

Supporting Information to:

Scaling and dynamic stability of model vicinal surfaces

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In the beginning of this supplement we provide a slightly different than the original version of Dimensional Analysis (DA) of a generalized continuum equation for the time evolution of the (crystal) surface height – the so called Pimpinelli-Tonchev-Videcoq-Vladimirova (PTVV) equation^{1, 2} in order to illustrate the concept of universality classes in step bunching, then we will provide numerical evidence for the attribution of the Liu and Weeks model³, a model aimed at the attachment-detachment at steps or kinetics limited (KL) regime of the instability, to the $\rho=-1$ universality class and how this picture is modified in the other limiting regime – the surface diffusion limited (DL) one by studying the model of Sato and Uwaha⁴ of vicinal growth destabilized by an inverted Ehrlich-Schwoebel effect.

Dimensional Analysis of the continuum equation

The PTVV equation was proposed for first time and analyzed using DA by Pimpinelli et al.¹. Here we will use the modified version of the equation as proposed later by Krug et al.²:

$$\frac{\partial h}{\partial t} + \frac{\partial}{\partial x} \left(K_1 m^\rho + \frac{K_2}{m^k} \frac{\partial^2}{\partial x^2} m^n \right) = \text{const} \quad (\text{A2})$$

where h is the (crystal) surface height, x is the spatial coordinate, K_1 and K_2 are material parameters that contain the details of the concrete destabilization mechanism and the magnitude of the step-step repulsion, respectively, $m \equiv (\partial h / \partial x)$ is the surface slope, ρ generalizes the destabilizing role of the step kinetics/diffusion asymmetry, n is the power of the step-step distance d in the step-step

repulsion law, $U \sim 1/d^n$, and k is triggering between the surface diffusion limited (DL) and attachment/detachment at steps (kinetics) limited (KL) regimes driving the time evolution. When introduced in the PTVV equation², it was postulated that k acquires the values of 0(1) for DL(KL) regime(s). We will show below based on numerical analysis of Liu and Weeks (LW) model³ which is introduced for the KL regime followed by same type study of the Sato and Uwaha model⁴ (both models are formulated in terms of ODE-systems, but the latter permits by varying the model parameters to reproduce both DL and KL regimes) that k is 1(2) for DL(KL) regime(s).

While PTVV assume a power-law dependence between the two perpendicular length-scales (along x and y) that eventually enter the model thus accounting for the model's self-affinity, we apply here another approach in the DA^{5, 6} assuming only that the dimensions along the two axis are different and introduce three (arbitrary) scales, the two perpendicular length-scales along x and y , and the time-scale:

$$[\xi_y] = L_y; [\xi_x] = L_x; [\tau] = T \quad (A3)$$

Further we introduce the dimensionless variables $H \equiv h/\xi_y; X \equiv x/\xi_x; T \equiv t/\tau$ and plug them into (A2):

$$\frac{\xi_y}{\tau} \frac{\partial H}{\partial T} + \frac{1}{\xi_x} \frac{\partial}{\partial X} \left(K_1 (\xi_y \xi_x^{-1})^\rho M^\rho + (\xi_y \xi_x^{-1})^{-k} \xi_x^{-2} \frac{K_2}{M^k} \frac{\partial^2}{\partial X^2} (\xi_y \xi_x^{-1})^n M^n \right) = \text{const} \quad (A4)$$

Now, if the initial equation obeys dimensional homogeneity, after dividing all terms by ξ_y/τ one will arrive at a dimensionless equation:

$$\frac{\partial H}{\partial T} + \frac{\partial}{\partial X} \left(\frac{\tau}{\xi_y \xi_x} K_1 (\xi_y \xi_x^{-1})^\rho M^\rho + K_2 \frac{\tau}{\xi_y \xi_x} (\xi_y \xi_x^{-1})^{-k} \xi_x^{-2} (\xi_y \xi_x^{-1})^n \frac{1}{M^k} \frac{\partial^2}{\partial X^2} M^n \right) = \text{const}_1 \quad (A5)$$

Since the three scales defined in (A3) were arbitrary we can set the two remaining coefficients containing parameters and scales (in front of the second and of the third term of the left hand side of (A5)) equal to 1 thus obtaining a system of two equations for the three scales:

$$\frac{\tau}{\xi_y \xi_x} K_1 (\xi_y \xi_x^{-1})^\rho = 1 \quad (A6)$$

$$K_2 \frac{\tau}{\xi_y \xi_x} (\xi_y \xi_x^{-1})^{-k} \xi_x^{-2} (\xi_y \xi_x^{-1})^n = 1 \quad (A7)$$

and a dimensionless equation without parameters:

$$\frac{\partial H}{\partial T} + \frac{\partial}{\partial X} \left(M^\rho + \frac{1}{M^k} \frac{\partial^2}{\partial X^2} M^n \right) = \text{const}_1 \quad (A8)$$

that could be in principle solved numerically.

By dividing the two equations, (A6) and (A7), to eliminate the time-scale, we get the relation

between the two length-scales:

$$\xi_y = \frac{K_1}{K_2} \xi_x^{\frac{2+(n-k)-\rho}{(n-k)-\rho}} \quad (\text{A9})$$

and defining the exponent α as:

$$\alpha = \frac{2+(n-k)-\rho}{(n-k)-\rho} \quad (\text{A10})$$

A brief inspection of the just obtained expression (A10) shows that the original equation does not account for the trivial case $\alpha \equiv 1$ for any finite value of n , therefore it describes a self-affine (B2-type¹⁵) and not a self-similar (B1-type¹⁵) time evolution. Now we obtain the connection between one of the length-scales and the time scale using (A6) and restricting ourselves to the universality class $\rho = -1$:

$$\xi_y = (K_1 \tau)^{1/2}; \beta = 1/2 \quad (\text{A11})$$

being independent of the values of n and k as observed yet by Liu and Weeks¹¹ and discussed later in detail². This result shows further that not only the scaling exponent but also the pre-factor is independent of n and k .

Further, combining (A9) and (A11), one obtains:

$$\xi_x = \left[\left(\frac{K_2}{K_1} \right)^2 (K_1 \tau) \right]^{\frac{n-k+1}{2(n-k+3)}}; 1/z = \frac{n-k+1}{2(n-k+3)} \quad (\text{A12})$$

Numerical analysis of the Liu and Weeks (LW) model

Here we extend our numerical study of the model of Liu and Weeks (LW)³. In their theoretical study of the sublimation Si(111) vicinal crystals controlled by the slow attachment/detachment rate of the adatoms to/from the steps they deduce a BCF-type equation (with non-transparent steps) for the velocity of a step in the step train:

$$\frac{dx_i}{dt} = (K_- \Delta x_i + K_+ \Delta x_{i+1}) + \frac{Kc_0\Omega}{2kT} n g h^{n+1} \Omega \left[3 \left(\frac{1}{\Delta x_i^{n+1}} - \frac{1}{\Delta x_{i+1}^{n+1}} \right) + \frac{1}{\Delta x_{i+2}^{n+1}} - \frac{1}{\Delta x_{i-1}^{n+1}} \right] \quad (\text{A13})$$

The first term is linear in the terrace widths $\Delta x_i \equiv x_i - x_{i-1}$ and setting $K_- > K_+$ leads to

destabilization of the initially uniform step train, $K_{\pm} \equiv \pm \frac{Kc_0\Omega F}{2kT} + \frac{1}{2\tau_e}$, where K is the

attachment/detachment rate, F is the electromigration force, only when negative it destabilizes a sublimating vicinal surface descending to the left, i.e step-down (SD), c_0 is the equilibrium concentration of the adatoms and τ_e is the average time the adatoms spend on the surface before

evaporating into the ambience, Ω is the area occupied by an adatom. The second term comes from taking into account⁷ the omnipresent step-step repulsions with magnitude of the repulsion energy g and h is the height of a monoatomic step, the canonical value of the step-step repulsions exponent n is 2. The coefficients K_{\pm} are simplified further by introducing the dimensionless quantity

$$b \equiv -\frac{Kc_0\Omega F\tau_e}{kT} :$$

$$K_{\pm} \equiv \pm \frac{Kc_e\Omega F}{2kT} + \frac{1}{2\tau_e} = \frac{1}{\tau_e} \left(\frac{\pm Kc_e\Omega F/kT + 1}{2} \right) \equiv \frac{1}{\tau_e} \left(\frac{mb + 1}{2} \right) \quad (A14)$$

to arrive at:

$$\frac{dx_i}{dt} = \frac{1+b}{2\tau_e} \left(\frac{1-b}{1+b} \Delta x_{i+1} + \Delta x_i \right) + u \left[3 \left(\frac{1}{\Delta x_i^{n+1}} - \frac{1}{\Delta x_{i+1}^{n+1}} \right) + \frac{1}{\Delta x_{i+2}^{n+1}} - \frac{1}{\Delta x_{i-1}^{n+1}} \right] \quad (A15)$$

where $u \equiv \frac{Kc_0\Omega}{2kT} ngh^{n+1}\Omega$. Now we non-dimensionalize (scale) eq. (A15) by introducing the

dimensionless coordinate and time $X \equiv \frac{x}{\xi}$ and $T \equiv \frac{t}{\tau}$:

$$\frac{dX_i}{dT} = \frac{\tau(1+b)}{2\tau_e} (\Delta X_i + B\Delta X_{i+1}) + \frac{\tau}{\xi^{n+2}} u \left[3 \left(\frac{1}{\Delta X_i^{n+1}} - \frac{1}{\Delta X_{i+1}^{n+1}} \right) + \frac{1}{\Delta X_{i+2}^{n+1}} - \frac{1}{\Delta X_{i-1}^{n+1}} \right] \quad (A16)$$

where $B \equiv (1-b)/(1+b)$. Since the scales for length and time ξ and τ are arbitrary we can set the coefficients in front of the two terms on the right hand side of (A16) equal to unity to obtain:

$$\frac{\tau(1+b)}{2\tau_e} = 1 \Rightarrow \tau \equiv \frac{2\tau_e}{(1+b)} \quad (A17)$$

$$\frac{\tau}{\xi^{n+2}} u = 1 \Rightarrow \xi \equiv \left[\frac{2\tau_e}{(1+b)} u \right]^{1/(n+2)} \quad (A18)$$

Thus the dimensionless version of the equation of LW model is obtained as:

$$\frac{dX_i}{dT} = (\Delta X_i + B\Delta X_{i+1}) + \left[3 \left(\frac{1}{\Delta X_i^{n+1}} - \frac{1}{\Delta X_{i+1}^{n+1}} \right) + \frac{1}{\Delta X_{i+2}^{n+1}} - \frac{1}{\Delta X_{i-1}^{n+1}} \right] \quad (A19)$$

Together with the system of equations (A13) is defined the initial vicinal distance $\Delta x_i = l_0$, one for the all values of i . It is non-dimensionalized to:

$$L_0 \equiv \left[\frac{2\tau_e}{(1+b)} u \right]^{-1/(n+2)} l_0 \quad (A20)$$

More details, and especially how the procedure above can be used to find the scaling pre-factors, can be found in ⁸.

In its classical formulation, where l_0 and τ_e are used as scales for non-dimensionalization, the equation of LW model is written as:

$$dx_i/dt = \Delta x_i(1+b)/2 + \Delta x_{i+1}(1-b)/2 + u' \left[3(\Delta x_i^{-(n+1)} - \Delta x_{i+1}^{-(n+1)}) + \Delta x_{i+2}^{-(n+1)} - \Delta x_{i-1}^{-(n+1)} \right] \quad (\text{A21})$$

where $u' = u\tau_e l_0^{n+2}$ and, hence, the connection between the two variants of non-dimensionalization is given by:

$$L_0 = \left[\frac{2u'}{(1+b)} \right]^{-1/(n+2)} \quad (\text{A22})$$

and

$$B = \frac{1-b}{1+b} \quad (\text{A23})$$

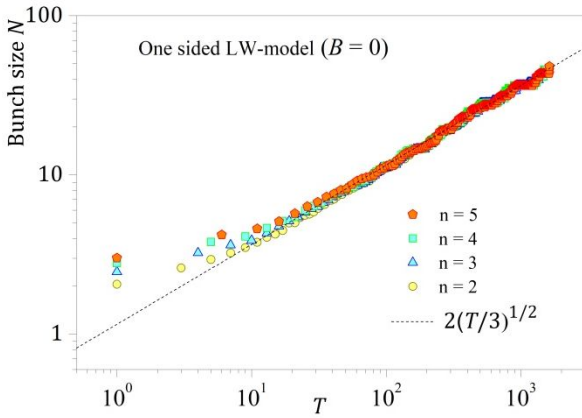


Figure S1 Number of steps in the bunch N as function of the rescaled time T for the one-sided case, $B=0$, of the LW model, the values of L_0 are given in the right panel, 3000 steps are used in this run.

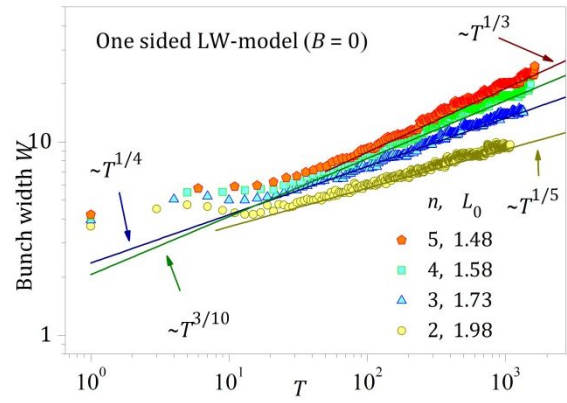


Figure S2 Bunch width W as function of the rescaled time T . It is the bunch width that distinguishes in between the values of the step-step repulsion exponent n , see (A12).

After being introduced, the LW model was studied further ^{2, 9-11} mainly with focus on the scaling relation between the minimum

distance in the bunch l_{\min} (maximal slope) and the number of steps in the bunch, $l_{\min} \sim N^{-\gamma}$. Unfortunately, the size scaling exponent γ cannot distinguish between the DL and KL regime of the instability, as shown by Krug et al. ². The numerical pre-factor in the time-scaling of the bunch size N was calculated¹⁰ for the case of $b=1$ ($B=0$) and, independently of the parameters used, the value of $\sqrt{1.5}=1.225$ was reported but only the parallel study of LW and MM reveals that it is $2/\sqrt{3}=1.1547$. Krug et al. ² studied both the time-scaling of N and the size-scaling of W to obtain

α as defined in (A10) but were unable to attribute the LW model to a universality class. No systematic parallel study of the time-scaling of the bunch width W and bunch size N within the LW model was attempted so far although experimental results are reported¹²⁻¹⁴ but in experiments one cannot vary the step-step repulsions exponent n . Here we study in parallel the two length-scales, bunch size N and bunch width W , necessary to describe thoroughly^{1, 15} the self-affine pattern evolution in the intermediate regime⁶ of the instability by using a unified monitoring protocol¹⁶. Thus we obtain the time-scaling exponents of the bunch size N and of the bunch width W by changing systematically the value of the step-step repulsions exponent n , see Figure S1 and Figure S2 (the values of L_0 are shown on the right one only).

Numerical study of the Model of Sato-Uwaha for unstable vicinal growth

The equations for step motion for the case of vicinal growth destabilized by inverse Ehrlich-Shwoebel effect (Gr+iSE) are first deduced and studied by Sato and Uwaha⁴. They integrate twice a stationary diffusion equation on a single terrace between two steps that contains only two terms – the usual diffusional one and the flux F while the effect of the step-step repulsion with a magnitude A and range n , $U = A/d^n$, d being the step-step distance, enters the boundary conditions in the usual form¹⁷. First we re-write them⁴ in a more familiar form:

$$\begin{aligned} \frac{dx_i}{dt} = & \frac{\Omega D_s}{d_+ + d_- + \Delta x_{i+1}} \left(\frac{F}{2D_s} \Delta x_{i+1}^2 + d_- \frac{F}{D_s} \Delta x_{i+1} \right) + \frac{n\Omega^2 A D_s c_e^0}{k_B T (d_+ + d_- + \Delta x_{i+1})} \left(\frac{1}{\Delta x_{i+2}^{n+1}} - \frac{2}{\Delta x_{i+1}^{n+1}} + \frac{1}{\Delta x_i^{n+1}} \right) + \\ & \frac{\Omega D_s}{d_+ + d_- + \Delta x_i} \left(\frac{F}{2D_s} \Delta x_i^2 + d_+ \frac{F}{D_s} \Delta x_i \right) + \frac{n\Omega^2 A D_s c_e^0}{k_B T (d_+ + d_- + \Delta x_i)} \left(\frac{1}{\Delta x_{i+1}^{n+1}} - \frac{2}{\Delta x_i^{n+1}} + \frac{1}{\Delta x_{i-1}^{n+1}} \right) \end{aligned} \quad (A24)$$

where the asymmetry of the adatom-to-step attachment is reflected by the different kinetic length-scales $d_{\pm} \equiv D_s/K_{\pm}$, K_+ (K_-) is the kinetic coefficient for adatom-to-step attachment from the lower (upper) and the step motion during vicinal growth is unstable when $K_- > K_+$ (easier attachment from the upper terrace). Now, after introducing the dimensionless variables $X_i \equiv \frac{x_i}{\xi}$; $T \equiv \frac{t}{\tau}$ and plugging them into (A24) we obtain:

$$\begin{aligned} \frac{dX_i}{dT} = & \frac{F\Omega\tau}{D_+ + D_- + \Delta X_{i+1}} \left(\frac{\Delta X_{i+1}^2}{2} + D_- \Delta X_{i+1} \right) + \frac{\tau n\Omega^2 A D_s c_e^0}{\xi^{n+3} k_B T (D_+ + D_- + \Delta X_{i+1})} \left(\frac{1}{\Delta X_{i+2}^{n+1}} - \frac{2}{\Delta X_{i+1}^{n+1}} + \frac{1}{\Delta X_i^{n+1}} \right) + \\ & \frac{F\Omega\tau}{D_+ + D_- + \Delta X_i} \left(\frac{\Delta X_i^2}{2} + D_+ \Delta X_i \right) + \frac{\tau n\Omega^2 A D_s c_e^0}{\xi^{n+3} k_B T (D_+ + D_- + \Delta X_i)} \left(\frac{1}{\Delta X_{i+1}^{n+1}} - \frac{2}{\Delta X_i^{n+1}} + \frac{1}{\Delta X_{i-1}^{n+1}} \right) \end{aligned} \quad (A25)$$

Thus in front of the two different types of terms we have two combinations of parameters that both can be set equal to 1 since the time- and length-scales were arbitrary:

$$F\Omega\tau = 1 \Rightarrow \tau = (F\Omega)^{-1}$$

$$\frac{\tau n \Omega^2 A D_s c_e^0}{\xi^{n+3} k_B T} = 1 \Rightarrow \xi = \left(\frac{n \Omega A D_s c_e^0}{k_B T F} \right)^{\frac{1}{n+3}}$$

and what is remaining from (A25) is:

$$\begin{aligned} \frac{dX_i}{dT} = & \frac{\left(\frac{\Delta X_{i+1}^2}{2} + D_- \Delta X_{i+1} \right) + \left(\frac{1}{\Delta X_{i+2}^{n+1}} - \frac{2}{\Delta X_{i+1}^{n+1}} + \frac{1}{\Delta X_i^{n+1}} \right)}{D_+ + D_- + \Delta X_{i+1}} + \\ & \frac{\left(\frac{\Delta X_i^2}{2} + D_+ \Delta X_i \right) - \left(\frac{1}{\Delta X_{i+1}^{n+1}} - \frac{2}{\Delta X_i^{n+1}} + \frac{1}{\Delta X_{i-1}^{n+1}} \right)}{D_+ + D_- + \Delta X_i} \end{aligned} \quad (\text{A26})$$

Thus, only three parameters remain – $D_- \equiv \frac{d_-}{\xi} \equiv \frac{D_s}{K_-} \frac{1}{\xi}$, $D_+ \equiv \frac{d_+}{\xi} \equiv \frac{D_s}{K_+} \frac{1}{\xi}$ and the dimensionless initial vicinal distance $L_0 \equiv l_0/\xi$ instead of the four as in the Sato and Uwaha's version. Note that in this form of the equations, (A26), the value of the flux F is hidden and enters only in the parameters used for non-dimensionalization. Let us re-write the new parameters in terms of the “old” ones:

$$L_0 \equiv \frac{l_0}{\xi} = \frac{1}{\left(\frac{D_s c_e^0}{F l_0^2} \frac{n \Omega A}{k_B T l_0^{n+1}} \right)^{\frac{1}{n+3}}}$$

and we see that namely L_0 is combination of two of the “old” parameters. For the parameter values studied by Sato and Uwaha L_0 acquires the value of $L_0 = \frac{1}{(4 * 0.4)^{\frac{1}{2+3}}} = 0.91$ while the other two

parameters, D_+ and D_- , are:

$$D_- = \frac{d_-}{\xi} = \frac{d_-}{l_0} \frac{l_0}{\xi} = \frac{d_-}{l_0} L_0 = 0.02 * 0.91 = 0.0182$$

$$D_+ = \frac{d_+}{\xi} = \frac{d_+}{l_0} \frac{l_0}{\xi} = 200 * 0.91 = 182$$

Here we use for the kinetics limited (KL) version of the phenomenon $D_+ = 517, 481, 456$, $D_- = 5.17, 4.81, 4.56$ and $L_0 = 0.517, 0.481, 0.456$ for $n = 2, 3, 4$ correspondingly. For the diffusion-limited (DL) version we use $D_+ = 151.6, 197.2, 220.8$, $D_- = 0.015, 0.02, 0.022$, $L_0 = 0.758, 0.986, 1.104$ for $n = 2, 3, 4$ correspondingly.

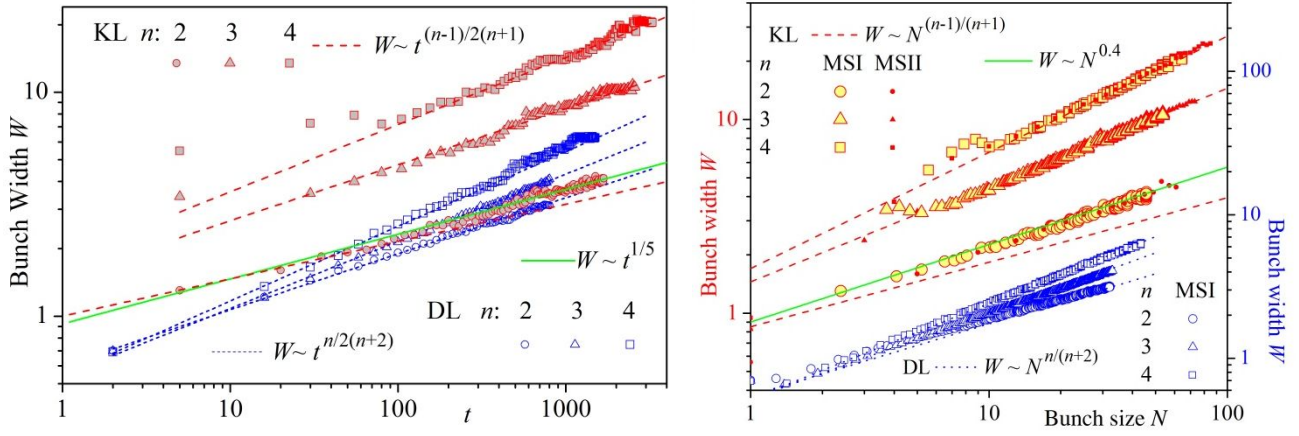


Figure S3 Results from the Sato and Uwaha (Gr+iSE) model for the time dependence of the bunch width (left) and for the scaling between the two spatial length-scales – bunch width and bunch height (right panel). Note the deviation from the predictions, (A12) and (A10), in the KL regime with $n=2$, same as in the LW-model, Figure S2.

Universality classes in Step Bunching

The parallel study of the time-scaling of bunch size N and bunch width W in the one-sided version of the LW model as illustrated in Figure S1 and Figure S2 strongly supports the attribution of the LW model to the $\rho=-1$ universality class (although one could argue that also the universality class of the “C+ - C-“ is possible based only on the value of β) but with the correction coefficient k as introduced by Krug et al.² should have the value of 2 for this, KL regime of the instability. The model of Sato and Uwaha, from another side, permits to study also the DL regime of the instability, see Figure S3, and obtain the values of $k=2$ (1) for KL (DL) regime with the notable exception for the case of $n=2$ in the KL regime – $\alpha=0.4$ instead of $1/3$ and $1/z = 1/5$ instead of $1/6$, same as obtained already for the LW model, Figure S2.

Note that the experimental findings in¹³, $\beta=1/2$, $1/z=1/4$, could be attributed both to the $\rho=-1$ universality class from the PTVV¹⁴ scheme and to the “C+ - C-“ universality class with n formally equal to 1 (without the correction k for PTVV), therefore, assuming that $n=2$ in real systems, the experimental system studied in¹³, Si homoepitaxy, is probably in the DL growth mode, $k=1$. Same set of exponents was reproduced by Omi et al.¹⁴ in growth experiments on Si(111)-(7x7) at 750°C. From the table below (and from the considerations above) are excluded the exponents connected with the minimal distance in the bunch l_{\min} since it was shown that γ , the size scaling exponent of l_{\min} in $l_{\min} \sim N^{-\gamma}$ cannot distinguish between the DL and KL regimes of the instability (for a recent review see¹⁹).

	C ⁺ - C ⁻ (DL) ¹⁶	Universality Classes PTVV ¹		Minimal model* (MM)
General <i>n</i>				
α ($W \sim N^{1/\alpha}$)	$(n+1)/n$	$(2+n-\rho)/(n-\rho)$		$(n+1)/n$
$\beta=\alpha/z$ ($N \sim t^\beta$)	1/2	$(2+n-\rho)/[2(n+1-2\rho)]$		1
z ($W \sim t^{1/z}$)	$2(n+1)/n$	$2(n+1-2\rho)/(n-\rho)$		$(n+1)/n$
<i>n=1</i>		$\rho = -2$	$\rho = -1$	
α ($W \sim N^{1/\alpha}$)	2	5/3	2	2
$\beta=\alpha/z$ ($N \sim t^\beta$)	1/2	5/12	1/2	1
z ($W \sim t^{1/z}$)	4	4	4	2
<i>n=2</i>		$\rho = -2$	$\rho = -1$	
α ($W \sim N^{1/\alpha}$)	3/2	3/2	5/3	3/2
$\beta=\alpha/z$ ($N \sim t^\beta$)	1/2	3/7	1/2	1
z ($W \sim t^{1/z}$)	3	7/2	10/3	3/2
<i>n=3</i>		$\rho = -2$	$\rho = -1$	
α ($W \sim N^{1/\alpha}$)	4/3	7/5	3/2	4/3
$\beta=\alpha/z$ ($N \sim t^\beta$)	1/2	7/16	1/2	1
z ($W \sim t^{1/z}$)	8/3	16/5	12/4	4/3

* when the destabilizing term is linear in the widths of the two adjacent to the step terraces as in the LW model, the numerical values are shifted systematically as compared to ¹⁸ because of the different definition of the repulsive term.

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