_0)^{-2}+(x_{m+1}-x_m+l_0)^{-2}]$, where $\mathcal{A}=2(1-\sigma^2)f^2/\pi E$, σ and E are the Poisson ratio, and Young modulus, and f a force. The quantity $\mathcal{A}$ measures the strength of the elastic interaction [16] and has a dimension of an energy multiplied by a length. From a dimensional analysis $f\sim Ea^2$ where a is an atomic length, so that $\mathcal{A}\sim Ea^4$. Typically $E\sim 10^{10}$ Pa, $a\sim(1-3)\times 10^{-10}$ m, and then $\mathcal{A}\sim(10^{-29}-10^{-30})$ J m. This is consistent with the experimentally measured value $\sim 10^{-30}$ J m for silicon [17].

Finally, if $J_{\pm}=D(\partial c/\partial x - Fc/k_B T)_{x=0^{\pm}}$ denotes the mass current across the step, the normal velocity is given by

$$v_n = \Omega(J_+ - J_-). \qquad (3)$$

The set of equations (1)–(3) completely describes step dynamics during sublimation. Before proceeding to the analysis, some remarks are in order. We disregard step meandering which will be the subject of a future work. We shall make use of the quasisteady approximation, which is valid

for all practical purposes. The diffusion field on a given terrace can easily be found to be given by $c = a\cosh(x/\alpha_0) + b\sinh(x/\alpha_0)$, $\alpha_0 = \sqrt{x_s^{-2} + 1/4\xi^2}$, $x_s = \sqrt{D\tau}$, $\xi = k_B T/F$ (it has a dimension of a length), and where a and b are integra-

tion factors which are easily determined by making use of Eqs. (2). Finally use of the continuity equation (3) provides us with the step position evolution as a function of neighboring steps

$$
(D\Omega c_{eq})^{-1}\dot{x}_m = \mathcal{D}^{-1}(\Delta x_{m+1})\left\{\alpha_0 e^{-\Delta x_{m+1}/2\xi}\left[1 + \frac{A}{3\Omega c_{eq}}\Delta_2\right] - \left[\alpha_0\cosh(\alpha_0\Delta x_{m+1})\right.\right.
$$

$$
\left.+ (1/2\xi + d_-/x_s^2)\sinh(\alpha_0\Delta x_{m+1})\right]\left[1 + \frac{A}{3\Omega c_{eq}}\Delta_1\right]\right\}
$$

$$
+ \mathcal{D}^{-1}(\Delta x_m)\left\{\alpha_0 e^{\Delta x_m/2\xi}\left[1 + \frac{A}{3\Omega c_{eq}}\Delta_0\right] - \left[\alpha_0\cosh(\alpha_0\Delta x_m)\right.\right.
$$

$$
\left.+ (-1/2\xi + d_+/x_s^2)\sinh(\alpha_0\Delta x_m)\right]\left[1 + \frac{A}{3\Omega c_{eq}}\Delta_1\right]\right\} \tag{4}
$$

where $d_{\pm} = D/\nu_{\pm}$ is the Schwoebel length, $\Delta x_{m+1} = x_{m+1} - x_m$, etc., $\mathcal{D}(x) = [1 + d_+ d_-/x_s^2 + (d_+ - d_-)/2\xi]\sinh(\alpha_0 x) + \alpha_0(d_- + d_+)\cosh(\alpha_0 x)$, $\Delta_i = \Delta x_{m+i}^{-3} - \Delta x_{m+i-1}^{-3}$ ($i = 0, 1, 2$), and $A = 6\Omega^2 c_{eq}\mathcal{A}/k_B T$. Equation (4) represents, in principle, an infinite set of equations. Rather than using a microscopic picture of step dynamics (by treating each step separately), our aim here is to use a "coarse-grained" image of step bunches by resorting to a continuum limit. For that purpose we assume that the interstep distance is much smaller than the diffusion length x_s. This holds, in particular, in the case of Si(111) for moderate temperatures. We shall simplify the analysis further by assuming $d_+ = d_- = d$. That is, we neglect the asymmetry due to the Schwoebel barrier. The asymmetry is caused by the electromigration force. It must be emphasized however that a finite sticking rate (d finite) is necessary in order to account for linear instability of the vicinal surface. Equation (4) is highly nonlinear. We shall truncate it to some order. For such an operation to be legitimate we assume that the growth rate of the instability is small enough (see below). This is satisfied by concentrating on the situation close to the instability threshold. Our strategy then is (i) to expand Eq. (4) in powers of Δx_m and (ii) to treat the subscript m as a continuous variable. Our treatment is valid for long wavelength modulations. Let us first extract from Eq. (4) only the linear part and let $mh \equiv x$ and $W(x) = x_{m+1} - x_m$. Equation (4) yields

$$
(1/\Omega D c_{eq})[W_t - (\Omega c_{eq}/\tau)W_x]
$$

$$
= -\beta_2 W_{xx} - \left(\frac{l_0^2\beta_2}{12} + \frac{A}{2l_0^4 d\Omega c_{ceq}}\right)W_{xxxx}
$$

$$
- \left(\frac{1}{6x_s^2} - \frac{A}{2\Omega c_{eq}\xi d l_0^3}\right)W_{xxx} \tag{5}
$$

where $\beta_2 = [1/(2\xi d) - A/(\Omega c_{eq}x_s^2 l_0^3)]$. The quantity l_0 is the zeroth order interstep distance. Note that we have truncated the expansion at the fourth derivative. In a multiscale analy-

sis developed below, it will be recognized that this is sufficient. There are two types of terms. The first type concerns the odd derivatives (which would not affect the stability—these are propagative terms)—and the second the even ones. (i) W_{xx} may be destabilizing or stabilizing according to whether $1/2\xi$ is smaller or larger than $dA/(\Omega c_{eq}x_s^2 l_0^3)$. In other words, electromigration (recall that $1/\xi \sim F$) destabilizes the vicinal surface (only if $F > 0$), while elasticity always stabilizes it. (ii) W_{xxxx} is always stabilizing for $\xi > 0$. It is composed of two contributions: one coming from electromigration and the second stems from elasticity. For very short interstep distances the latter dominates. Note that the first derivative in Eq. (5) can always be absorbed in W_t by means of a Galilean transformation $x \to x - v_0 t$ (where $v_0 = \Omega c_{eq}/\tau$ is nothing but the step velocity in the original vicinal train for a unit interstep distance—recall that x is scaled by h), and we shall omit it in the following.

Our treatment is expected to be valid in the long wavelength limit. As we shall see immediately, this situation is encountered close to the instability threshold. The critical condition for the onset of instability is obtained for $1/2\xi = dA/(\Omega c_{eq}x_s^2 l_0^3)$. As in other contexts [18], we introduce a small parameter $\epsilon = 1 - 2dA\xi/\Omega c_{eq}x_s^2 l_0^3$ which measures the distance from the threshold. In Fourier space ($W \sim e^{iqx + \omega t}$), we obtain from Eq. (5) $\omega \sim \epsilon q^2 - q^4 + iq^3$. The real part controls the instability. The fastest growing mode (obtained by setting $\partial Re\,\omega/\partial q = 0$) corresponds to a wave vector which scales as $\sqrt{\epsilon}$ (in real space this corresponds to the long wavelength regime) and the corresponding growth rate scales as ϵ^2. The imaginary part of ω would scale as $\epsilon^{3/2}$ and it dominates in principle (see below). This means that in a multiscale analysis we must introduce a short time associated with propagation, and a long time, the scale of which determines that of the amplification or the attenuation of the instability. The total time $\mathcal{T} = \epsilon^2 t_1 + \epsilon^{3/2}t_2$, where t_1 is the long time and t_2 the short one. Now we go back to Eq. (4) and pursue our expansion in a manner very similar to that developed in [18]. The first nonlinear term that appears is of the form WW_x, and the next one $W_x^2 + WW_{xx}$. Both terms scale as W^2. However, the first term contains only one
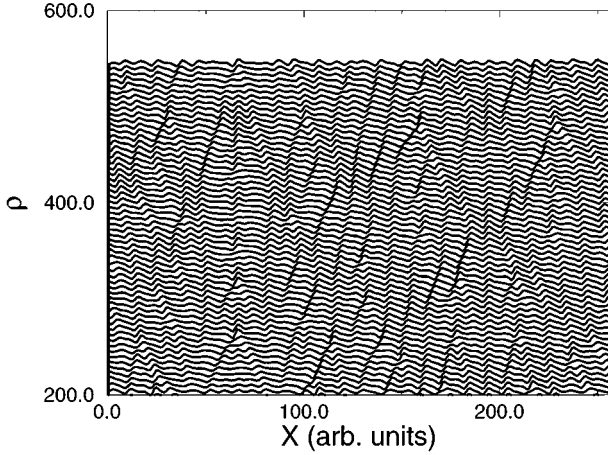
FIG. 1. The spatiotemporal portrait of the step density for $\beta=0.5$. The pattern is chaotic. The vertical coordinate represents the step density at different times as a function of the coordinate along the vicinal direction. The time unit is arbitrary.

derivative, and it is this one which dominates (in the long wavelength regime we are interested in). We can then show using a balance between the nonlinear term and the linear ones that the amplitude will scale as $W\sim\epsilon^{3/2}$. Rather than using the quantity W, we may as well use the steps density $m=1/W\equiv m_0+m_1$, where m_0 is the initial density (in the vicinal regime). Using Eq. (4) together with the nonlinear term, it is a simple matter to show that to leading order the steps density obeys the following equation:

$$\rho_T=-\rho_{XX}-\rho_{XXXX}-\beta\rho_{XXX}-\rho\rho_X \qquad (6)$$

where we have set $\mathcal{T}=\epsilon^2 D(\Omega c_{eq})^2 l_0^4/(2Ad\xi^2)t$, $X=\sqrt{\epsilon}l_0[\Omega c_{eq}/(A\xi)]^{1/2}x$, $\rho=\epsilon^{3/2}m_1 x_s^4 l_0^2(2\Omega c_{eq})^{1/2}/[(2\xi)^{3/2}A^{1/2}d^2]$, and $\beta=\epsilon^{-1/2}[l_0^2 d(\xi\Omega c_{eq})^{1/2}/(3x_s^2 A^{1/2})-l_0^{-1}A^{1/2}/(\xi\Omega c_{eq})^{1/2}]$. That the right hand side of Eq. (6) can be written as a derivative is no surprise; this simply expresses the conservation of steps. In the absence of the dispersive term ρ_{XXX}, Eq. (6) reduces to the Burger equation, which is a variant of the Kuramoto-Sivashinsky (KS) equation [18]. Note, for example, that the KS equation describes the meandering of an advancing isolated step [18]. When the destabilizing term (ρ_{XX}) together with the smoothing one ρ_{XXXX} are both absent, Eq. (6) reduces to the Korteweg–de Vries equation (KDV) [19]. Thus Eq. (6) is a mixture of the KS and the KDV equations. The KS equation is known to produce spatiotemporal chaos, while the KDV one gives rise to solitons.

It is thus an important question to see the consequence of the competition between solitons and chaos. Equation (6) has been solved numerically by means of a gear backward difference, where the derivatives are evaluated in Fourier space. Equation (6) possesses the following linear spectrum (we seek perturbations of the form $\rho\sim e^{ikX+W\mathcal{T}}$): $W=k^2-k^4+i\beta k^3$. The cutoff wave number (determined by $Re\,\omega=0$) is given by $q_c=1$. This means that if the extent of the system L is smaller than $2\pi/q_c\equiv\lambda_c$ then no instability takes place. In practice the system extent is very large ($L>>\lambda_c$) and one has to solve Eq. (6) for L large. Several simulations have been performed with L ranging from $10\lambda_c$ to $40\lambda_c$. Figures
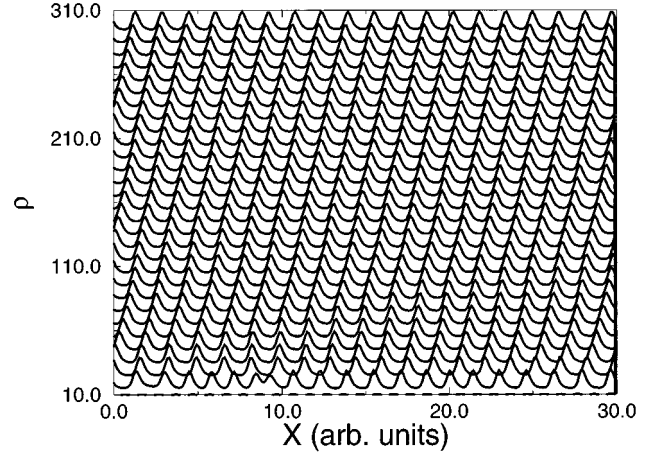

FIG. 2. The same quantity as in Fig. 1 but for $\beta=3$. The bunches are ordered.

1 and 2 show the spatiotemporal portrait of the step density for two different values of the dispersive term β. For small β (Fig. 1, $\beta=0.5$) we obtain spatiotemporal chaos. That is to say the pattern would not have any intrinsic order: the bunches would be spatially disordered. On increasing β, there is an emergence of more pronounced pulses with a tendency towards an ''ordering'' of the bunches. Figure 2 shows the pattern for $\beta=3$. It is interesting to note that when starting from a pattern with $\beta=0$ (which is disordered), and then switching on to $\beta\sim1$ we observe the birth of a localized pulse which propagates sideways. The successive passages of the ''soliton'' on the initially disordered pattern leaves behind it a more ordered structure. It seems as if the ''soliton'' acts as a sort of order selector. This phenomenon bears a strong resemblance to the situation encountered during directional growth of a nematic phase at the expense of the isotropic phase [20]. In that problem there are some circumstances where a ''solitarylike'' wave propagates along the cellular structure which had initially a rather strong wavelength dispersion. The successive passages of the ''solitary'' wave reduces drastically the dispersion. The ''soliton'' there is believed to play the role of a wavelength selector.

We are investigating experimental regimes in order to evaluate the coefficient β, and thus to decide which type of structure would be expected in a real situation (disorder, order, or a somewhat intermediate situation). This coefficient stems from the dispersion of the wave train indicating thereby that the phase velocity is different from the group velocity of the bunches. The definition of β seems to imply that this coefficient is large since it scales as $\epsilon^{-1/2}$. Our treatment is valid as long as the whole coefficient β is not much larger than unity. A value of the order of 2 or 3 is sufficient to produce regular pulses. On the one hand, while our regime is asymptotic, it may be legitimate even for $\epsilon\sim0.1$—or even larger—thus $\epsilon^{-1/2}\sim3$. On the other hand, from rough estimates we find that the prefactor of $\epsilon^{-1/2}$ may be as small as 0.1 by using realistic values that enter its expression, and that therefore β may easily be rendered as small as $\beta\sim1$ [21] in a given experiment. We shall give further details in an extended paper. A precise confrontation with experimental results would be decisive in order to guide further developments. We are now studying the evolution of the mean pulse

width and the mean pulse-pulse distance as a function of temperature, and we hope to report along these lines in the near future.

In summary we have shown from the Burton-Cabrera-Frank model which incorporates electromigration, that close to the step-bunching instability threshold the bunch dynamics obey an equation which is a mixture of the Kuramoto-Sivashinsky (or Burger) equation and the Korteweg–de Vries one. The increase of the dispersive term leads to a transition from disorder to order with more and more pronounced pulses. This analysis has given a qualitative behavior of the evolution of the bunches on a coarse-grained scale. While we have restricted our attention to the case where electromigration is the driving source for step bunching, we believe that the equation we have derived here is generic and

should arise irrespective of the details of the underlying physics. Among other open questions are (i) how the step meandering affects bunch dynamics, (ii) what is the role of fluctuations, and to what extent ordered pulses (if they are to arise) can suffer from statistical fluctuations?

Note added in proof. After this work was submitted we received a paper by M. Sato and M. Uwaha [22], who dealt with the problem of step bunching induced by the Schoebel barrier. Their equation is identical to ours. At the same time V. Hakim (private communication) pointed out to us that Eq. (6) was met, and some of its properties considered, in other contexts [23,24].

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