**Abstract**

Existing theories describing faceted growth and ablation of ice crystals have significant limitations in terms of the connection to underlying physical processes. In filling that knowledge gap, the present model provides insights and predictive power about faceted growth and ablation that have not been possible previously. For example, the model exhibits Turing-like pattern behavior, in the sense that the horizontal distance between successive ice layers atop a growing facet is predicted to increase in proportion to the square root of the surface diffusivity.

1. **Prior theories of faceted ice crystal growth and ablation**

The BCF picture is clearly wrong between 240 K and melting, in that it assumes deposition atop a crystalline surface, whereas in reality the surface of real ice is covered by a quasi-liquid layer (QLL) whose efficiency at capturing incoming water vapor molecules is close to 100%.

The quasi-liquid continuum model introduced by some of the authors in 2016 (N2016) recasts the problem as an ice surface represented by two mesoscale variables (see Fig. 1). Variable represents the total thickness of the ice surface, while variable represents the thickness of the quasi-liquid part of . N2016 represents the dynamics of these processes in the form of two coupled differential equations, one for and one for . (A third variable may be computed from these: .) Key atomistic processes incorporated in N2016 were: (i) vapor deposition and ablation, (ii) surface diffusion of the quasi-liquid across the facet, and (iii) the interconversion of quasi-liquid and ice.

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| **Figure 1**. Visual representation of mesoscale variables , , and , and processes affecting them, in the N2016 (and present) model. Dashed arrows represent processes affecting how these variables evolve over time. |

A key advancement of the N2016 model over prior theories of crystal growth is that runs (“trajectories”) of solutions to the differential equations constituting the model exhibited unbound growth at facet corners (i.e., dendritic growth associated with snowflake formation), as well as faceted growth (a pattern of steady-state growth across an entire facet. The mechanism by which the latter occurred was termed “diffusive slowdown,” in which excess deposition of water vapor at facet corners is compensated by an emergent surface morphology change.

N2016 suffered from several limitations, however, of which the most important for our present purpose is that the time scale embedded in process (iii), the interconversion of quasi-liquid and ice, was fixed relative to processes (i) and (ii); in real crystal facets, these relative time scales may vary from facet to facet, or as a function of temperature. The revised model corrects this deficiency, as described below.

1. **A revised quasi-liquid continuum model**

The model introduced here is defined by

(1a)

(1b)

Some notes about this model:

* represents the idea that surface diffusion depends on the thickness of the quasi-liquid only; the underlying ice is considered immobile on time scales considered here.
* is the rate of exchange of water between the facet and the vapor phase (i.e. deposition and ablation).
* is the fractional difference between the rate of ablation and that of deposition (i.e., the surface supersaturation), given by … relationship to and .
* defines the thickness of quasi-liquid when it is in equilibrium with the underlying ice. Here (as in N2016) we use the sinusoidal form

(2)

* is a first-order relaxation constant describing the time scale at which quasi-liquid/ice equilibrium is achieved. That is, if we imagine an initial situation having an amount of quasi-liquid given by , then equilibration after a time occurs according to

(3)

* If one takes the time derivative of Eq. (3), and assumes that is small, the second term on the right-hand side of Eq. (1b) results.

The difference between the present model and N2016, therefore, lies in the treatment of the quasi-liquid equilibration just described, i.e., the use of Eq. (1b) rather than Eq. (5b) of N2016 for the time evolution of the quasi-liquid layer.

With this revision, we are able to parameterize the rate of quasi-liquid/ice equilibration relative to processes (i) and (ii). Specifying a small value for , for example, would represent the idea that quasi-liquid/ice equilibration is fast compared to diffusion and exchanges with the vapor phase, while large would mean the opposite. We do not have reliable independent guides for determining , but we do have a guidepost: because the “diffusive slowdown” mechanism for stabilization of faceted ice growth described in N2016 required that quasi-liquid/ice equilibration be slow compared to surface diffusion, we should not be surprised if we find that large leads to stable growth scenarios. We return to this topic below.

1. **Informing theory with SEM experiments**

Also motivating a revised theory is the fact that new information is available from mesoscale experiments – specifically, the ability to construct quantitative surface morphology of hexagonal ice crystals grown and imaged in the chamber of a scanning electron microscope. We will refer to this process as “SEM/GNBF retrieval.”

1. **Questions addressed in this paper**

The premise of this paper is to explore predictions of the model embodied by Eqs. (1a-b), hand in hand with observations of growing and ablating hexagonal ice crystals at the mesoscale made by SEM/GNBF retrieval of surfaces morphology of those ice crystals. Questions we seek answers to include:

1. *Existence of faceted ablation*. Is there such a thing as faceted ablation, and if so, does the model support such a phenomenon?
2. *Dependence of surface morphology on supersaturation, diffusion coefficient, and other parameters.* Is it possible to make general statements about how surface morphology varies as a function of these parameters?
3. *Facet concavity and convexity*. What does concavity of convexity of a facet tell us about the conditions surrounding a crystal?
4. *Resilience*. SEM observations show that faceting is resilient in the sense that a crystal with rough surface morphology can be restored to smooth morphology when exposed to highly supersaturated conditions. Is the model similarly resilient?
5. *Does facet roughness have an intrinsic characteristic length scale*? More particularly, is there a difference between the roughness that appears under supersaturated conditions, vs subsaturated conditions, and if so, does the model help us understand that difference?
6. *What about differences between facets*?
7. **Implementation details**

Python, accelerated with Numby. Surface morphology extraction using the GNBF formalism …

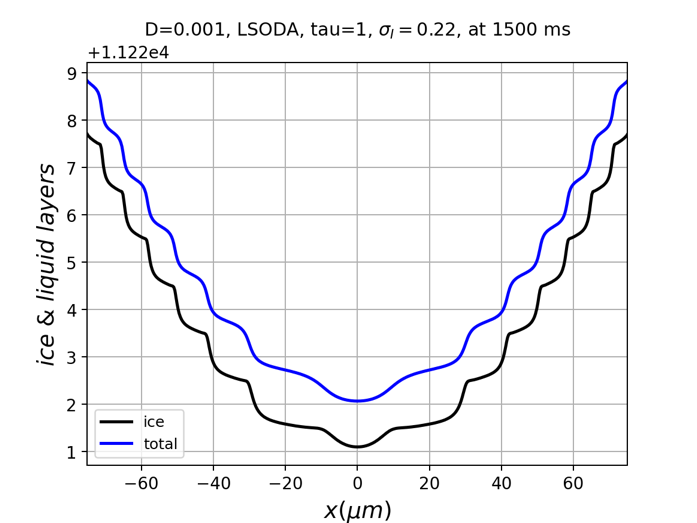
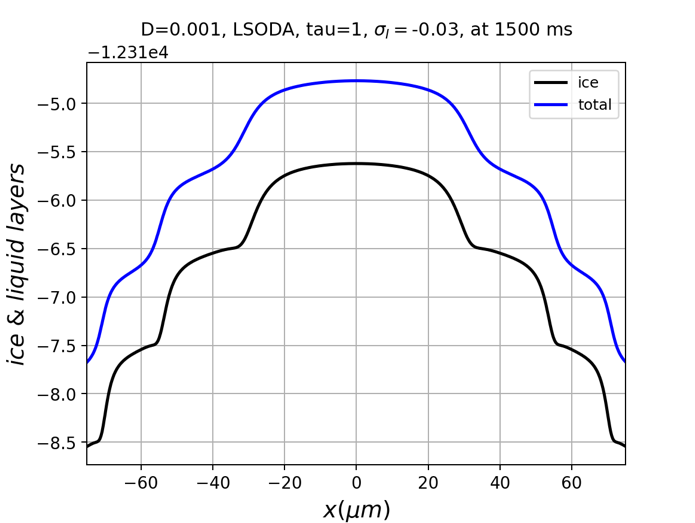
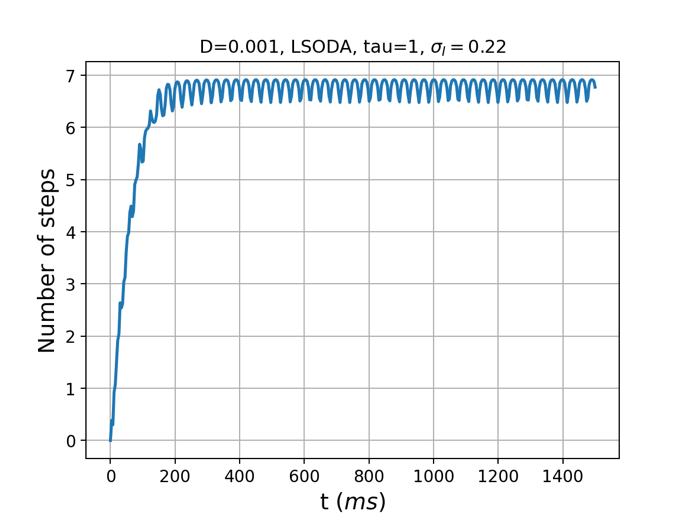
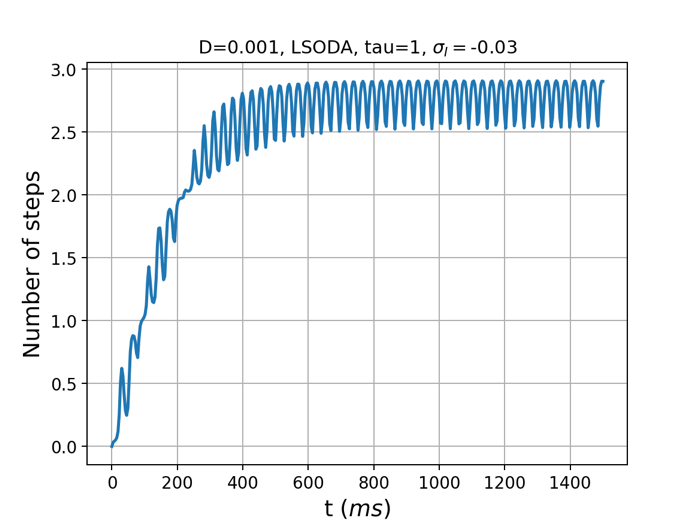
Figure 2 displays supersaturation profiles …

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| **Figure 2**. Supersaturation and subsaturation profiles. |

1. **Results**

*I. Existence of faceted ablation*

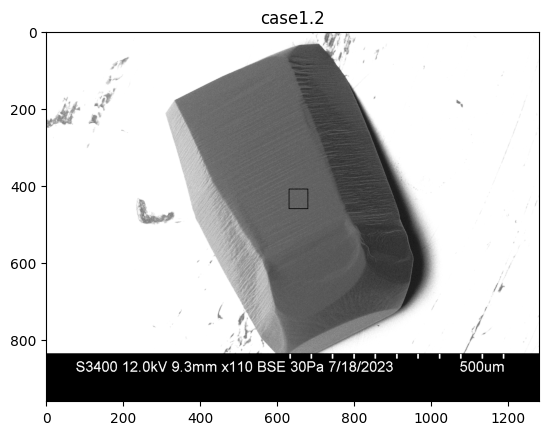
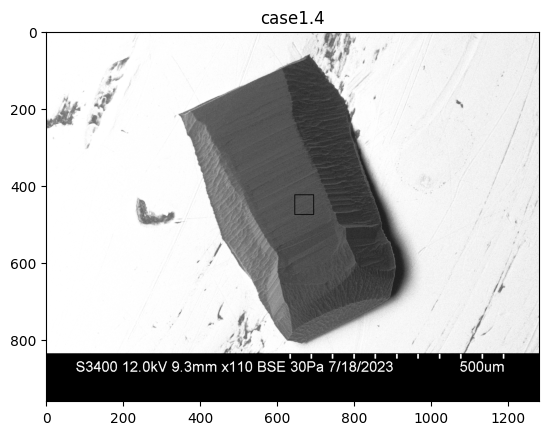
Figure 3 shows a modeled ice crystal surface under ablating (left) and growing (right) conditions. Focusing first on the ablation scenario, we see that the facet achieves a steady-state profile – i.e., faceted ablation – after about . The center of the crystal is about three molecular layers thicker than at facet boundaries. Turning to the growth scenario on the right, we see that the facet also achieves steady state, with the facet center about seven molecular layers thinner than at facet boundaries.



**Figure 3**. Stabilization of faceted ablation (left panels) and faceted growth (right panels).

Figure 4 displays SEM images of an ice crystal under ablating and growing conditions. Since the ablating crystal retains its faceted structure, we can conclude that faceted ablation does indeed occur. The figure shows, moreover, that faceted ablation occurs even when the surface is rough (e.g., the prismatic facets in the figure) as well as when the surface is smooth (basal facet).

In fact, we observe faceted ablation quite frequently in SEM images of ablating ice crystals.



**Figure 4**. An ice crystal under ablating (left) and growing (right) conditions.

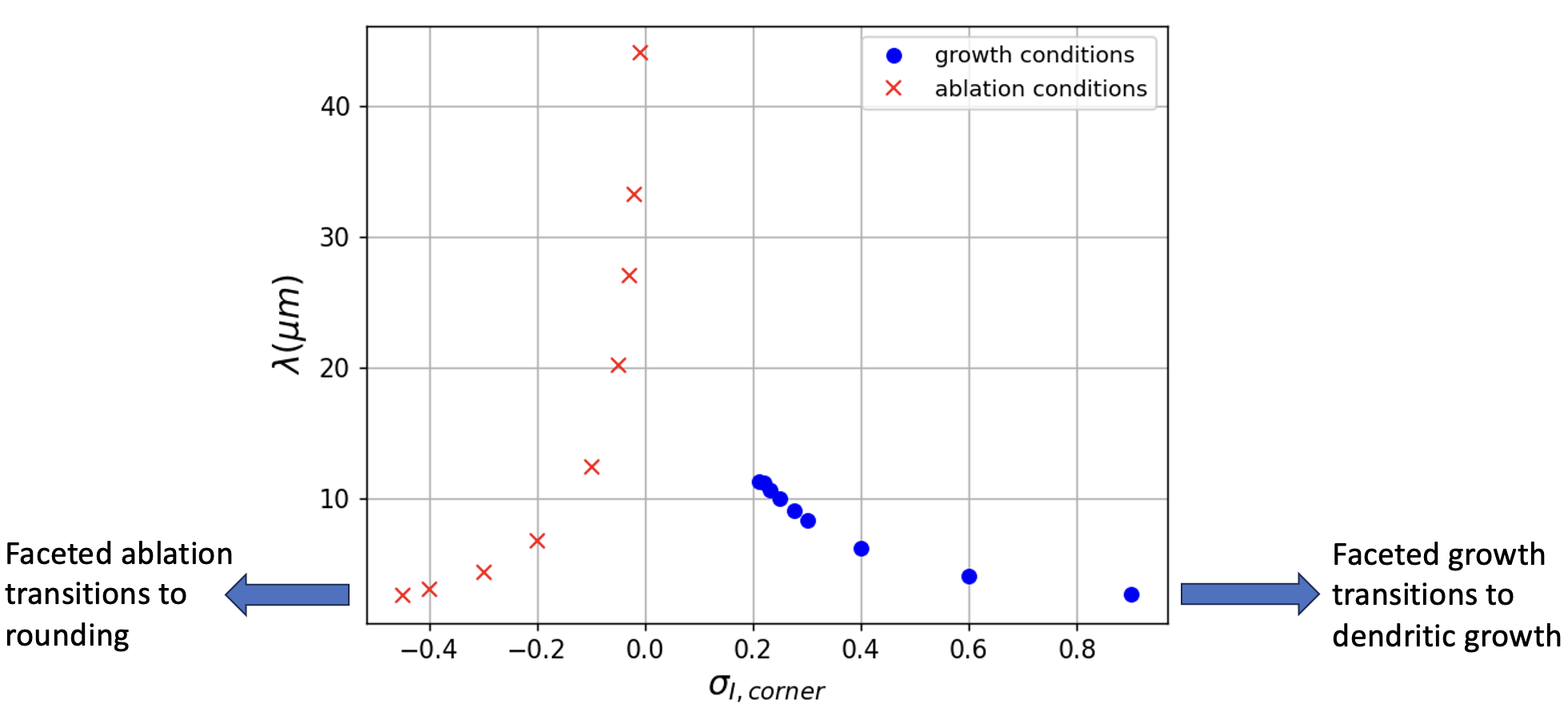
*II. Dependence of surface morphology on model conditions.*

A useful metric for describing the morphology of steady state profiles such as those appearing in Fig. 3 is the horizontal distance between successive molecular layers. Here we define a mean value of that distance as

(4)

Values of are shown in Fig. 5, based on a series of simulations in which the supersaturation at the corner of the crystal () is varied, and all other parameters fixed. Focusing first on the right-hand side, we see that when conditions are supersaturated relative to the most volatile microsurface of the model (microsurface II) (e.g., in the top panel of Fig. 2), steady-state spatial wavelengths start a little over , and get smaller (more steps in a profile …) with increasing supersaturation …. Eventually, at high enough supersaturation, faceted growth yields to dendritic growth.

On the left-hand side of Fig. 5 are displayed results when conditions are subsaturated relative to the least volatile microsurface of the model (microsurface I). We see that under these ablation conditions, steady-state spatial wavelengths much higher – over – and decrease for more extreme subsaturations farther to the left. Proceeding to the left … eventually yielding to rounding …

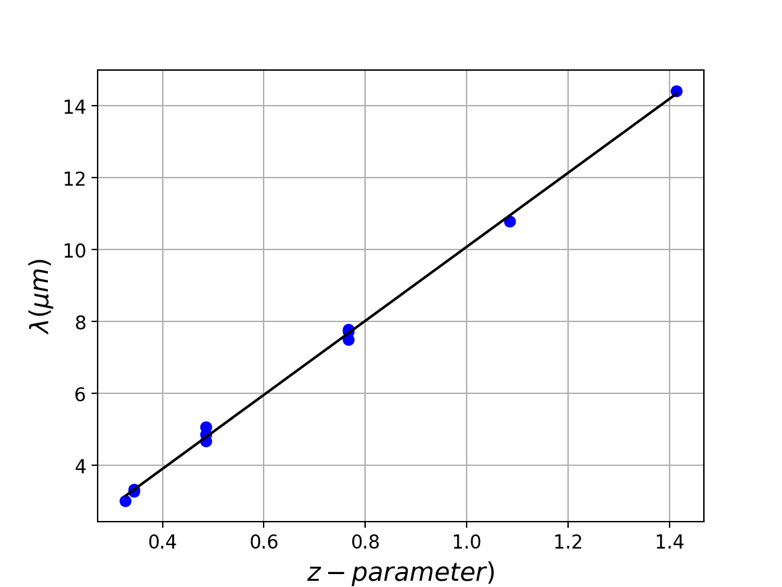


**Figure 5**. Spatial wavelength () as a function of corner supersaturation .

Figure 6 shows the dependence of on a parameter , defined by

(5)

where is the surface diffusion coefficient, is the facet center reduction in supersaturation compared to facet corners, and is the kinetic deposition velocity. We see that … The observation that is proportional to is noteworthy because of the connection to Turing patterns ….



**Figure 6**. Surface layer wavelength () as a function of parameter for a range of values of , , and . The corner supersaturation is fixed at for all points.

*III. Facet concavity and convexity*

An SEM image of a growing crystal, and GNBF reconstruction of a portion of its basal facet, are shown in Fig. 7. GNBF reconstruction of this surface reveals a distinct concavity, on the order of 1000s of layers in a comparable horizontal span, hence .

We can use the model to interpret this result. Returning to Fig. 3, we note that facet concavity implies that this crystal is growing. This is certainly the case: subsequent images taken of this crystal revealed expanding boundaries against the metal substrate to which the crystal is attached. Moreover, inspection of Figs. 5 and 6 would lead us to the following possible inferences:

1. Water vapor is at a high concentration in the SEM chamber, leading to large ;
2. Air in the chamber is at a high pressure, inhibiting water vapor transport across the facet, hence large ; or
3. The temperature is such that the ratio is small.

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| **Figure 7**. SEM image of a growing crystal (left) with its basal surface retrieved (right) | |

*IV. Facet resilience*

Facet resilience is defined here as the ability of a facet to recover from a perturbation. Actual ice crystal facets exhibit high resilience. For example, in SEM experiments, facets that have been roughened by some perturbation (e.g., by higher temperature) are commonly observed to be restored to smoothness within less than a minute upon reversal of the perturbation. An example is shown in Fig. 8

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| **Figure 8**. SEM image of a rough facet restored to smoothness. |

Figure 9 is an examination of facet resilience of the model. The blue curve is a timeline of , beginning with a flat profile but subject over time to the inverse parabolic supersaturation curve indicated by the dashed line in Fig. 2b. It evolves to the steady-state profile shown in the lower-left inset, characterized by . The gray curve begins with the same initial, flat profile, but is subject over time to the sinusoidal supersaturation curve indicated by the solid line in Fig. 2b. It also evolves to a steady state – the “perturbed” profile shown in the upper-right inset – but that profile is quite different from the first. When the saturation curve is replaced by the original (dashed line in Fig. 2b), we see that the perturbed state evolves back to the original steady state.

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| **Figure 9**. Examination of model facet resilience. |

*V. Does facet roughness have an intrinsic characteristic length scale*?

The model presented here is not, unfortunately, capable of the extreme depth variations that mesoscopic roughening represents, because the vertical relief is too great to resolve in our model. There is, however, a hint that the characteristic wavelength of roughness is an intrinsic property of the crystal, rather than the result of some imposed variability (e.g., a repeating variability in the overlying vapor field). That hint is the fact that the model predicts a substantial difference in growth vs ablation values (as shown in Fig. 5), in parallel with the difference between growth vs ablation characteristic distances (as shown in Fig. 4).

*VI. What about differences between facets?* Because facets have distinct underlying crystal cell structures, we can expect that their quasi-liquid properties (in the model, and ) will also be distinctive. Moreover, numerical studies show that the thickness of a single “layer” of ice leads to a proportional increase in .