Supporting information

Extended model for anisotropic spiral growth

In the main text we assumed that a spiral step starts to propagate at a constant rate after reaching its critical length, $d_{c,i}$. However, as pointed out in an early paper by Budevski et al. [SI-1], this is not entirely correct. The rate of a propagating step, $v_{st,i}$, of a polygonized growth spiral is namely determined by its effective supersaturation, $\Delta \mu_{eff}$,

$$v_{st,i} = \beta_i [\exp(\Delta \mu_{eff} / kT) - 1], \quad \text{(SI-1)}$$

which again is a function of its length, $l$. Here $\beta_i$ is the kinetic constant for step propagation, $kT$ has the usual meaning and the step length $l$ is defined in figure 4 of the main text. An expression for $\Delta \mu_{eff}$ follows from equation (5), which expresses the change in Gibbs energy, $\Delta G$ upon advancement of a step with length, $l$, over a distance $\delta$. Starting from this equation for step 2 in the main text

$$\Delta G = (\gamma_1 + \gamma_3) \delta - \frac{\Delta \mu_{eff} \delta h_{st}}{\Omega}, \quad \text{(5)}$$

and expressing $\delta$ in terms of the number of growth units, $N$, added to the crystal surface

$$\delta = \frac{N \Omega}{lh_{st}}, \quad \text{(SI-2)}$$

gives

$$\Delta G(N) = (\gamma_1 + \gamma_3) \frac{N \Omega}{lh_{st}} - \Delta \mu N. \quad \text{(SI-3)}$$

Differentiation of (SI-3) with respect to $N$ gives the effective supersaturation for growth of a spiral step as a function of its length

$$\Delta \mu_{eff}(l) = -\frac{\partial \Delta G(N)}{\partial N} = \Delta \mu - \frac{(\gamma_1 + \gamma_3) \Omega}{lh_{st}}. \quad \text{(SI-4)}$$

Rewriting and using expression (7) in the main text for the critical step length,

$$d_{c,2} = \frac{(\gamma_1 + \gamma_3) \Omega}{\Delta \mu h_{st}} \quad \text{(7)}$$

leads to
\[ \Delta \mu_{\text{eff},i} = \Delta \mu \left[ 1 - \frac{d_{c,i}}{l} \right]. \quad (\text{SI-5}) \]

The same holds for the other steps

\[ \Delta \mu_{\text{eff},i} = \Delta \mu \left[ 1 - \frac{d_{c,i}}{l} \right]. \quad (\text{SI-6}) \]

Combination of (SI-1) and (SI-6) gives the step velocity of the different spiral steps as a function on their –finite- length, \( l \)

\[ v_{a,i} = \beta_i \left[ \exp \left( \frac{\Delta \mu}{kT} \left[ 1 - \frac{d_{c,i}}{l} \right] \right) - 1 \right]. \quad (\text{SI-7}) \]

**Figure SI-1.** Calculation of the pattern of an anisotropic, polygonized spiral by numerical integration. The figure shows the successive stages of spiral formation mapped at constant time intervals. The first step, 1, proceeds at a constant rate; the steps that follow propagate at a rate that depends on their width. It can be recognized that the steps close to the dislocation outcrop propagate slower.

In contrast to the case of a constant step velocity for step length \( l > d_{c,i} \), in the present case no analytical solution can be obtained and numerical integration is needed to calculate the spiral pattern (figure SI-1). In this we start with one straight step, \( i = 1 \), ending at the emergence point of the dislocation at the crystal surface. This step of “infinite” length
advances with constant velocity $v_{st,1} = \beta_1[\exp(\Delta\mu/kT) - 1]$. During each infinitesimal time increment, $\Delta t$, the step moves forward over a distance $v_{st,1}\Delta t$. In this period step 2 forms at the dislocation outcrop; if this step reaches a length $l = d_{c,2}$, it starts to propagate as well. In first instance slowly, but its rate increases as its length increases, following equation (SI-7). From the moment that step 2 advances, step 3 is formed, which remains immobile until its length exceeds $d_{c,3}$. By continuing this numerical integration, considering the length dependent step velocity finally a stationary spiral pattern is obtained, of which the step spacing at the centre is somewhat less than further away.

Using the four different step distances, $w_i$, obtained from experiment and the step energies calculated using STEPLIFT [SI-2, SI-3] as summarized in table SI-1 and a program written in MATLAB code [SI-4], we come to the spiral shown in figure SI-2. In our model we assumed that the kinetic constants $\beta_i$ are proportional to $w_i$, as these step distances are measured at larger distances from the spiral centre. The spiral shape itself is independent of the absolute values of $\beta_i$, it only depends on the relative values of this parameter. By applying a supersaturation of $\Delta\mu/kT = 0.40$ in the model, we come to the observed values of $w_i$. This value is very close to the value of 0.42, deduced from the Monte Carlo simulation.

![Figure SI-2](image.png)

**Figure SI-2.** Calculated spiral pattern using the parameters given in Table SI-1. The supersaturation is kept at $\Delta\mu/kT = 0.40$. 

SI-3
Table SI-1. Parameters used for the calculation of the spiral shown in figure SI-2

<table>
<thead>
<tr>
<th></th>
<th>Step 1</th>
<th>Step 2</th>
<th>Step 3</th>
<th>Step 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Observed step distances (µm)</td>
<td>--</td>
<td>0.16</td>
<td>1.72</td>
<td>0.23</td>
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<tr>
<td>Relative values kinetic constant: ( w_j/w_1 )</td>
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<td>1.0</td>
<td>10.8</td>
<td>1.44</td>
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<tr>
<td>Step energies (kcal/mol.Å)</td>
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<td>3.40</td>
<td>2.29</td>
</tr>
<tr>
<td>Volume growth unit, ( \Omega (Å^3) )</td>
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<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Step height (Å)</td>
<td>12.5</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
</tbody>
</table>

References


