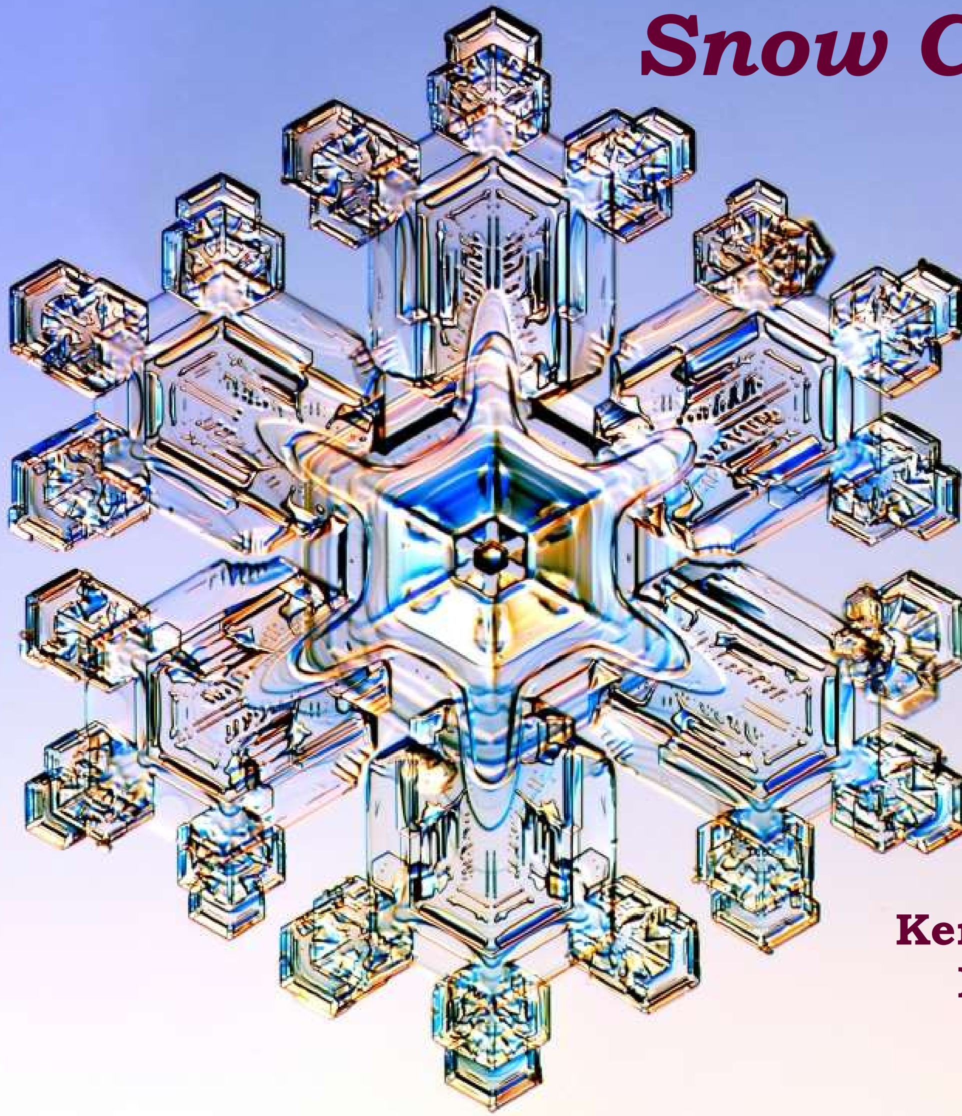
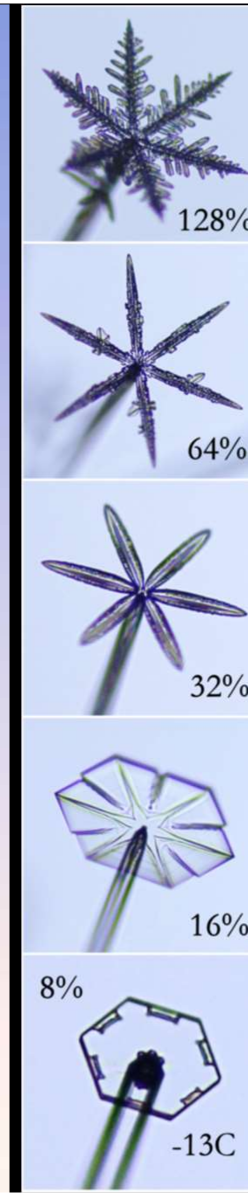


Snow Crystals: III

A Comprehensive Physical Model of Snow Crystal Growth

Kenneth G. Libbrecht
Dept. of Physics
Caltech



A COMPREHENSIVE MODEL OF SNOW CRYSTAL GROWTH

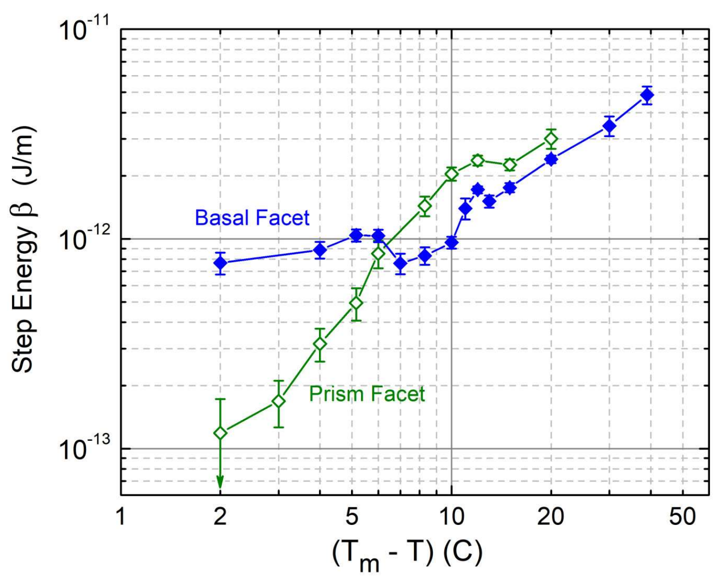
Basic Components

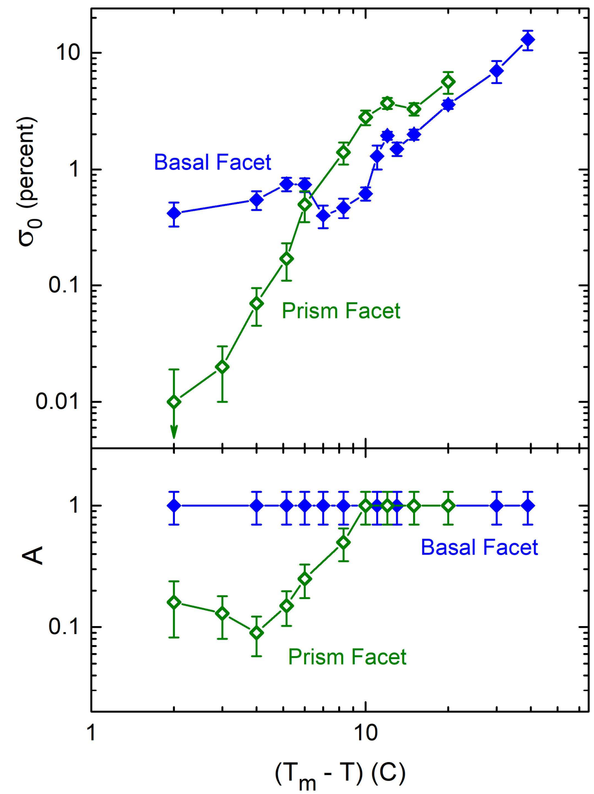
- Particle diffusion (through air)
- Molecular attachment kinetics

  $\alpha \approx 1$ on rough surfaces

  $\alpha = Ae^{-\sigma_0/\sigma_{surf}}$ on broad facets

Lesser contributions from : latent heating, surface energy effects, …

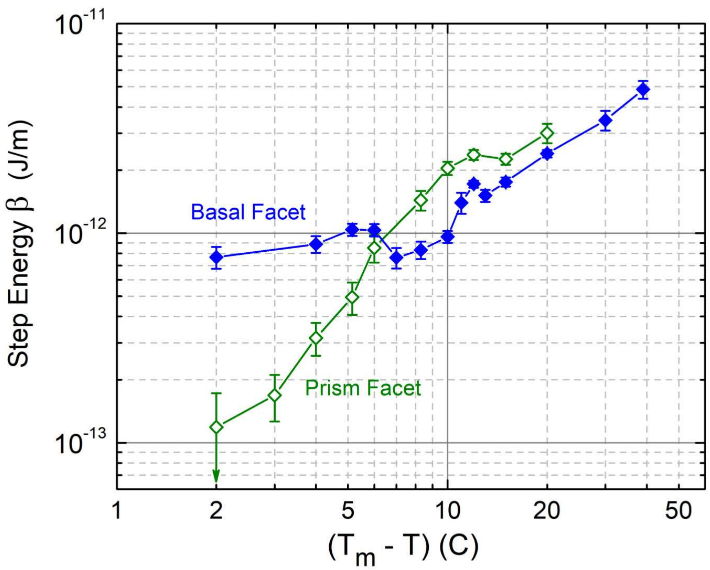


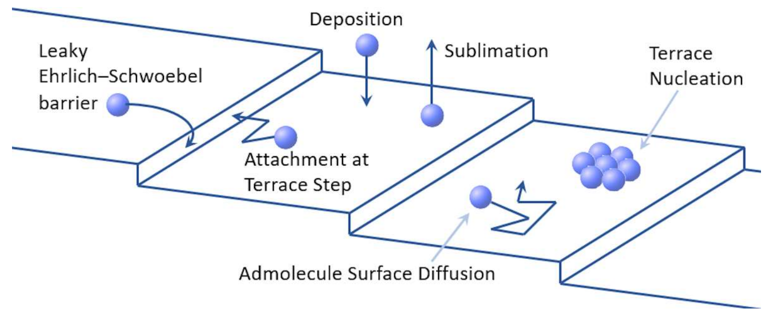


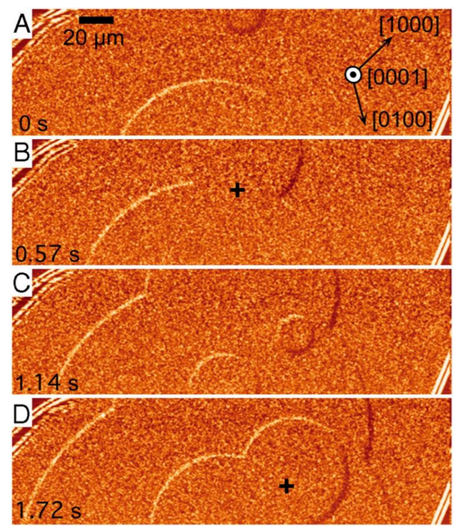
Best measurements to date…
Nothing radical here…
Additional verification?

Other ways to measure step energies?

Possibly… equilibrium quantity
But difficult to measure

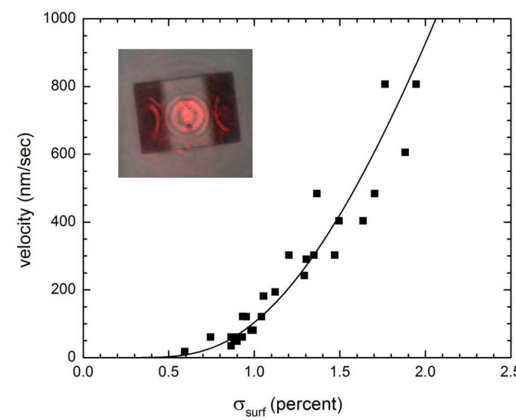




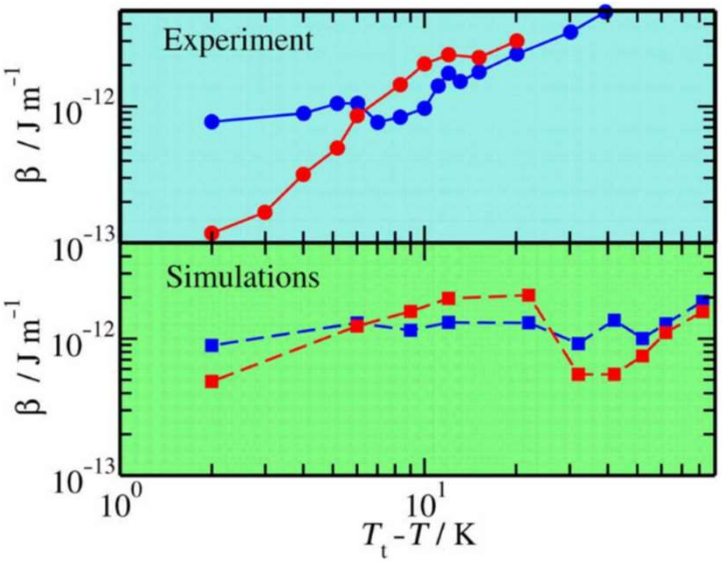


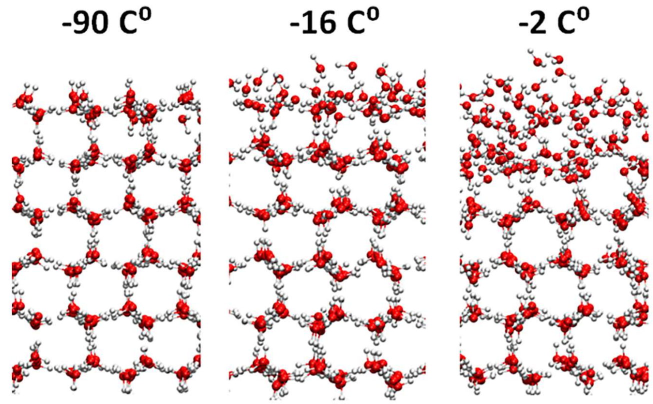
G. Sazaki et al., Elementary steps at the surface of ice crystals visualized by advanced optical microscopy, PNAS 107, 19702-19707, 2010.

Independent experiments would be helpful!

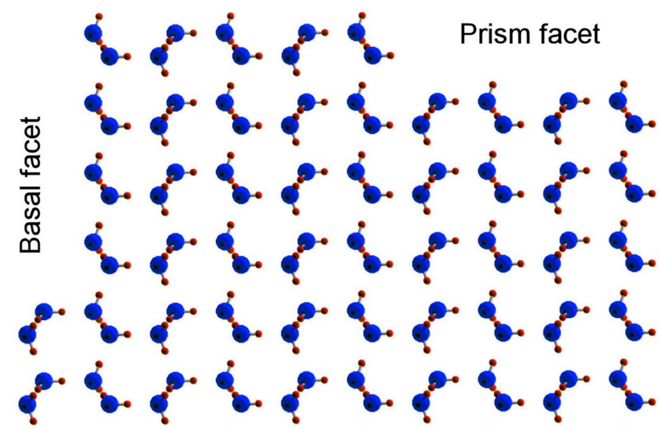


Verify terrace step energies using MD simulations?





← "robustness" analysis?



Measure β with computational chemistry
yes, but unknown accuracy (so far)
***Step energy = *equilibrium* property
Lots of interest in getting water right…
Much room for improvement, AI, etc.

Pablo Llombart, Eva G. Noya, and Luis G. MacDowell, Surface phase transitions and crystal growth rates of ice in the atmosphere, Science Advances 6, no. 21, eaay9322, DOI: 10.1126/sciadv.aay9322, 2020. Also arXiv:2004.10465, 2020.

SDAK: Structure Dependent Attachment Kinetics

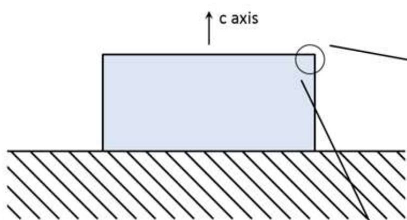
Hypothesis:

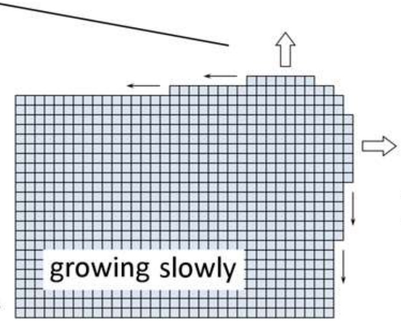
Large facet → normal terrace nucleation model (measured σ_0) … done

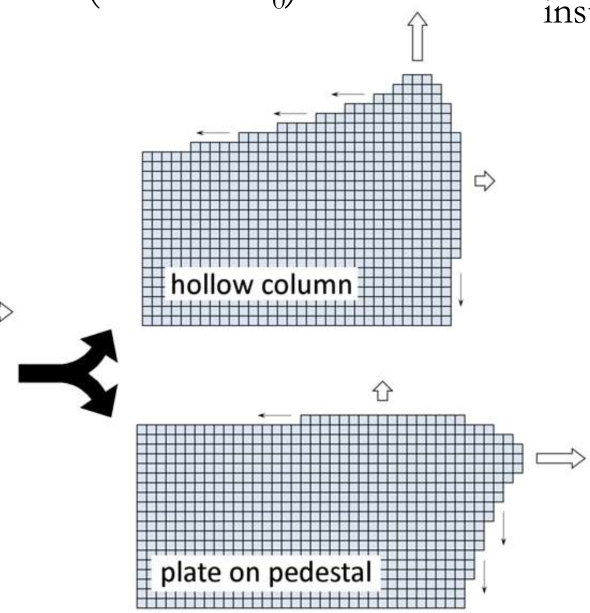
Narrow facet (edge) → higher α, smaller nucleation barrier (reduced σ_0)

→ an ***edge-sharpening instability*** *(in air)*

Not the Mullins-Sekerka instability!







Assume SDAK hypothesis…

→ Abrupt changes in *anisotropy* of the attachment kinetics → thin edges

→ Can have abrupt morphological changes with temp

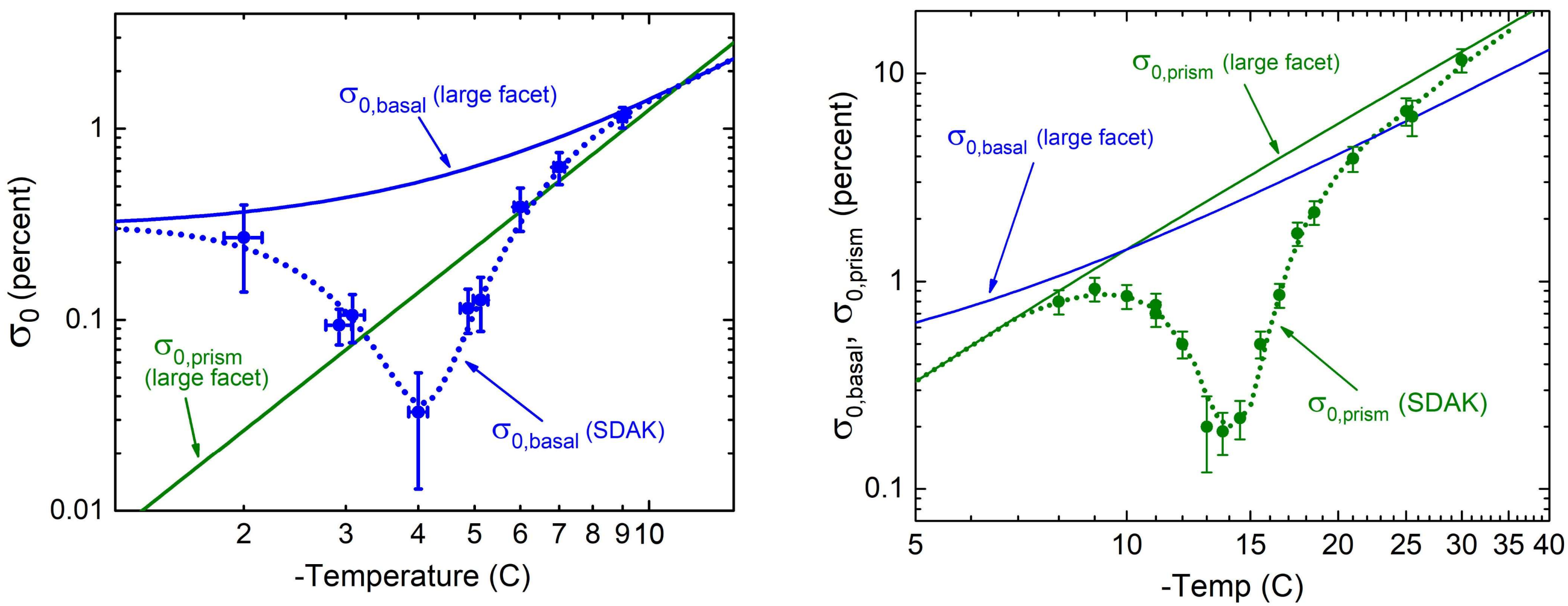
→ Explains this aspect of Nakaya diagram

→ Explains why no narrow facets in vacuum

KGL, Explaining the formation of thin ice-crystal plates with structure-dependent attachment kinetics, J. Cryst. Growth 258, 168-175, 2003.

SDAK: Test hypothesis using measurements

Putting all the data together → a pair of "SDAK dips"



Assume SDAK hypothesis … narrow facets ≠ broad facets

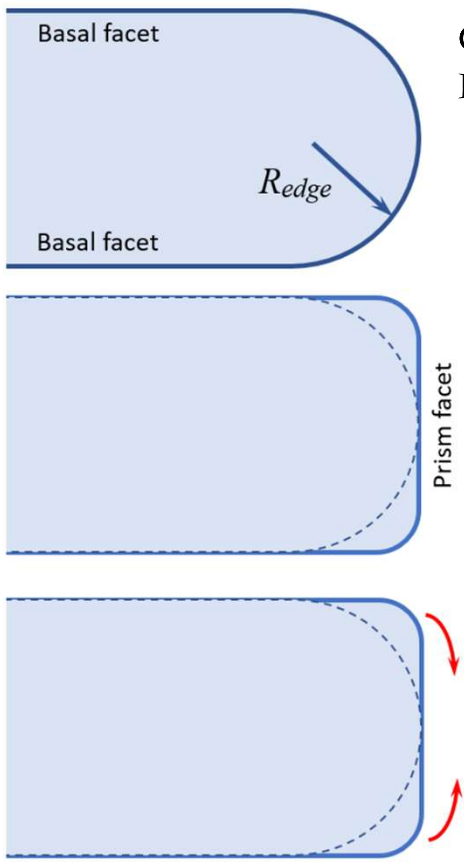
Measure σ_0 → a "repackaging" of the Nakaya diagram

…**Parameterized** and **Quantified**

Based on growth data, still empirical

Suitable for creating computational models

SDAK: A physical model



Consider the edge of a platelike snow crystal

Low supersaturation → basal surfaces do not grow appreciably

→ rounded prism edge ~stable equilibrium

Increase supersaturation slightly

→ corners become less rounded

→ surface energy out of equilibrium

Allow surface admolecule diffusion (to prism surface only)

→ energetically favorable to move molecules to prism facet

→ prism admolecule density anomalously high (if narrow facet)

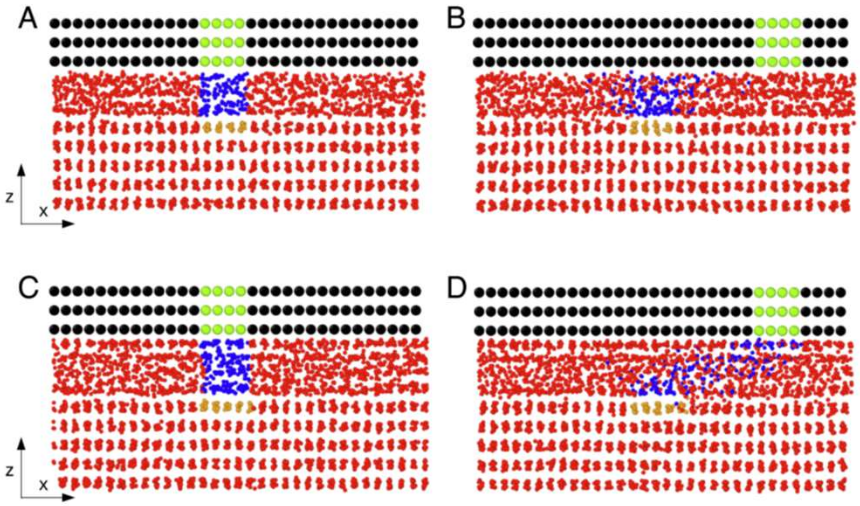
→ enhances terrace nucleation

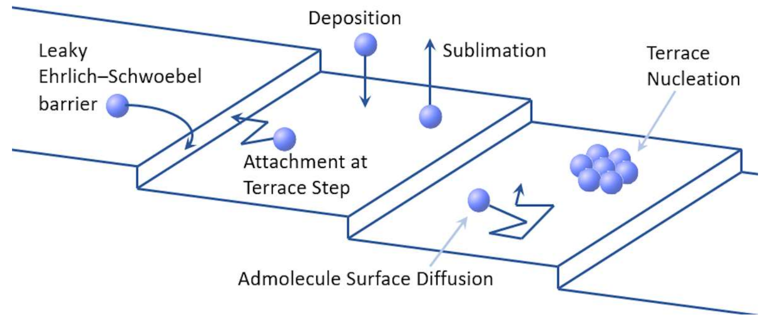
→ lowers effective $\sigma_{0,prism}$ (but only on narrow prism facets)

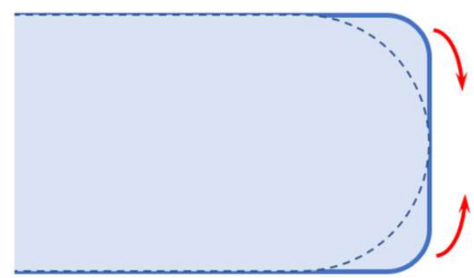
→ SDAK effect!

KGL, A quantitative physical model of the snow crystal morphology diagram, arXiv:1910.09067, 2019.

Verify SDAK model with MD simulations?







MD models can examine (vs Temp, facet)
- lateral diffusivity?
- QLL viscosity?
- Ehrlich-Schwoebel barrier

Robustness of MD model is key

Baran Łukasz, Pablo Llombart, Wojciech Rżysko, and Luis G. MacDowell, Ice friction at the nanoscale, Proc. Natl. Acad. Sci. 119, e2209545119, 2022.

Possible to model physical dynamics of driven surface diffusion?

→ Build model of SDAK mechanism

But surface transport challenging…

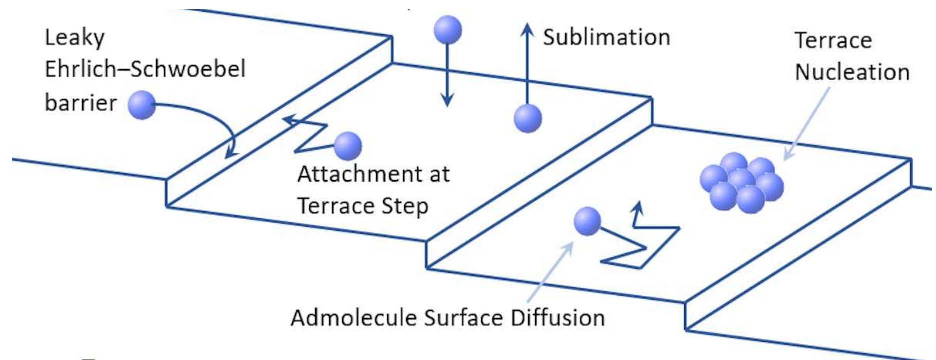
Verify SDAK model features with surface probes?

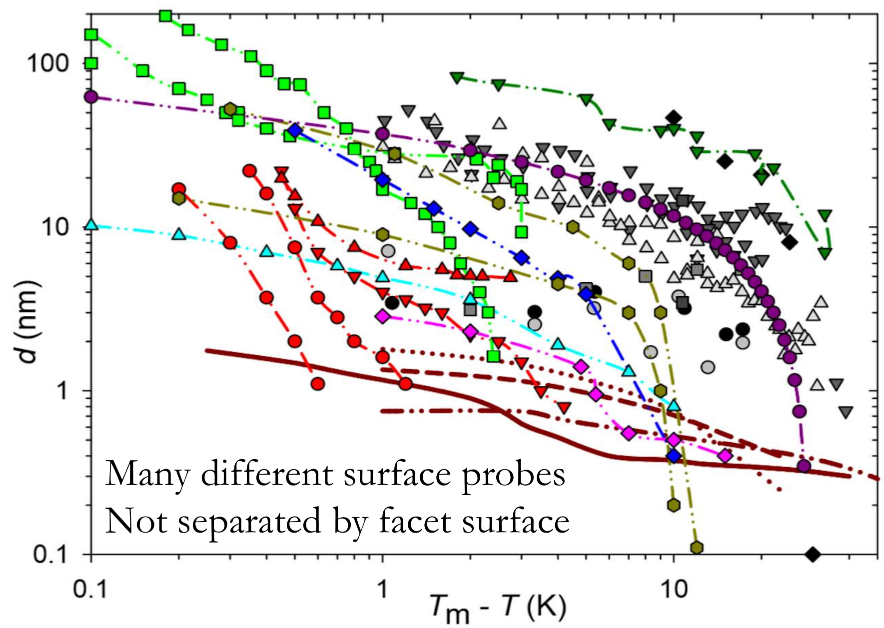
Perhaps measure
> Ehrlich-Schwoebel barrier height as function of temperature on basal and prism facets?
> QLL viscosity, surface diffusion?

Measure other properties of **surface premelting** as function of temperature?

Best to do identical measurements on basal and prism facets.

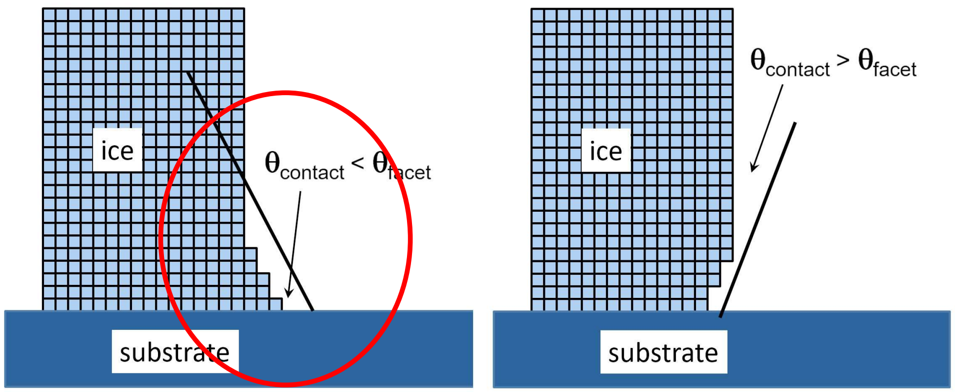
Look for "onset" of premelting at -4C (basal) and -14C (prism)

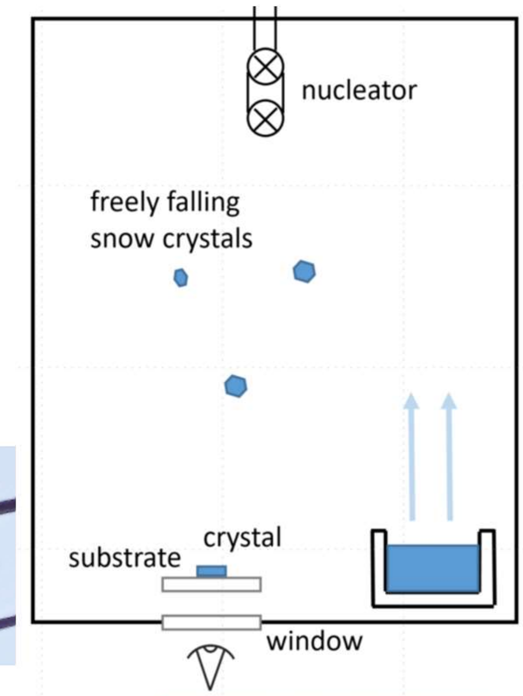




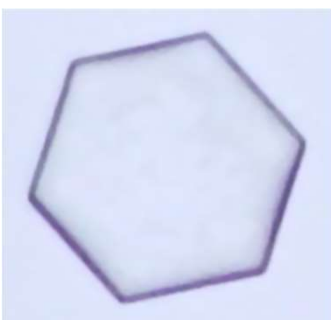
Uncertainty analysis?

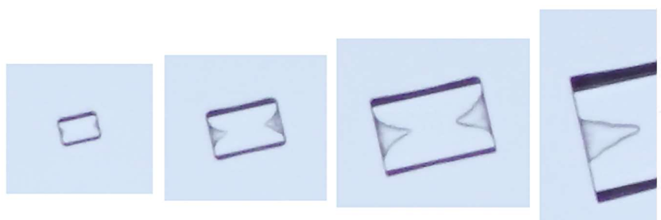
A method for creating large basal and prism facets





Small plate
→ large plate





Small column with one prism facet against surface
→ large prism facet parallel to substrate
Can grow either basal or prism facets
Facets slowly grow upward → fresh surface

A COMPREHENSIVE MODEL OF SNOW CRYSTAL GROWTH

Essential Components

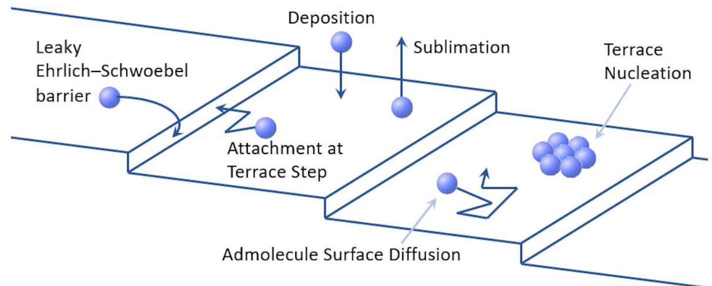
Suggests many new avenues of investigation…

- Particle diffusion (through air)
- Molecular attachment kinetics

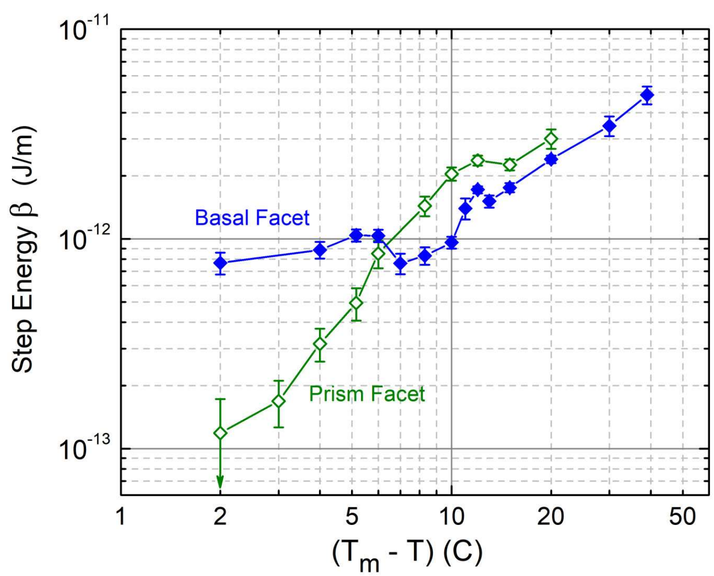
  $\alpha \approx 1$ on rough surfaces

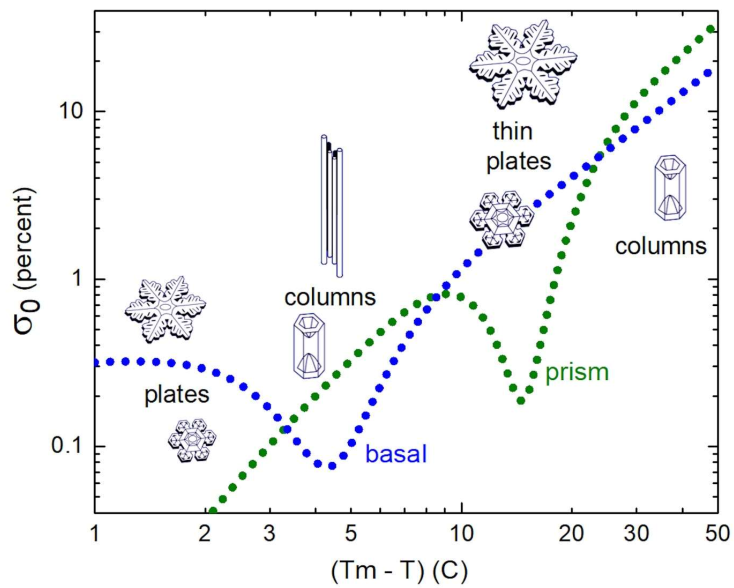
  $\alpha = Ae^{-\sigma_0/\sigma_{surf}}$ on broad facets

  SDAK on narrow facets



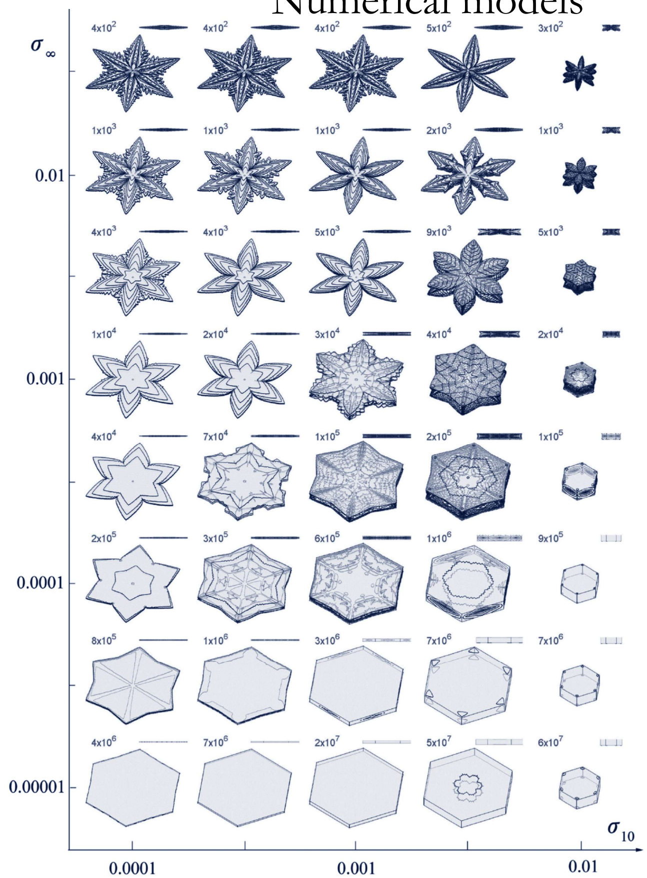
Lesser contributions from: latent heating, surface energy effects, …



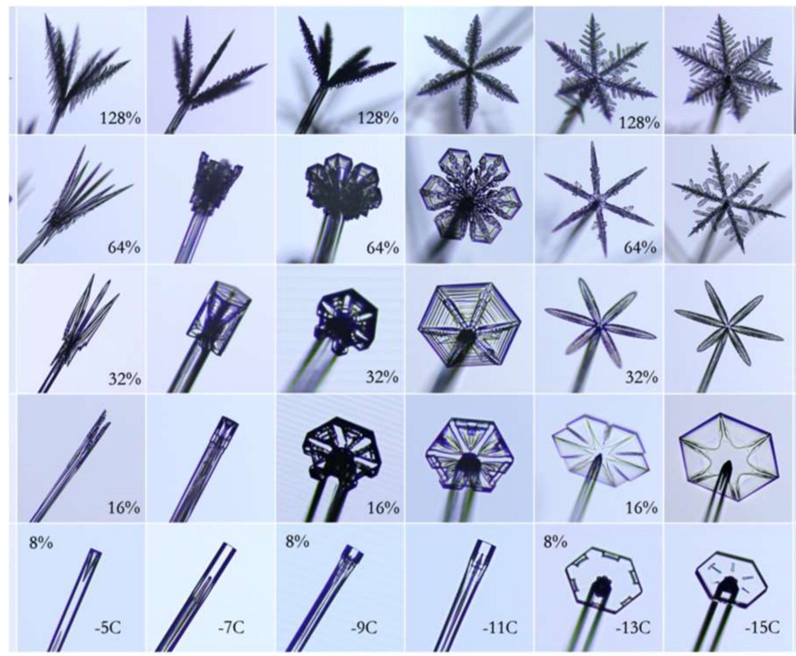


Computational Snow Crystals

Numerical models



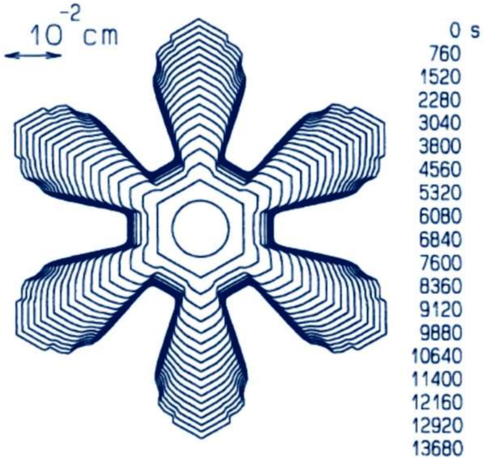
Laboratory grown

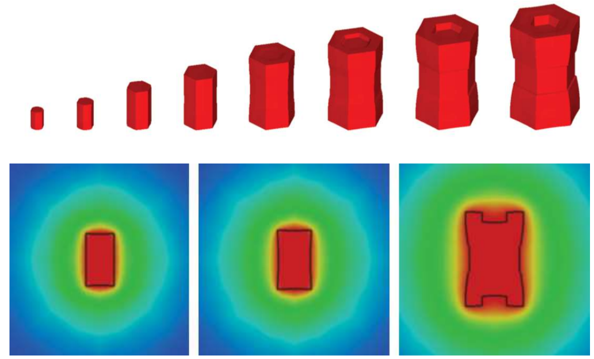


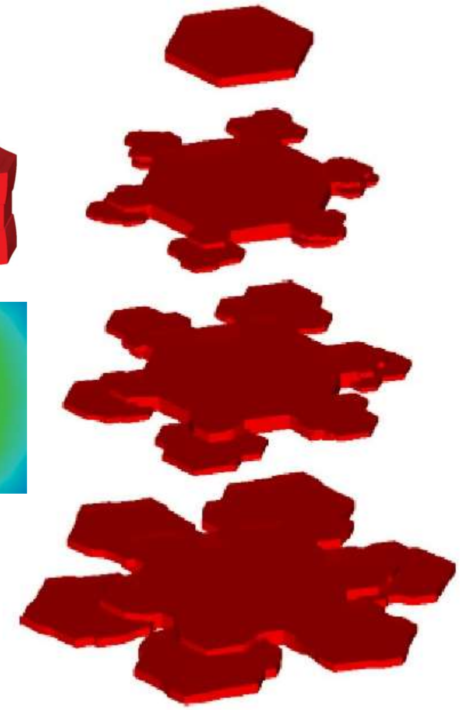
Can we bring these two sides together? (So far… no)

Goal: model physically realistic faceted + branched crystal structures

Computational Snow Crystals







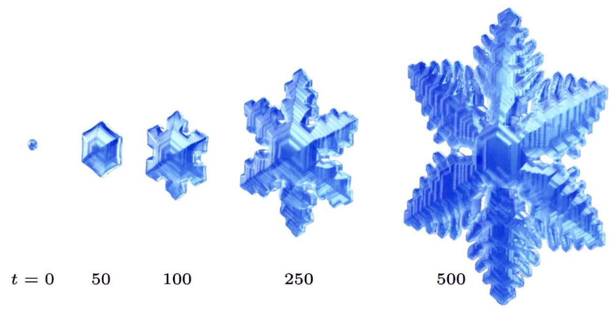
2D model
Good physical basis
Anisotropic attachment kinetics

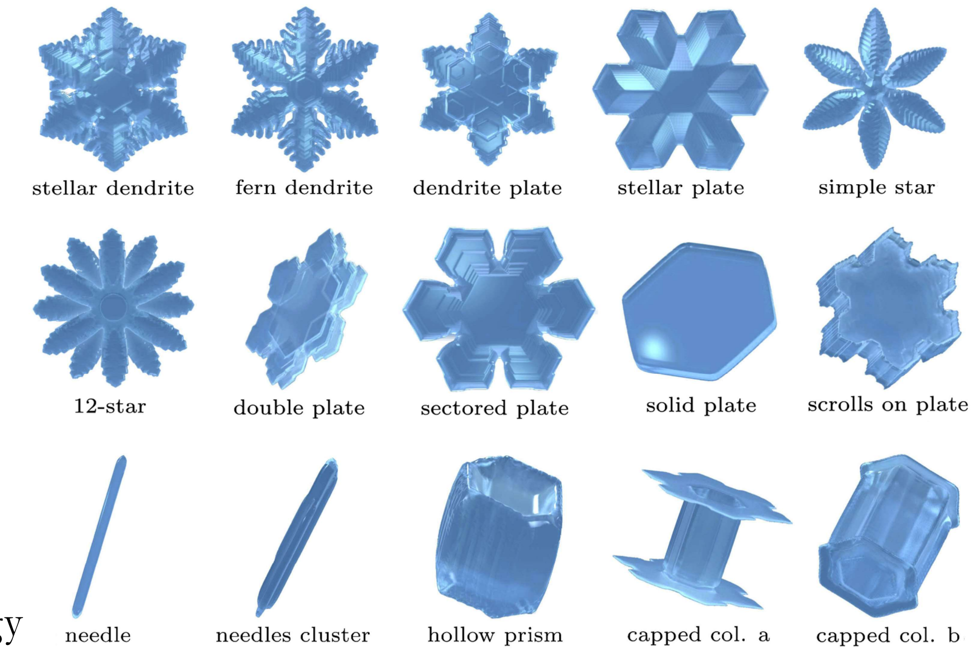
3D model
Front-tracking method
Poor physical basis
(Highly anisotropic surface energy not realistic)

Etsuro Yokoyama and Toshio Kuroda, Pattern formation in growth of snow crystals occurring in the surface kinetic process and the diffusion process, Phys. Rev. A 41, 2038-2049, 1990.

John Barrett, Harald Garcke, and Robert Nürnberg, Numerical computations of faceted pattern formation in snow crystal growth, Phys. Rev. E86, 011604, 2012.

<u>Computational Snow Crystals</u>





3D model
Phase-field method
Poor physical basis
(Highly anisotropic surface energy not realistic)

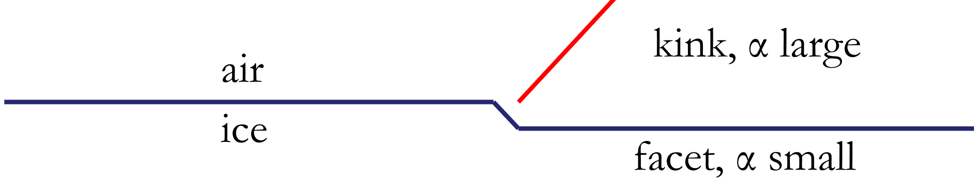
Gilles Demange et al., A phase field model for snow crystal growth in three dimensions, Computational Materials 3, 15, 2017.

In all models, solving the diffusion equation in air is easy
The devil is in the boundary conditions…
And need to use realistic physics!

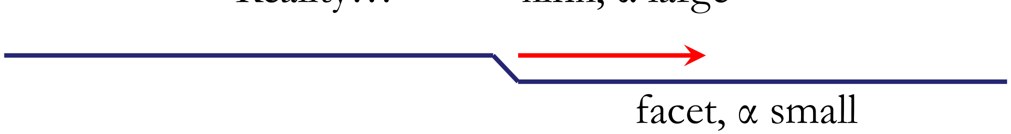
Computational instabilities

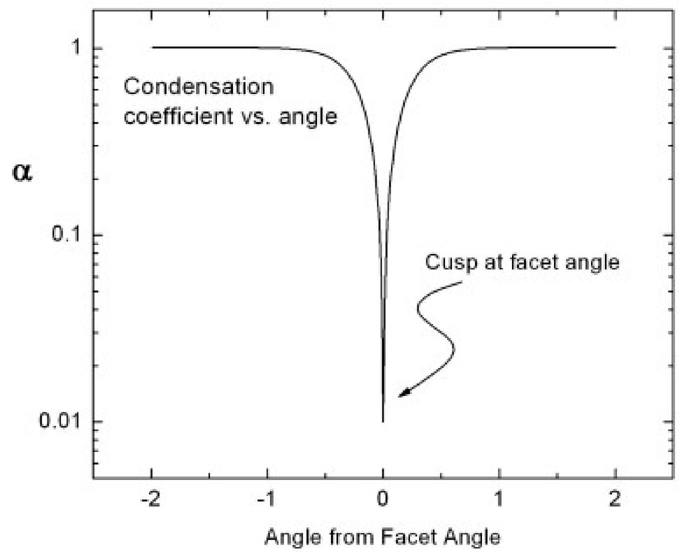
Highly anisotropic attachment kinetics difficult to model with continuum methods

Front tracking model:
Model output…



Reality…



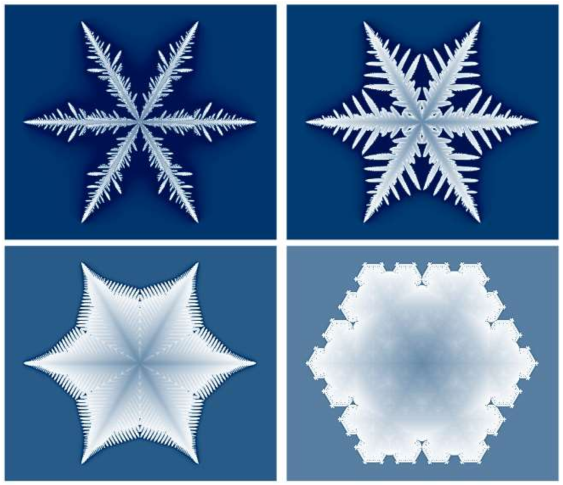


Deep cusp in the attachment coefficient
→ Numerical instabilities
Resolution ???

Much early model development in growth from melt, weak anisotropy, no faceting
No physically realistic modeling of faceted + branched systems (yet)…

Cellular Automata

Best method to date for handling highly anisotropic attachment kinetics



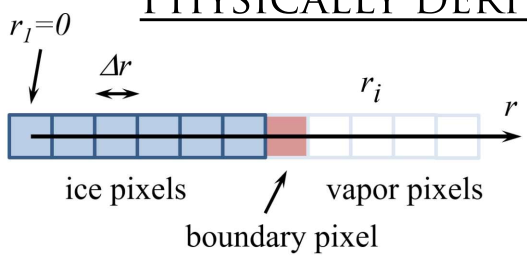
2D model
Poor physical basis

C. A. Reiter, A local cellular model for snow crystal growth, Chaos, Solitons, and Fractals 23, 1111-1119, 2005.

3D model
Not-too-bad physical basis
Highly anisotropic attachment kinetics

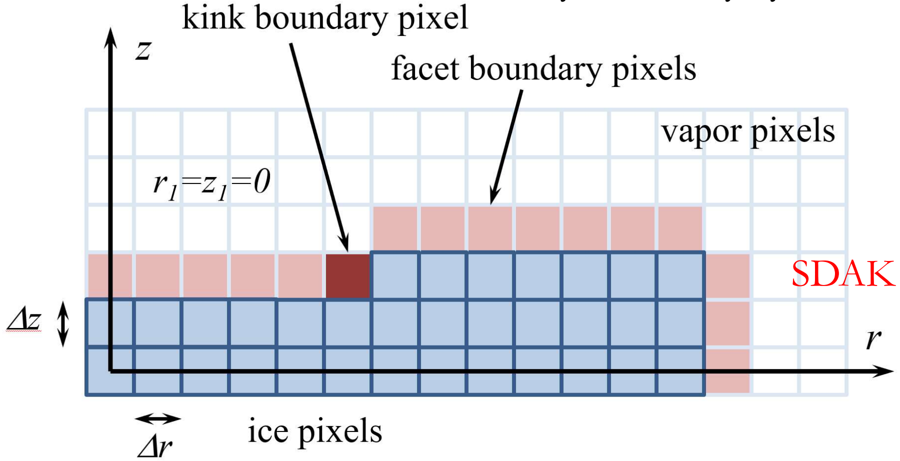
Janko Gravner and David Griffeath, Modeling snow-crystal growth: A three-dimensional mesoscopic approach, Phys. Rev. E79, 011601, 2009.

Physically derived Cellular Automata



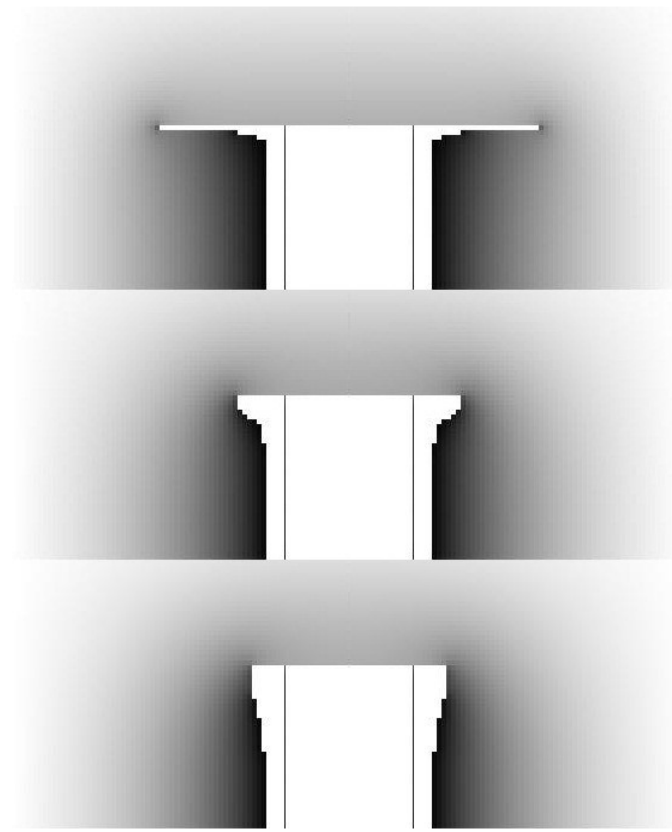
1D model
Reproduce spherical solution

2D model
Cylindrically symmetric



KGL, Quantitative modeling of faceted ice crystal growth from water vapor using cellular automata, J. Computational Methods in Phys., ID-174806, 2013. (Preprint at arXiv:0807.2616, 2008.)

Need surface energy to stabilize plates…

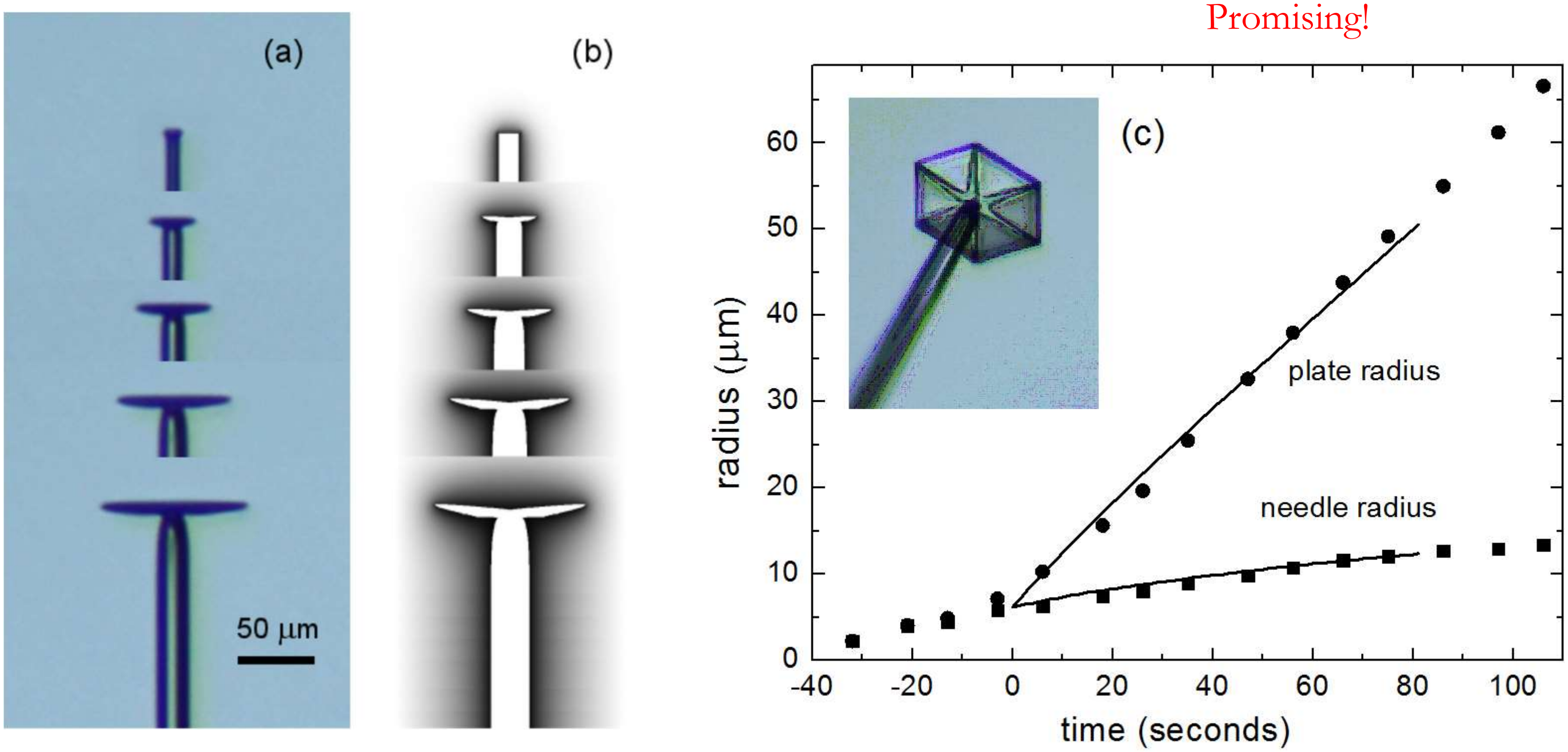


2D Modeling of Plate-on-Needle Growth

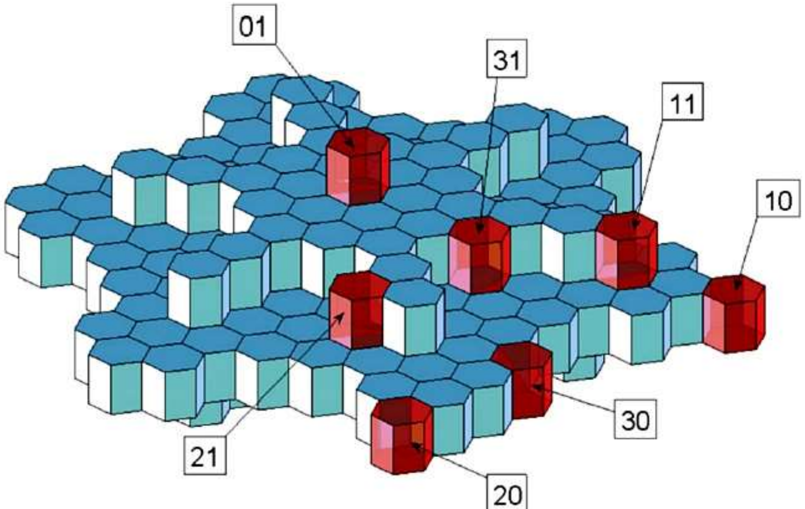
Cylindrically symmetric: Quantitative analysis

Including Structure-Dependent Attachment Kinetics (SDAK)

Promising!



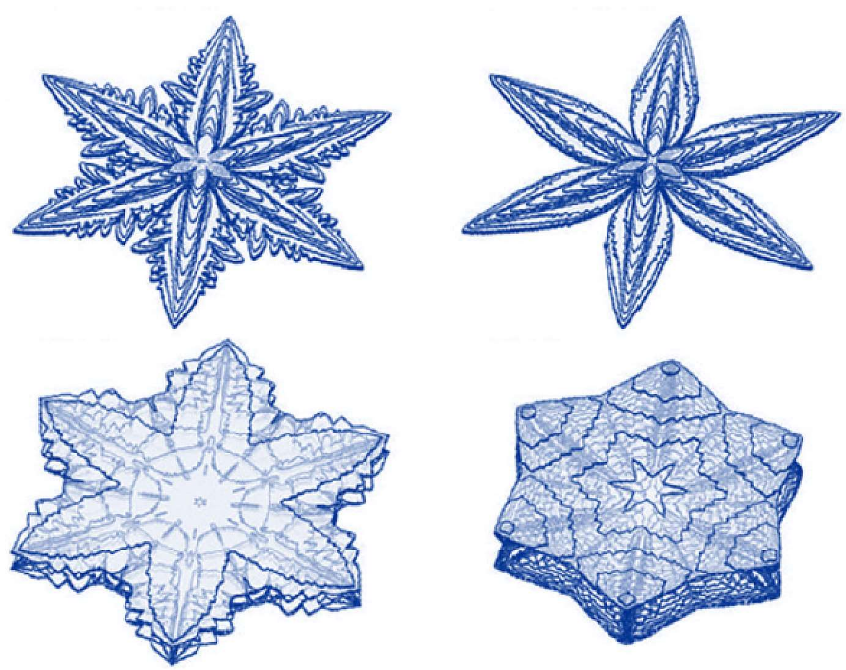
3D Cellular Automata



3D model
Many different types of boundary pixels

SDAK has not yet been implemented…
(but no obvious roadblocks)

Getting close… not quite ready for comparing with observations

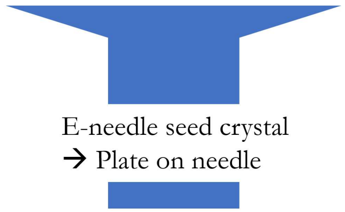


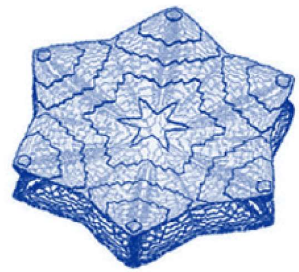
J. G. Kelly and E. C. Boyer, Physical improvements to a mesoscopic cellular automaton model for three-dimensional snow crystal growth, Cryst. Growth & Design 14, 1392-1405, 2014. Preprint at arXiv:1308.4910.

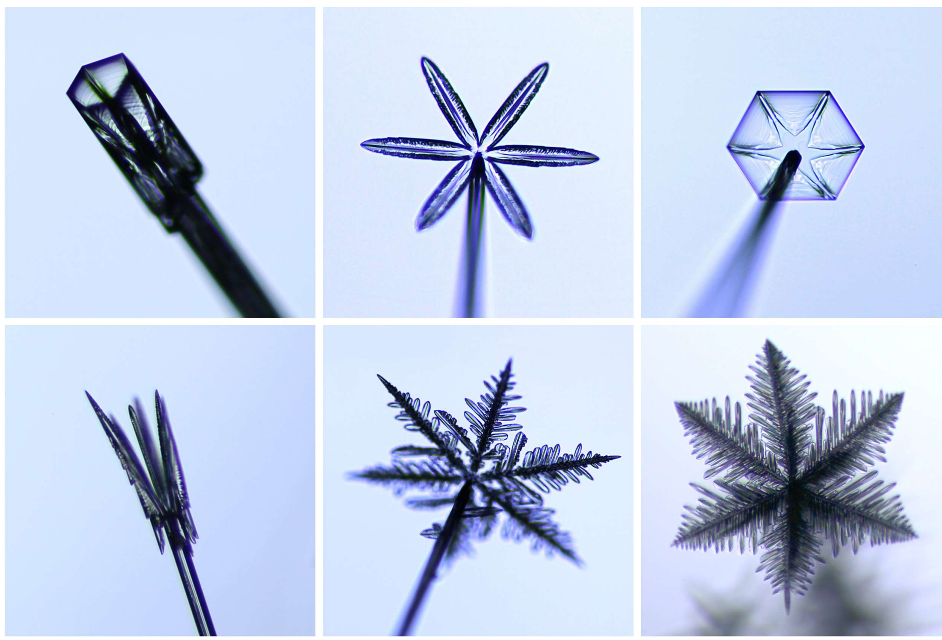
Comparing Models with data: "Electric" Ice Needles

ESI "double-plate" problem (symmetry)

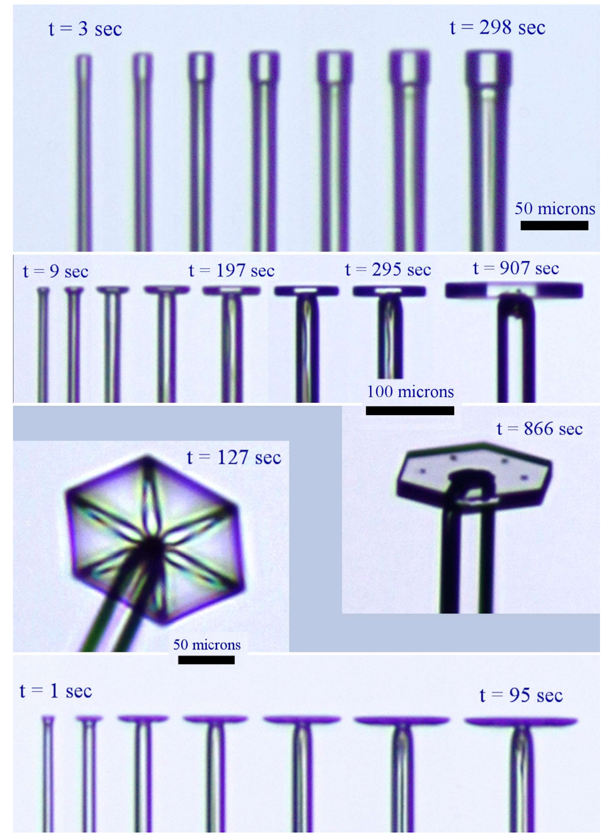
Small prism seed crystal
→ Double plate





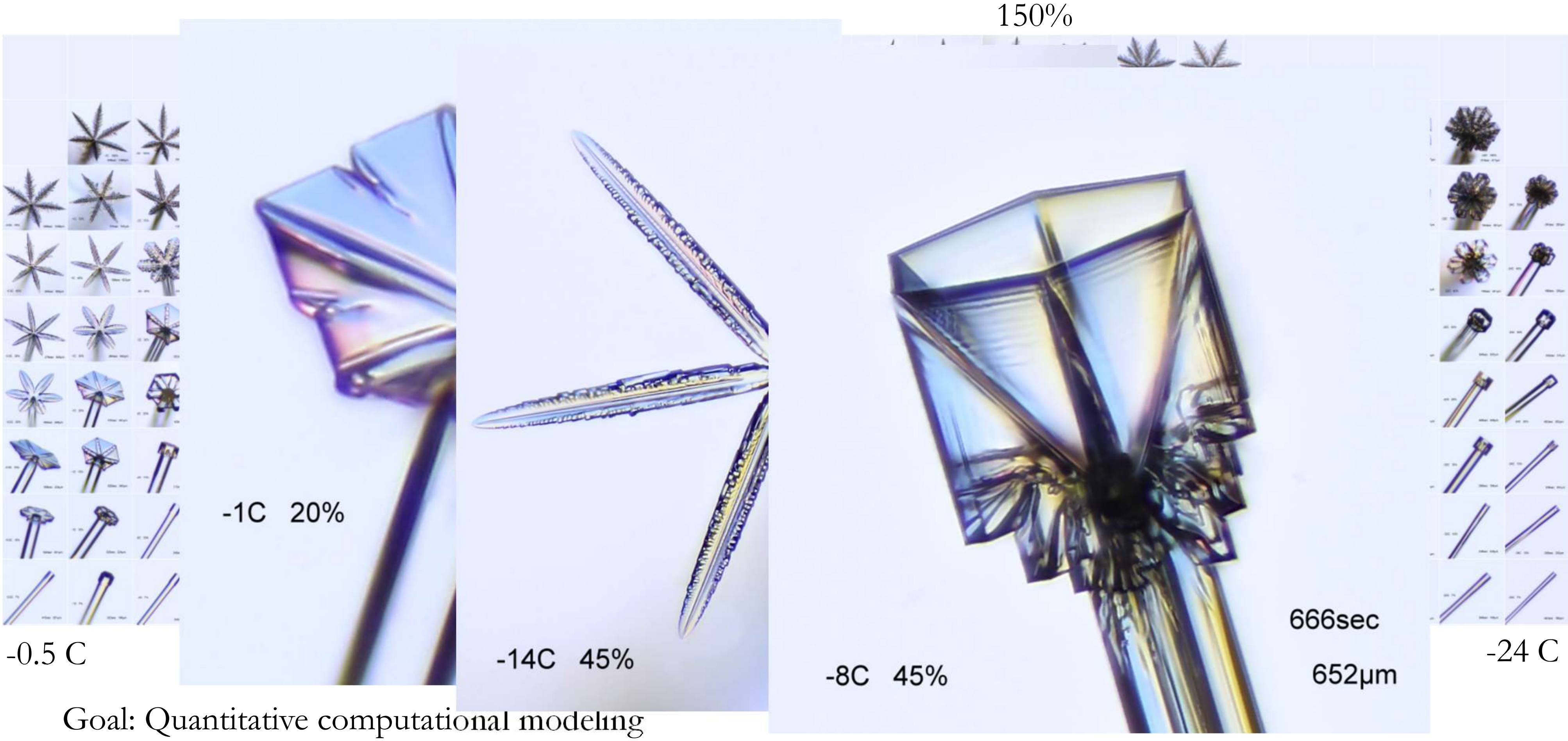


Can explore many morphological behaviors



Targeted experiments

Quantifying the Nakaya Diagram



Goal: Quantitative computational modeling

Reproduce robust growth morphologies and growth rates

Quantifying the Nakaya Diagram

150%

-0.5 C

7%

-24 C

Goal: Quantitative computational modeling

→ Reproduce robust growth morphologies and growth rates … a challenging task!

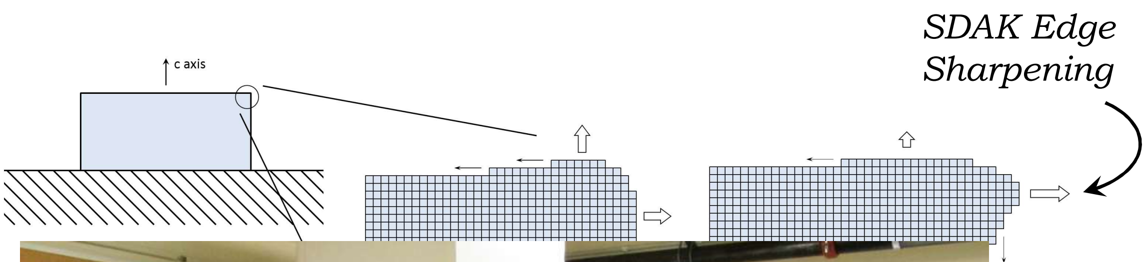
…to what end?

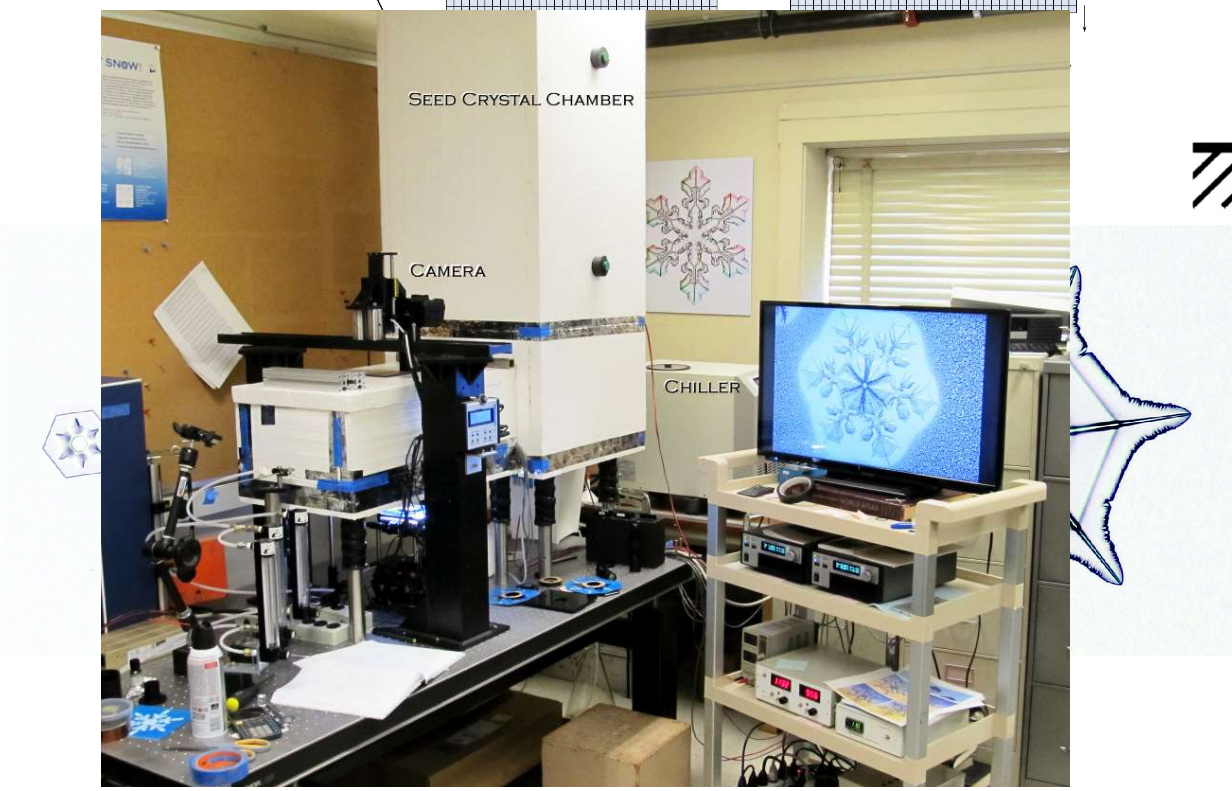
Rainbow Physics

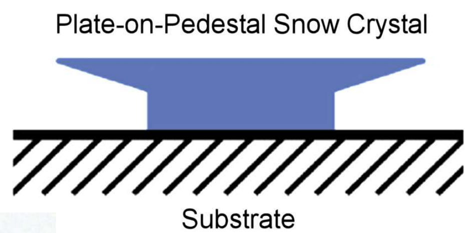
Aristotle (Greece, ~350 BC)
Seneca the Younger (Rome, ~65 AD) – droplets
Shen Kuo (China, ~1060) – droplet theory
Qutb al-Din al-Shirazi (Persia, ~1260) – droplet reflections
Kamāl al-Dīn al-Fārisī (Persia, ~1300) – sphere experiments
Roger Bacon (England, 1268) – droplet colors
Theodoric of Freiberg (Germany, 1307) – primary, secondary bows
Willebrord Snell (Netherlands, 1621) – refraction
Rene Descartes (France, 1637) – reflection+refraction, caustics
Isaac Newton (England, 1672) – dispersion → colors
Thomas Young (England, 1803) – diffraction → supernumerary rainbows
George Biddell Airy (England, ~1820) – refraction theory
Gustav Mie (Germany, 1908) – scattering theory

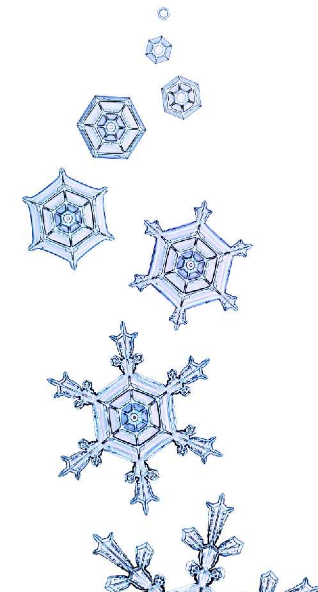
Steve Nelson (Fayfoto, Boston MA)

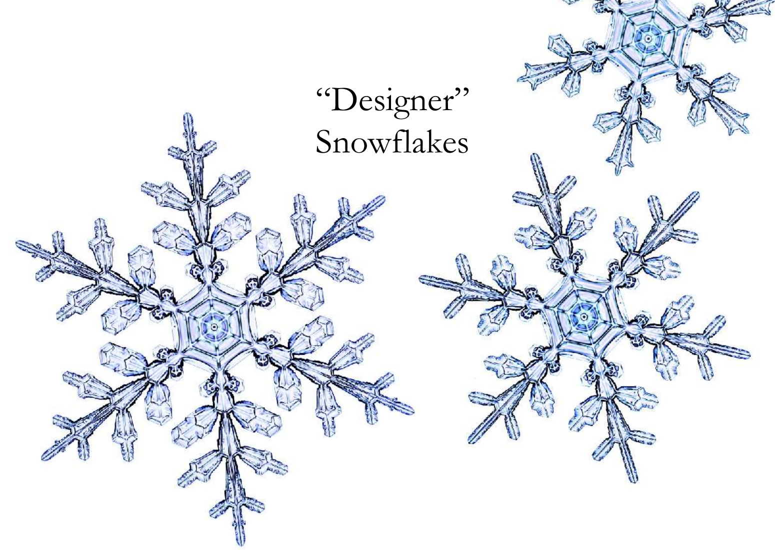
Snowflake Engineering – Plate-on-Pedestal Snow Crystals

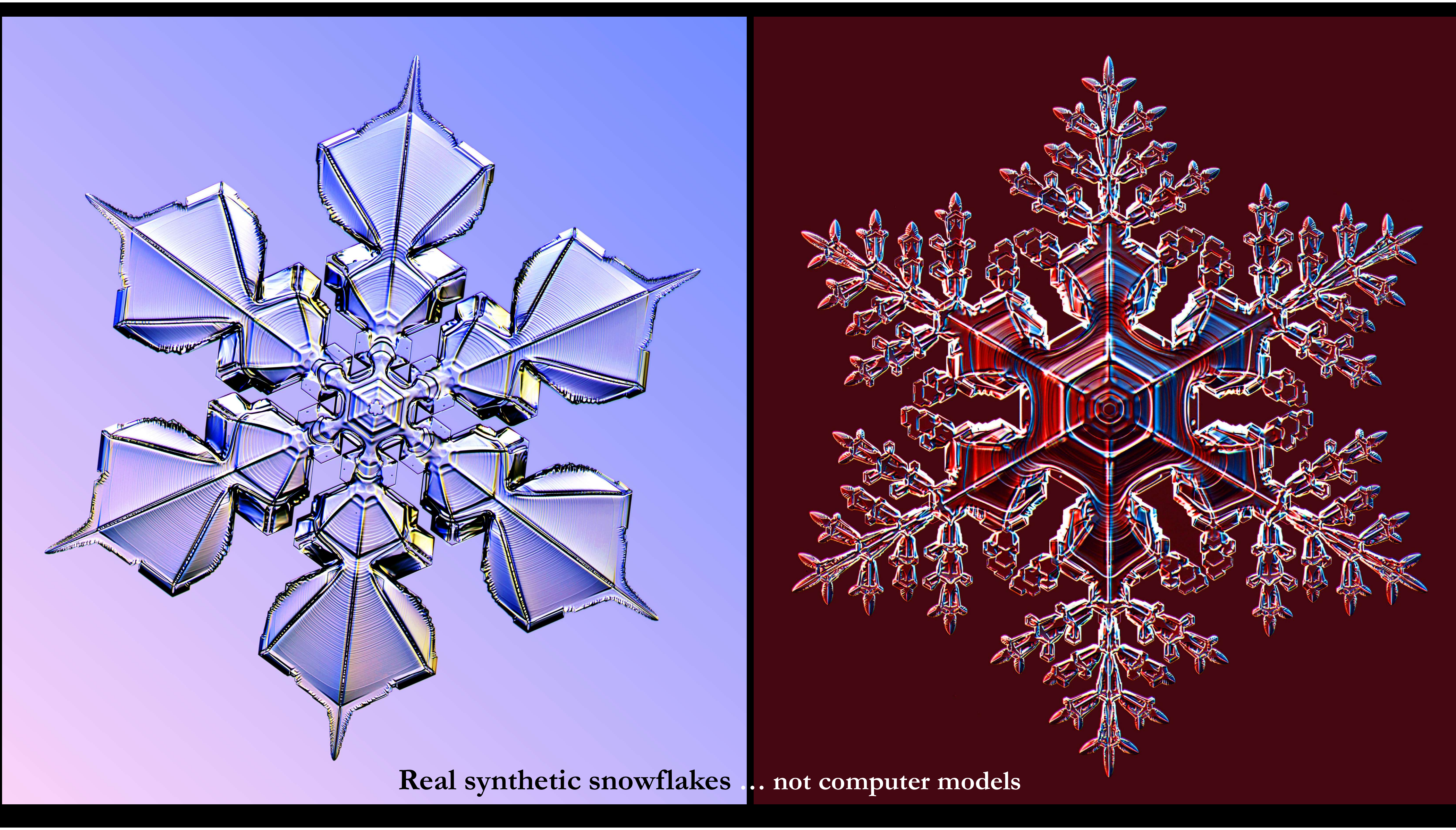




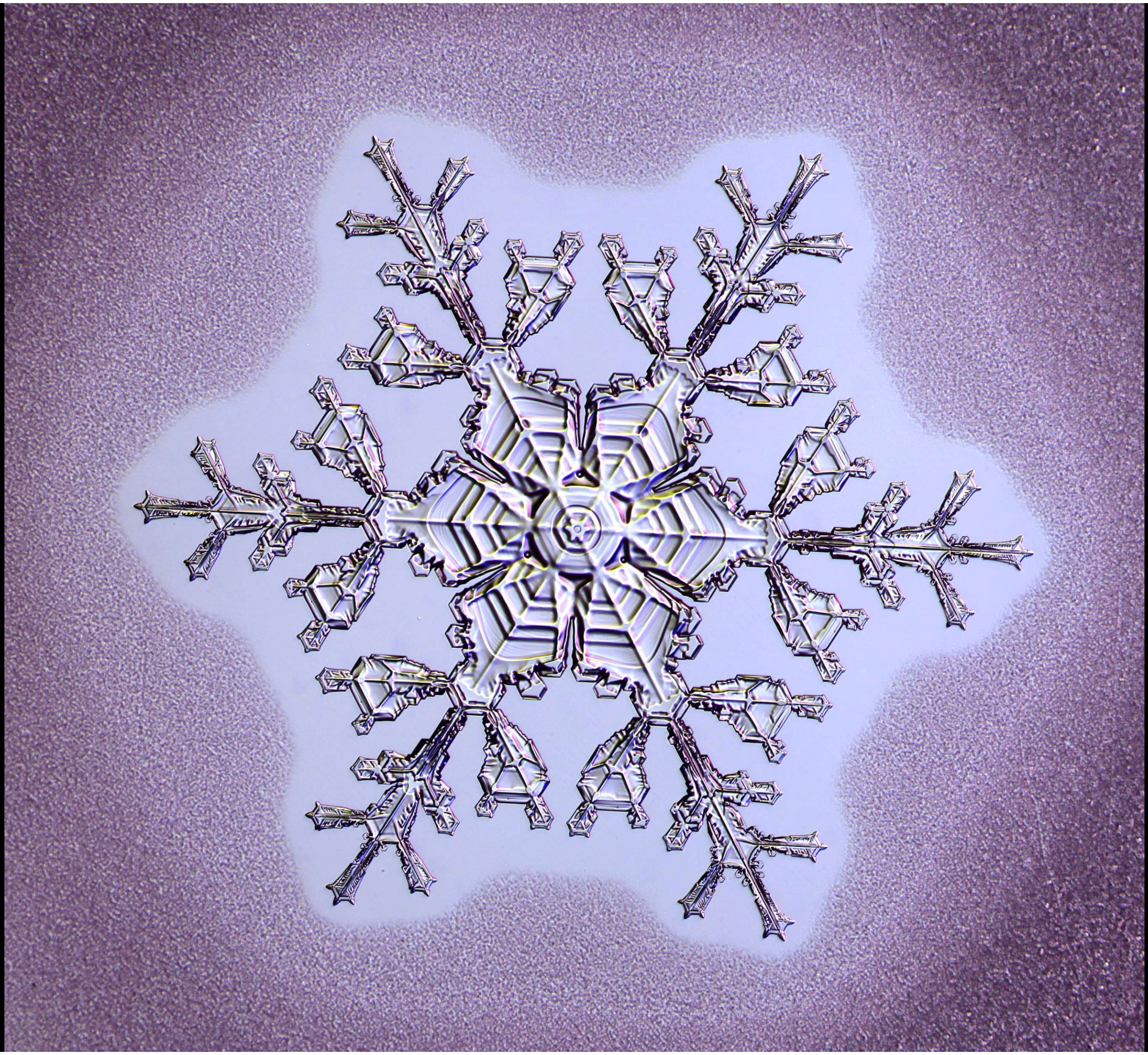


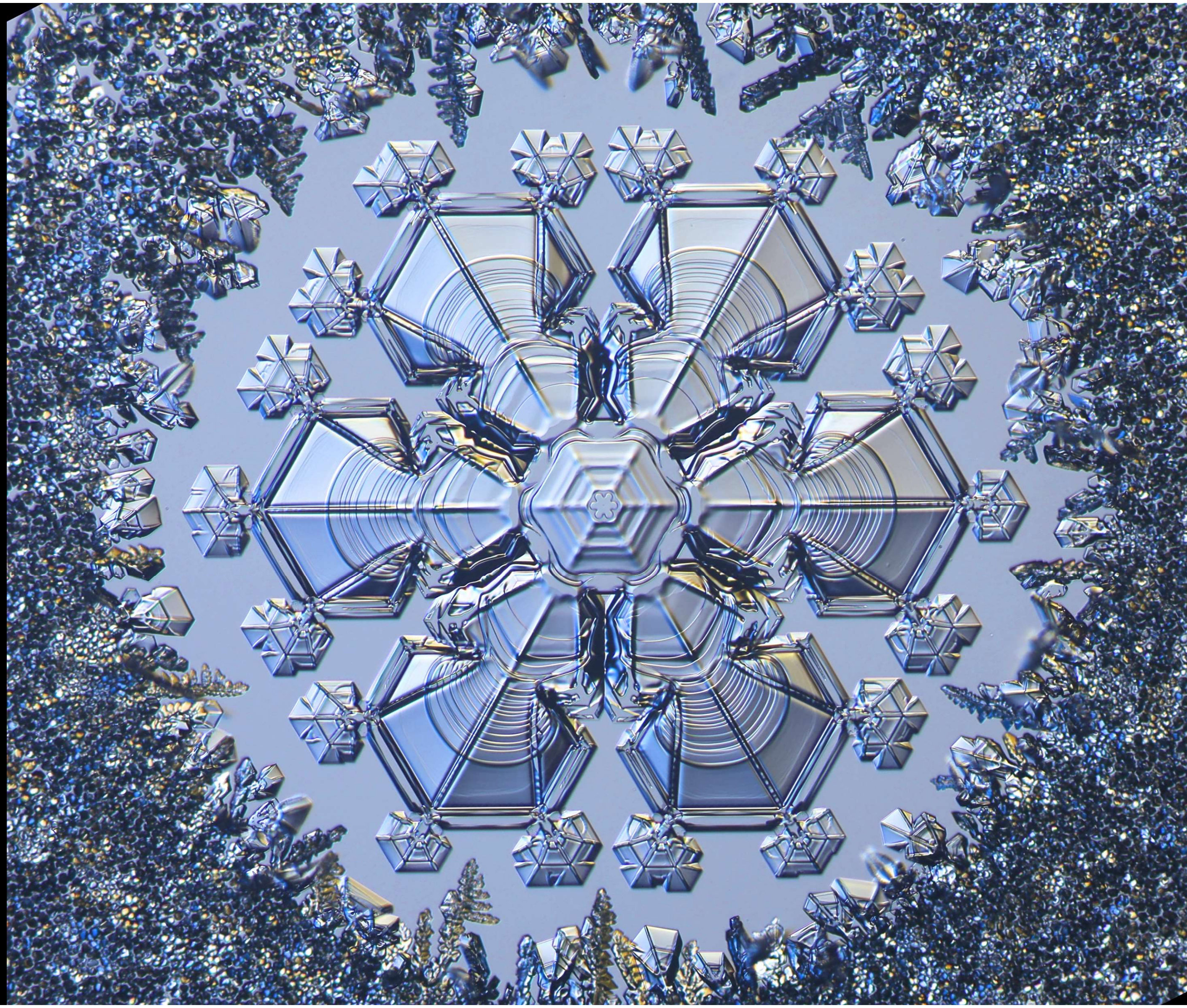


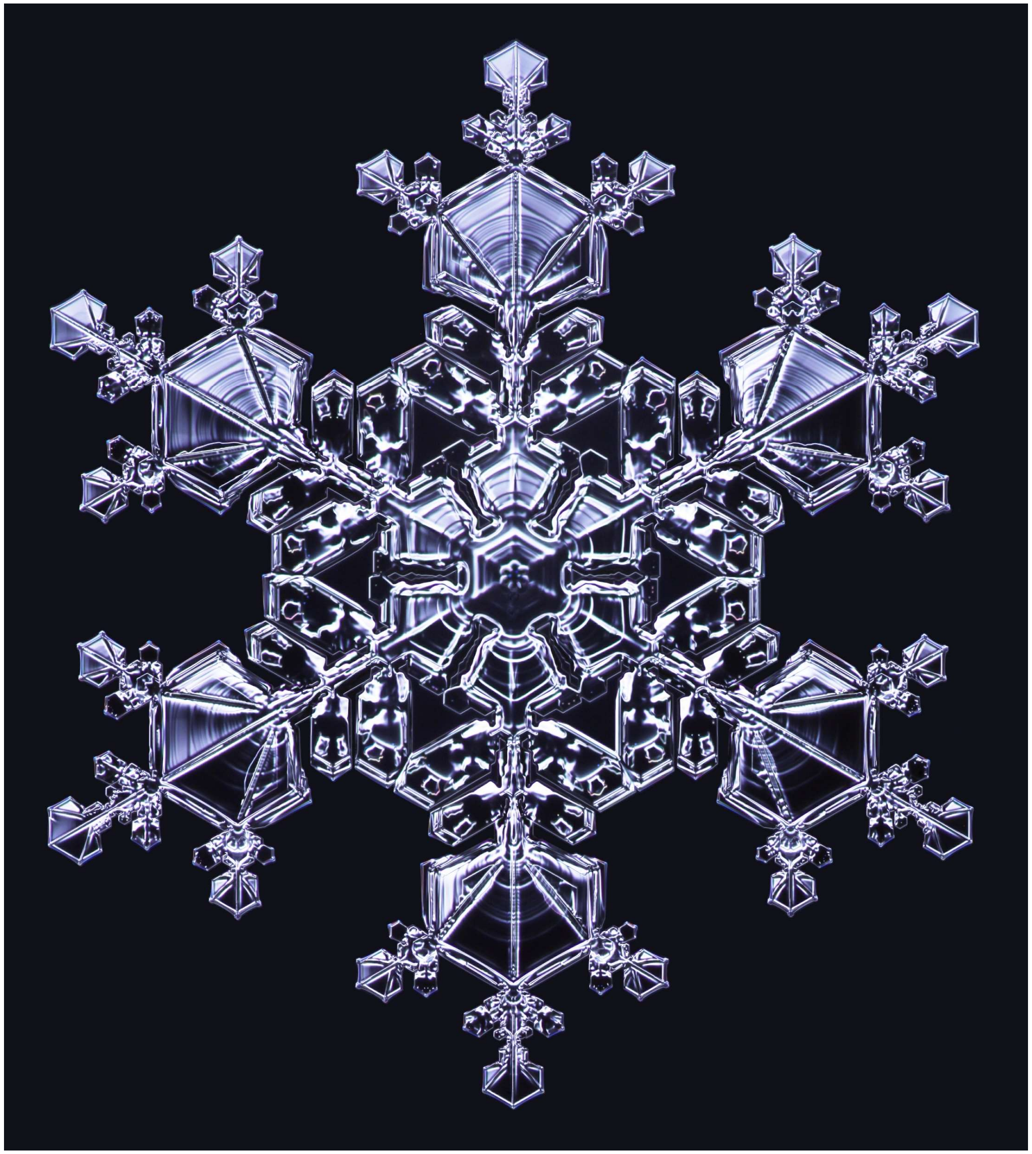


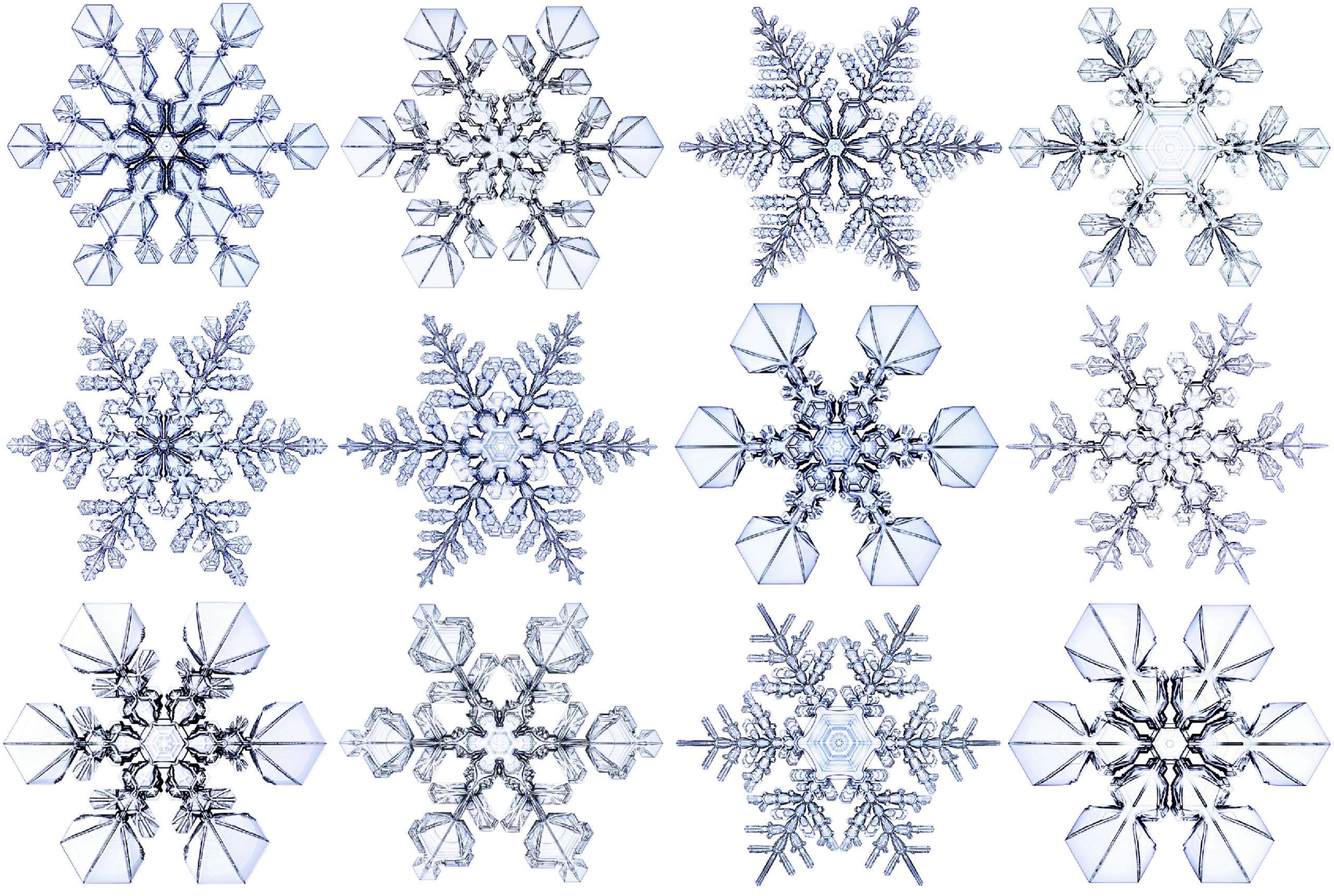


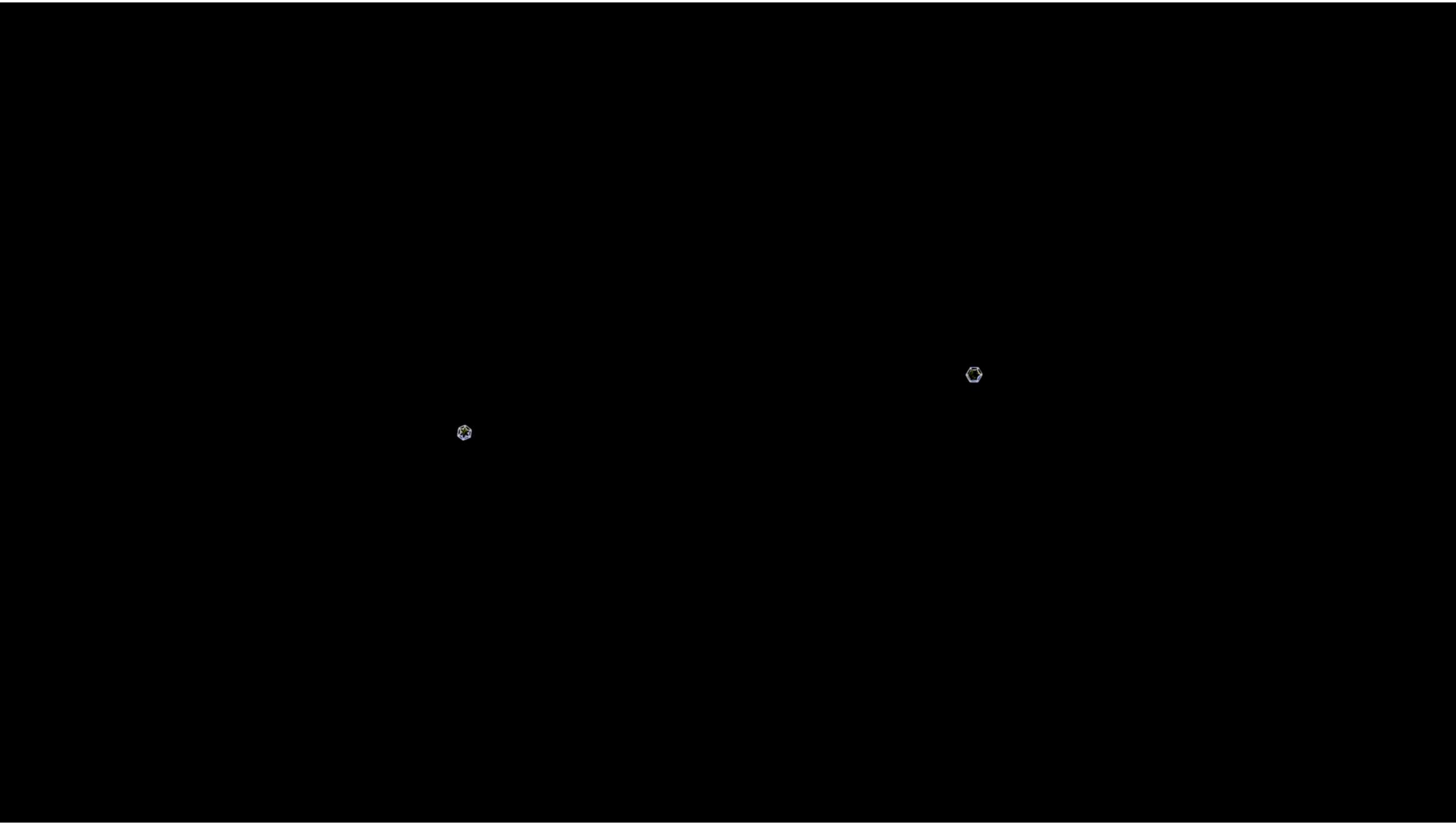
Real synthetic snowflakes … not computer models









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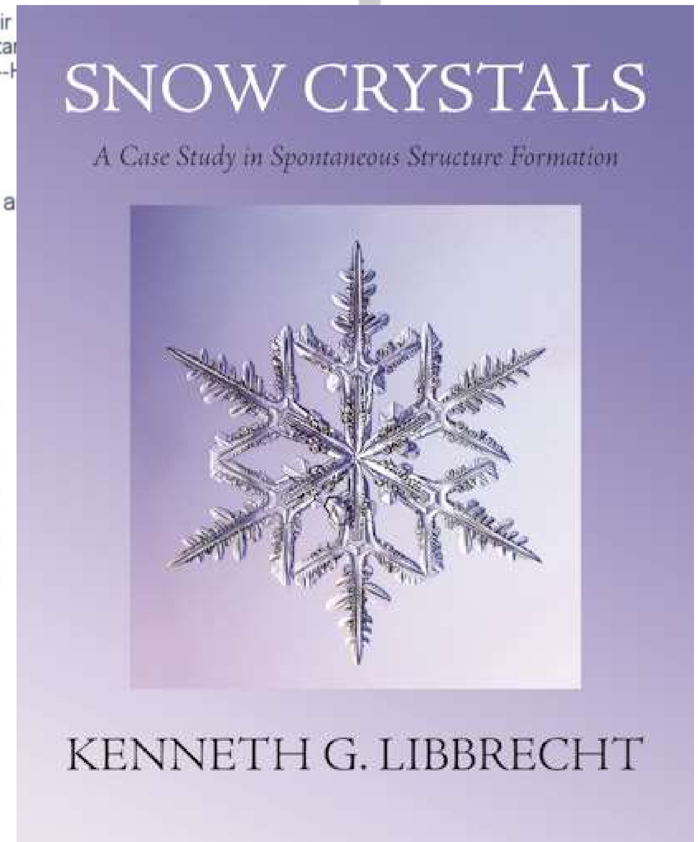
???
<< Check out this entertaining YouTube video about snow-crystal science, made by the good folks at *Veritasium*.
>> You can find the FULL story about the science of snow crystal formation in my magnum opus at right (weighting in at 456 pages)


SNOW CRYSTALS
A Case Study in Spontaneous Structure Formation
KENNETH G. LIBBRECHT