Reconstructed Morphology of Rough Ice Facets by Variable Pressure Scanning Electron Microscopy

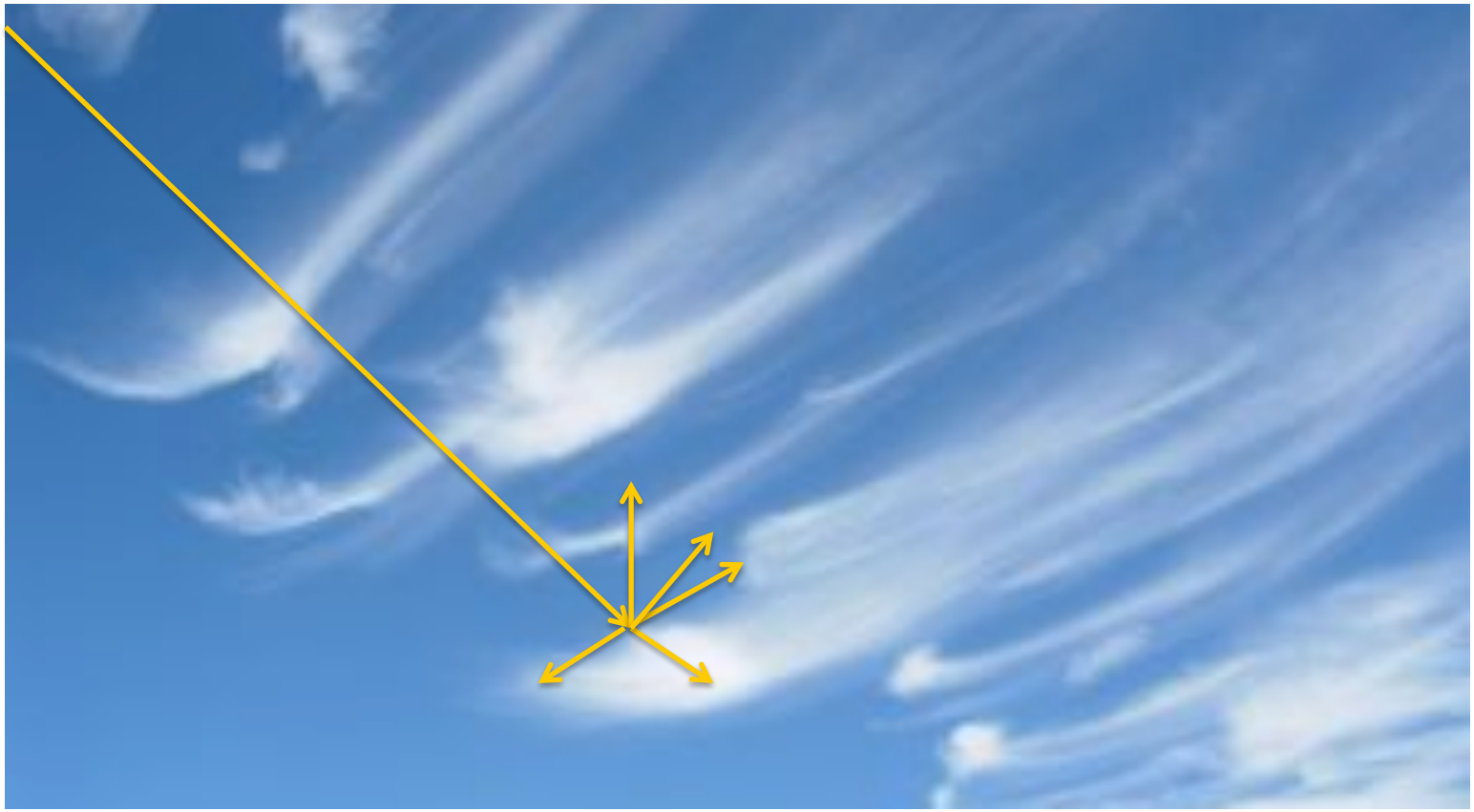
Steven Neshyba, David Roesel, Martina Roeselová, Ivan Gladich, Alicia Burns, Chelsea Jaeger, Drew Harrison

Outline

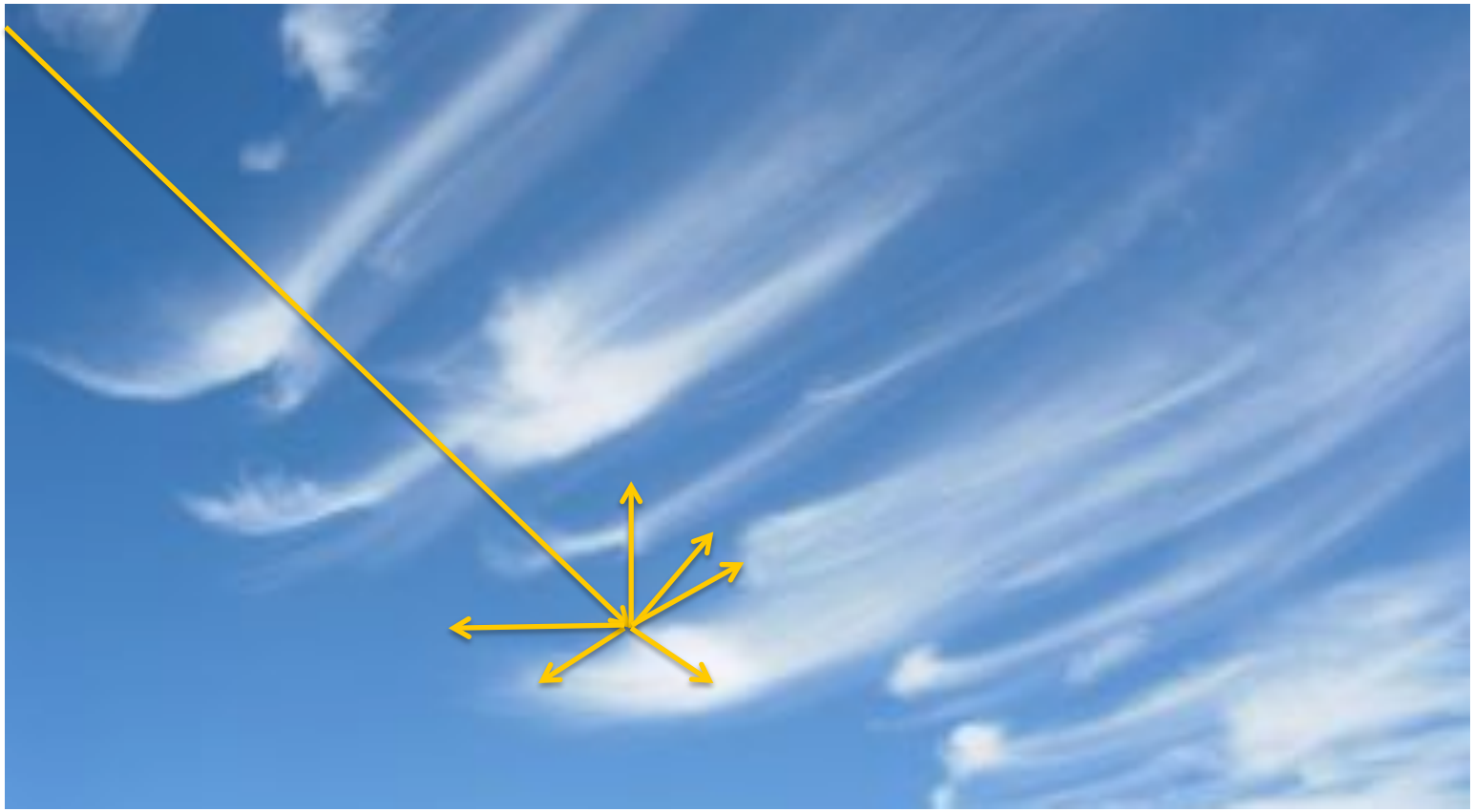
- Cirrus clouds in climate modeling and remote sensing
- Variable pressure scanning electron microscopy experiments
- Molecular dynamics simulations

Cirrus clouds are important for climate because of their light-scattering properties.

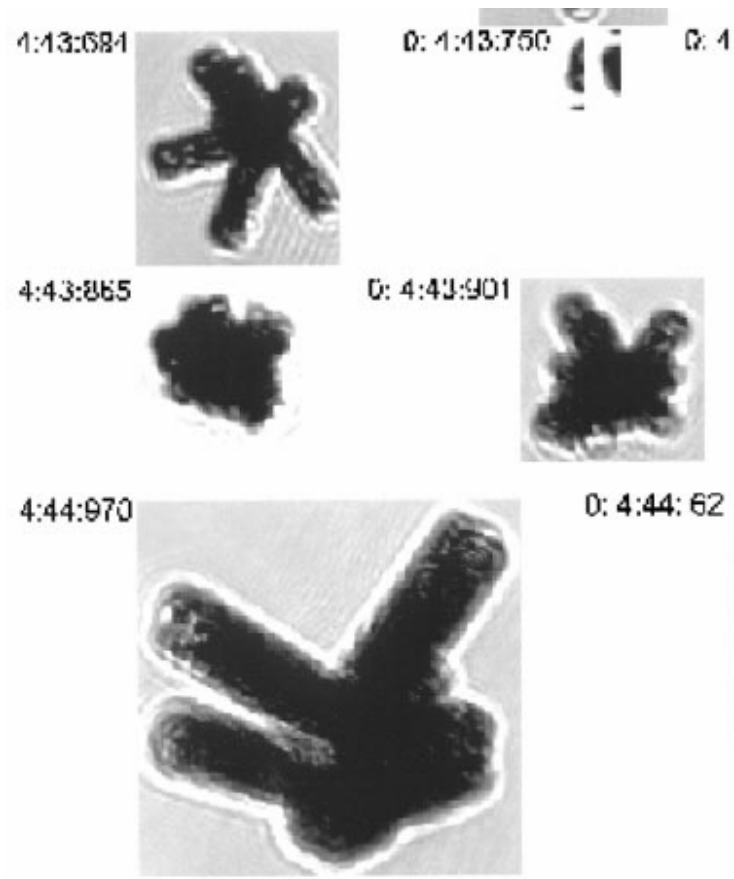
Ice crystals comprising cirrus clouds scatter light



But real cirrus ice crystals scatter more light back than theory predicts. Why?



Micrometer-scale facet roughening is capable of causing more back-scattering, but most in situ images lack sufficient resolution to tell if surfaces are rough



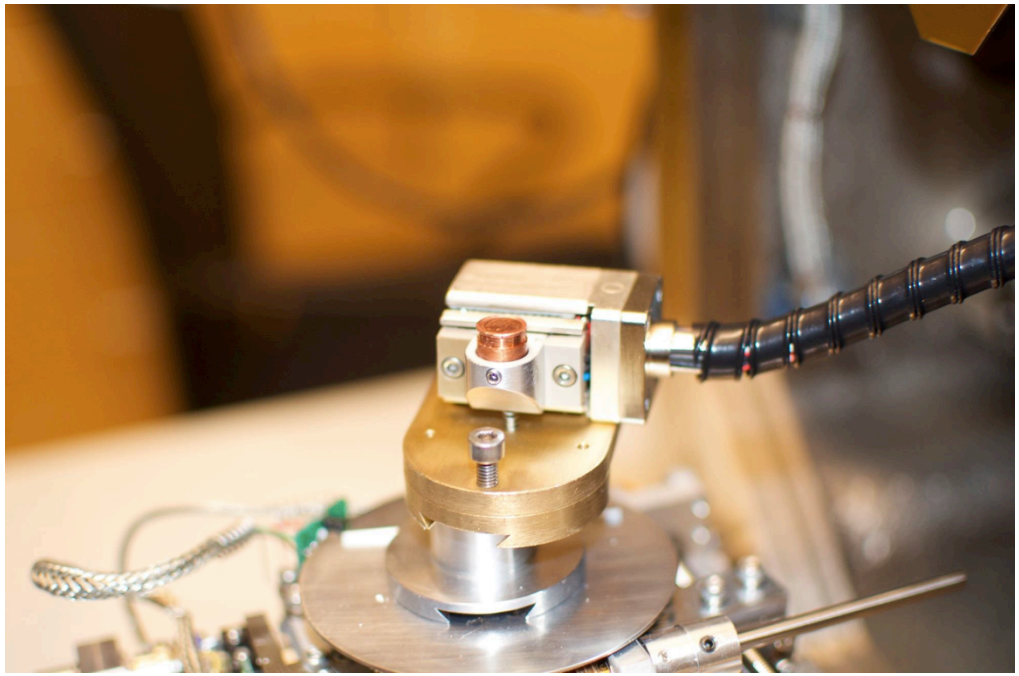
Airplane-deployed Cloud Particle Imager results (Gerber et al, J. Atmos. Sci. 2000)

VPSEM has much higher resolution, and can operate in the presence of water vapor – so you can grow ice as well as ablate it

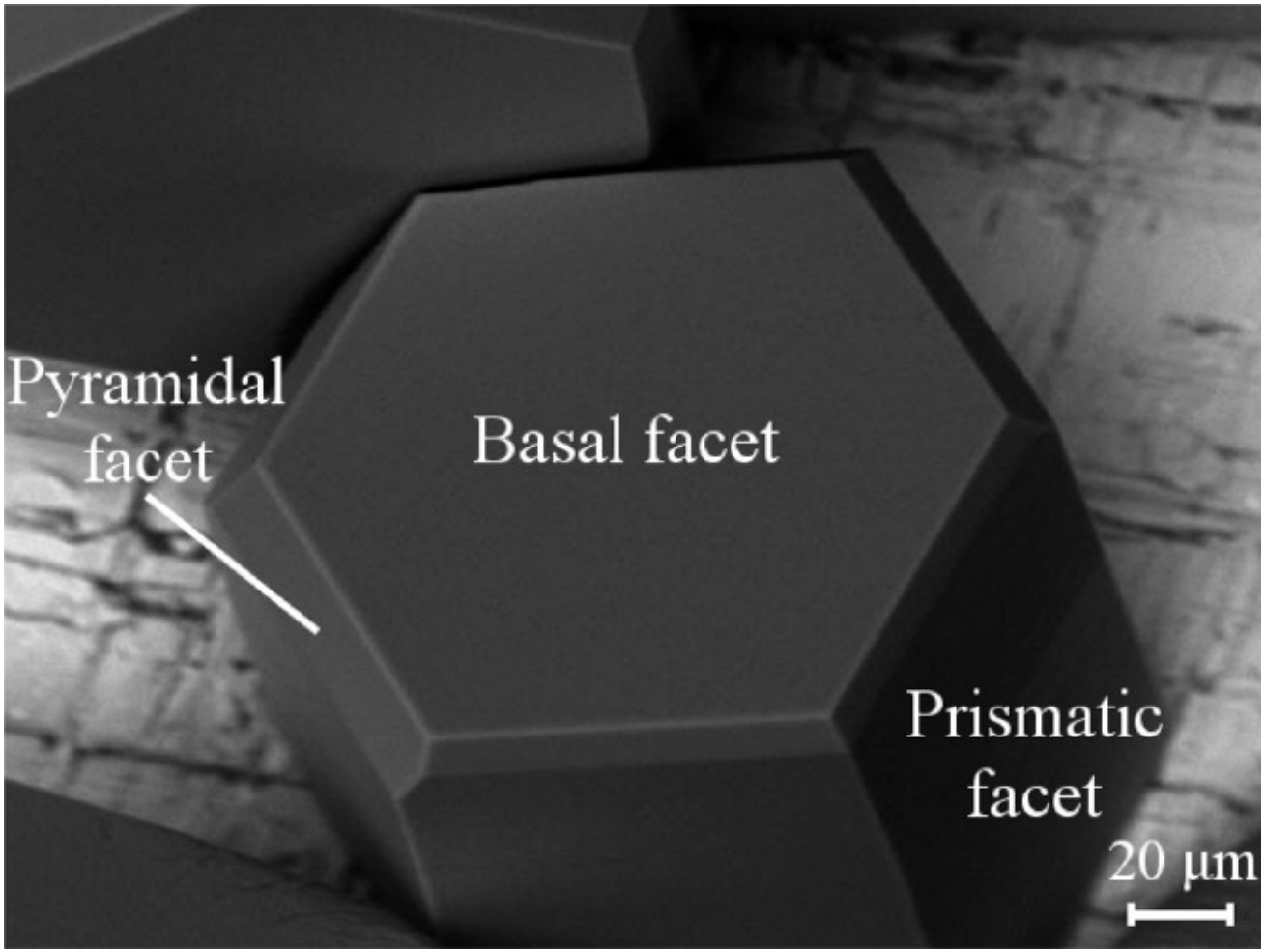
- Hitachi S-3400 N VP-SEM
- Deben Ultra-Cool MK3 w/ Peltier cooling element
- copper imaging stage

Chamber pressure is variable (up to 300 Pa) and determines $\mathbf{T_{eq}}$ of ice

$\mathbf{T_{stage}}$ is variable (down to -45°C) and determines the supersaturation of water vapor with respect to ice on the cold stage

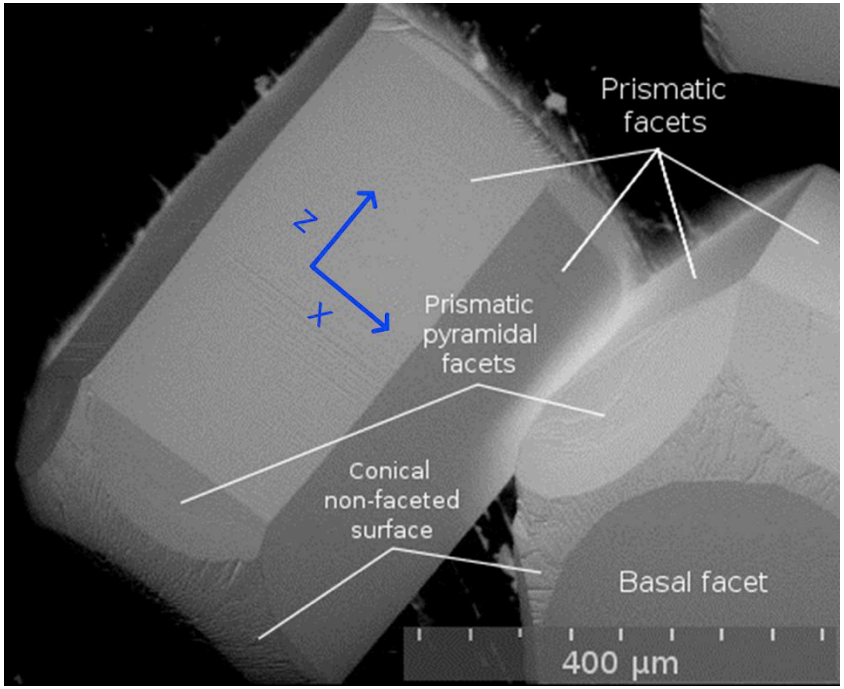


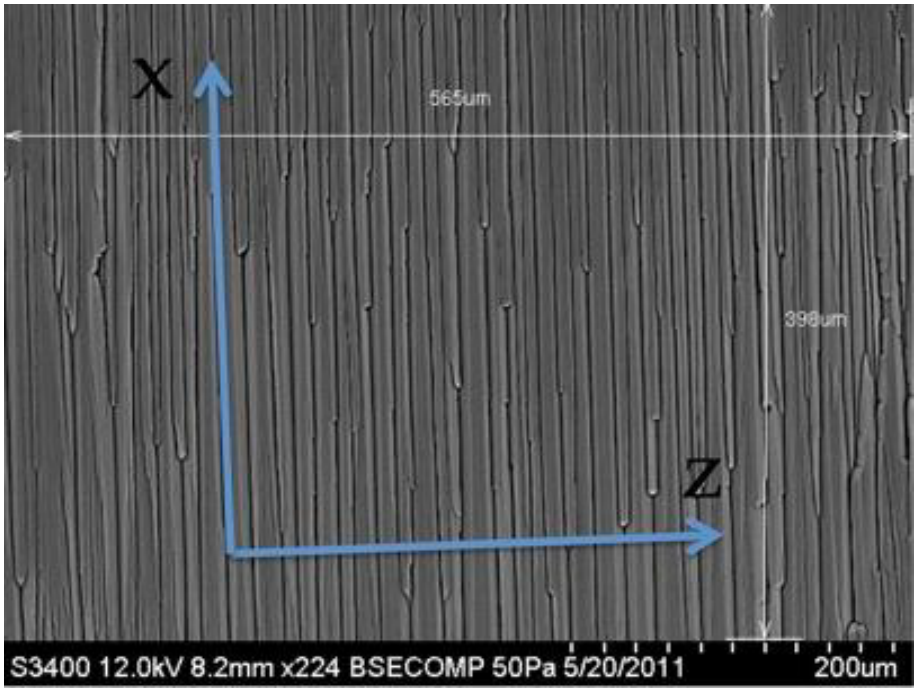
Here, $T_{stage} << T_{eq,}$ so supersaturation is positive, and the ice crystal is growing



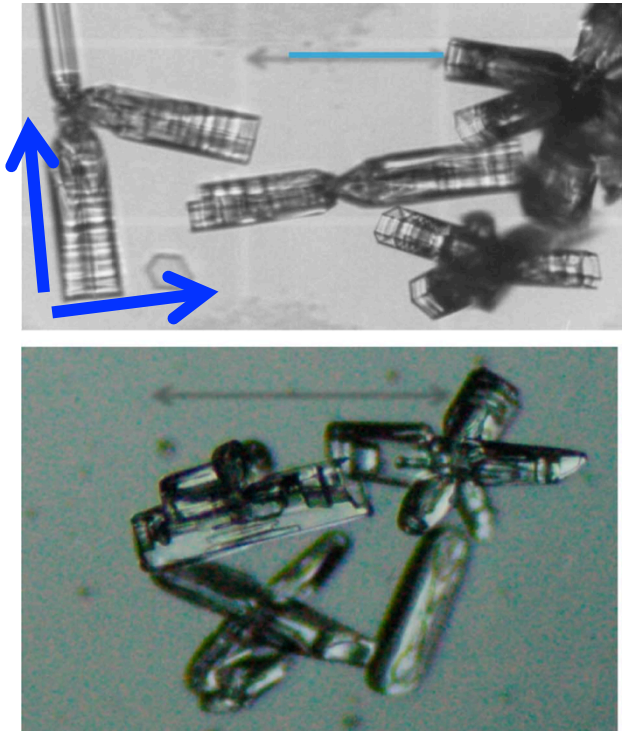
(JPC 2011)

Roughness on prismatic facets has distinct annular symmetry





Annular roughening symmetry on prismatic ice facets has been documented elsewhere

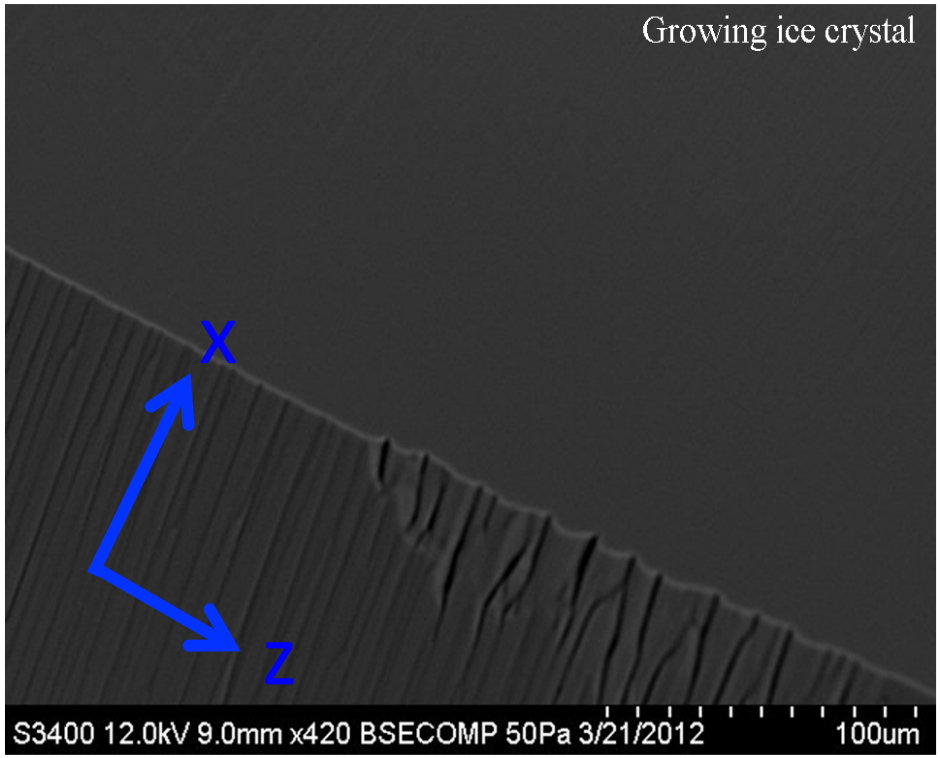


Atmospheric ice at South Pole and Summit, Greenland (Walden et al 2003, 2012).

Fig. 6. Etch pits formed on the prismatic plane (1010). The arrow indicates c-axis direction. (× 1000.)

Prismatic etch pits imaged using SEM and Formvar™ (Kuroiwa - J Glac. 1969)

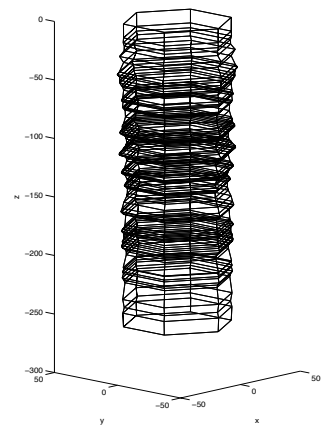
The flexibility to grow as well as ablate allows us to manipulate crystals in ways that admit quantitative analysis

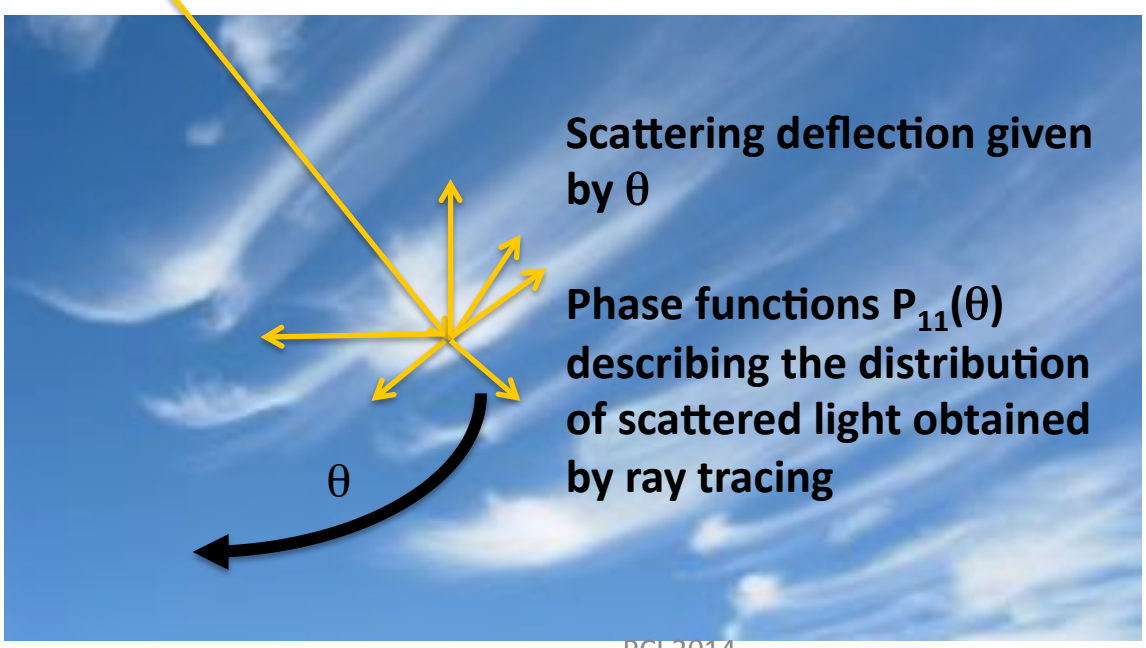


Adjacent prismatic ice facets (Pfalzgraff et al, 2010)

Model of a hexagonal ice column with VPSEM-derived roughening

Models can be used in ray-tracing studies to calculate scattering phase functions

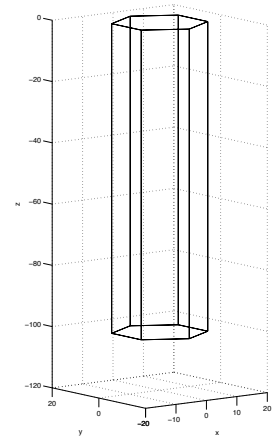


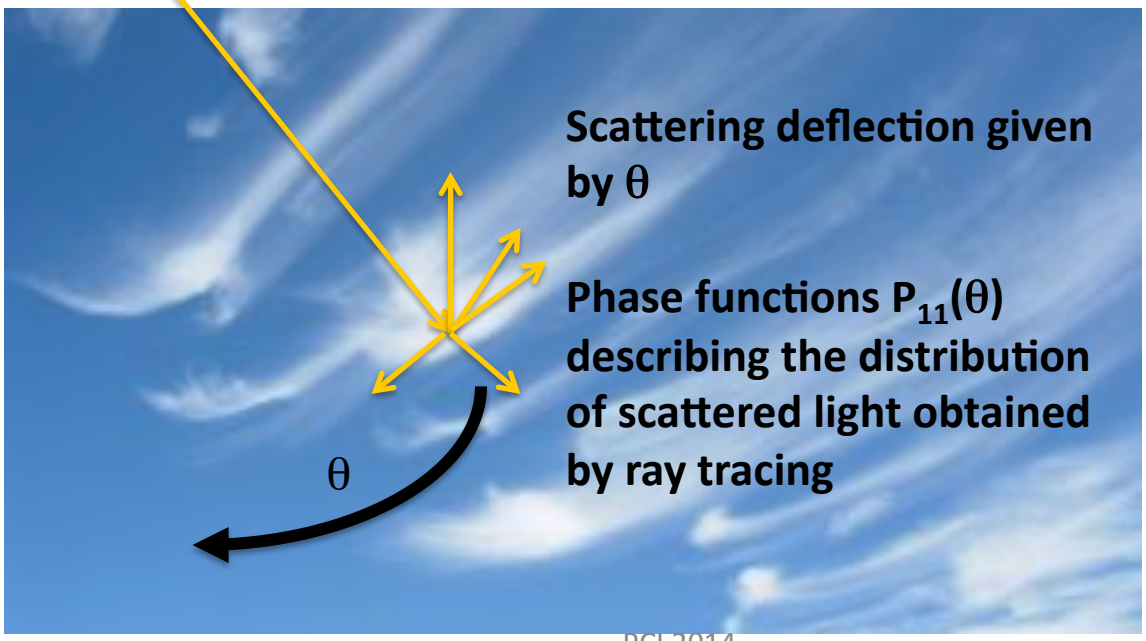


Scattering deflection given by θ

Phase functions $P_{11}(\theta)$ describing the distribution of scattered light obtained by ray tracing

Models can be used in ray-tracing studies to calculate scattering phase functions





Scattering deflection given by θ

Phase functions $P_{11}(\theta)$ describing the distribution of scattered light obtained by ray tracing

Resilience of certain halos to roughening has been an ongoing source of puzzlement in the remote sensing community

The prevalence of the 22° halo in cirrus clouds

Bastiaan van Diedenhoven

Columbia University, Center for Climate System Research, New York, NY

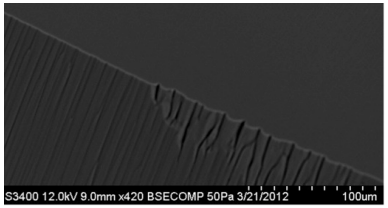
NASA Goddard Institute for Space Studies, New York, NY, USA

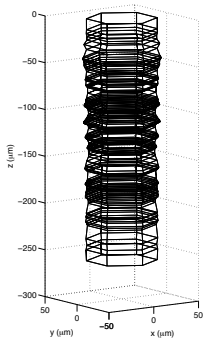
Abstract

Halos at 22° from the sun attributed to randomly-orientated, pristine hexagonal crystals are frequently observed through ice clouds. These frequent sitings of halos formed by pristine crystals pose an apparent inconsistency with the dominance of distorted, non-pristine ice crystals indicated by in situ and remote sensing data. Furthermore, the 46° halo, which is associated with pristine hexagonal crystals as well, is observed far less frequent than the 22° halo. Considering that plausible mechanisms that could cause crystal dis-

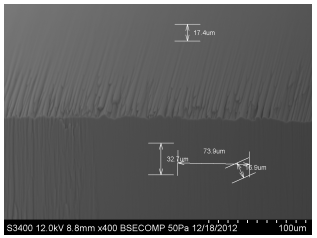
Annular-symmetric roughening retains the 22° halo

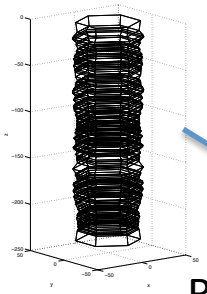
I
ablating;
deep





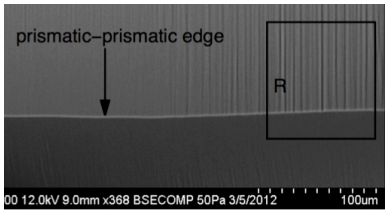
II
growing;
deep

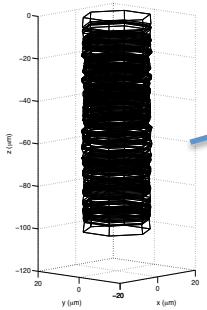




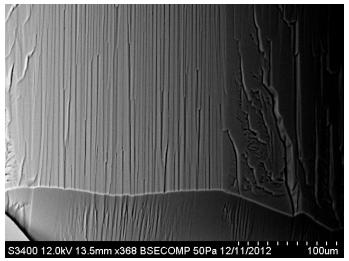
Ray tracing

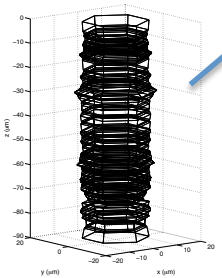
III
growing;
shallow

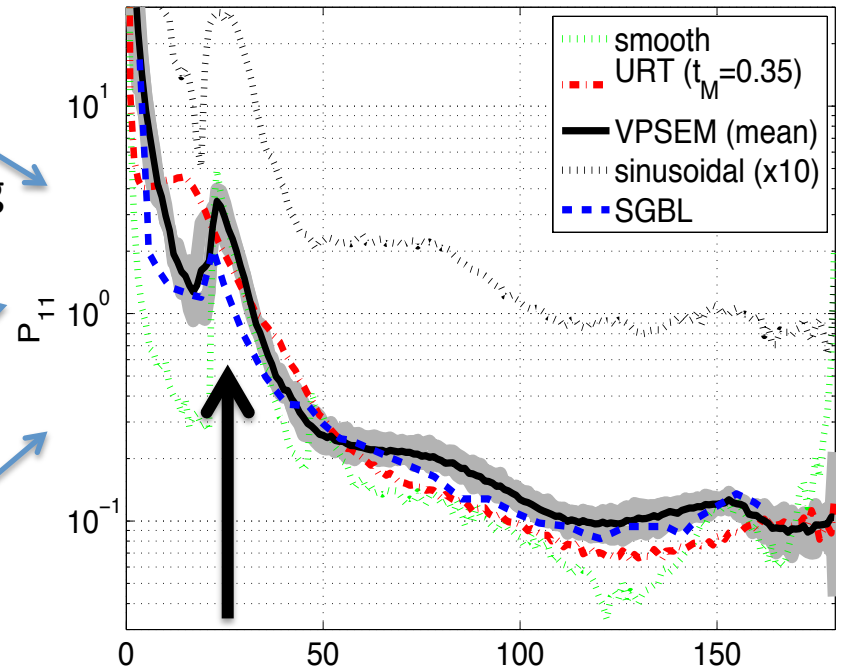




IV
growing;
shallow



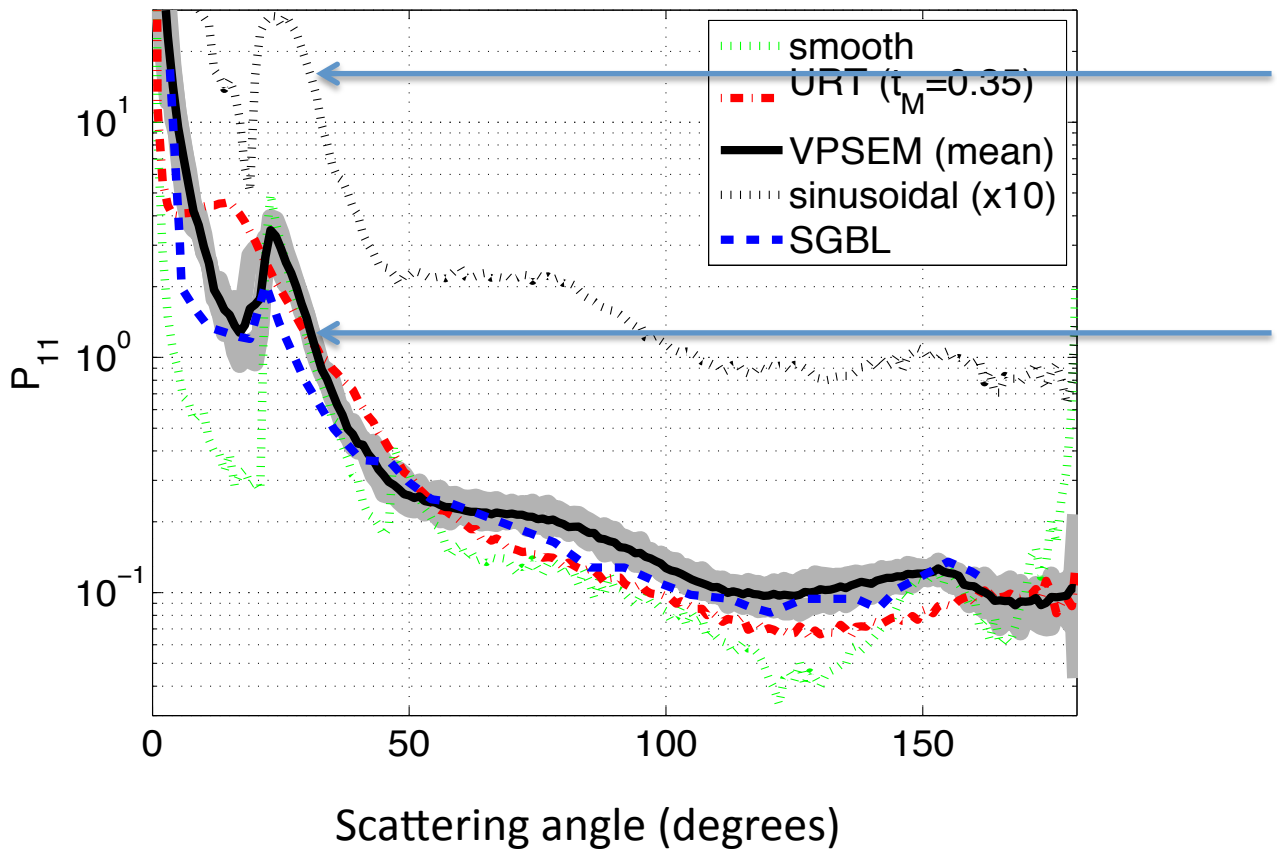


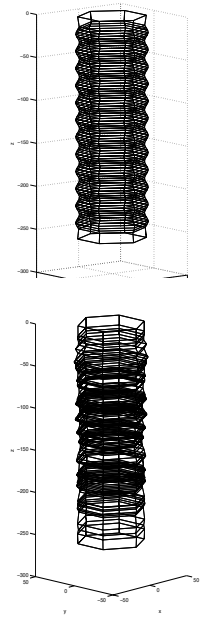


Scattering deflection angle (degrees)

JGR 2013

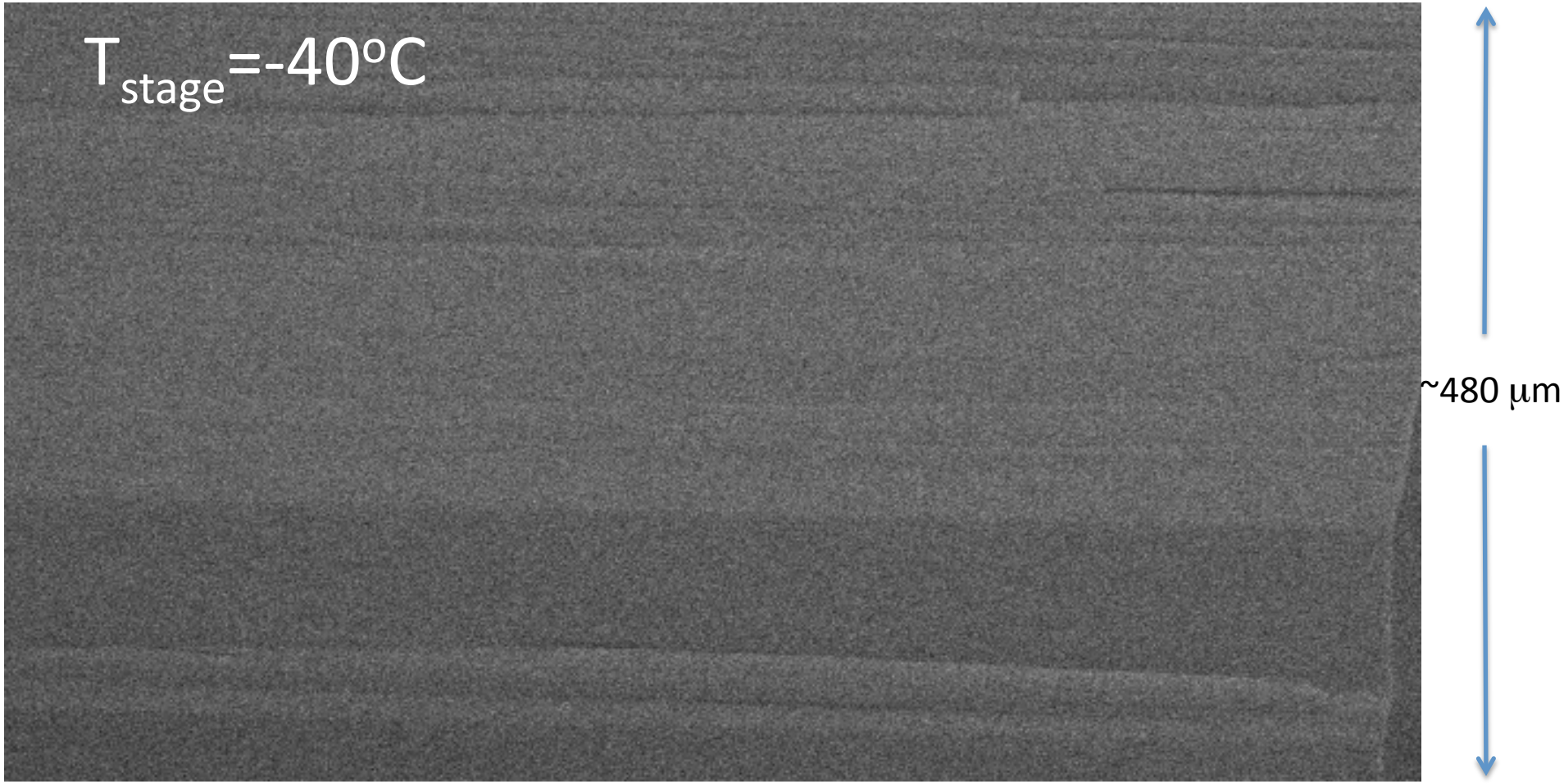
Actually the resilience of the 22° halo seems to be due mostly to the symmetry, rather than the details, of the roughening



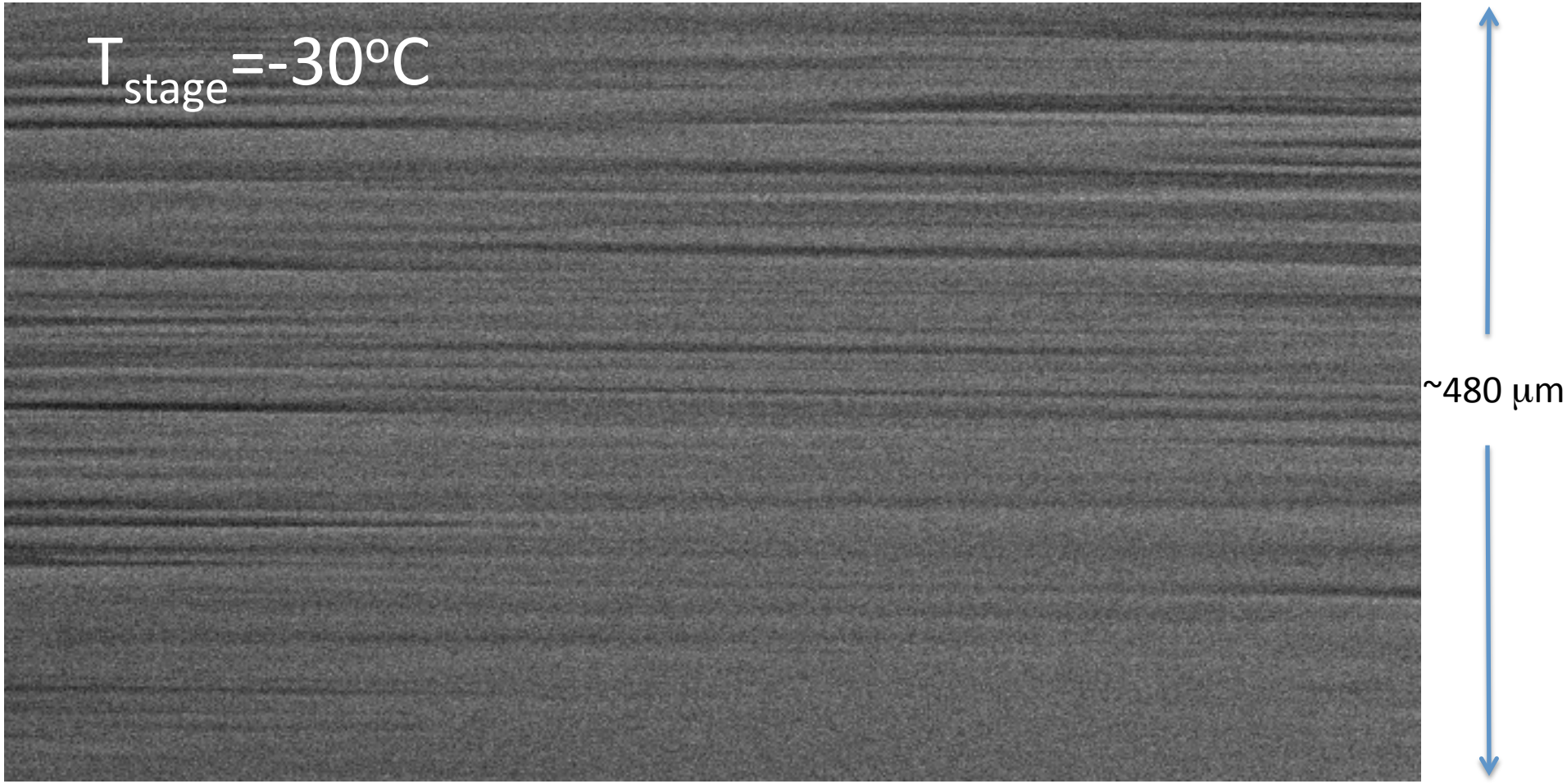


How does the roughening depend on atmospherically important variables?

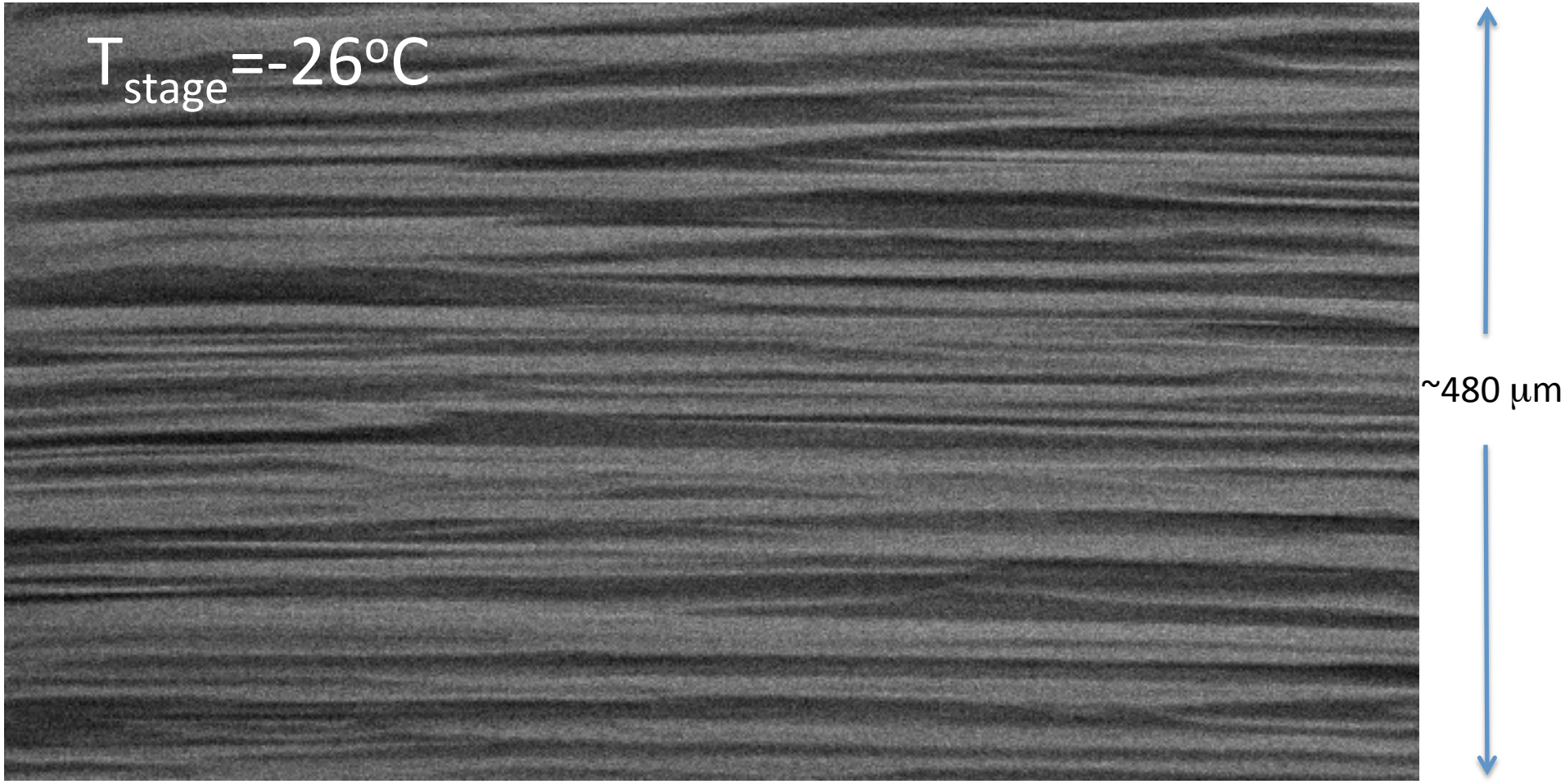
Fixing chamber conditions to T_{eq}=-26°C (P=100 Pa) while varying T_{stage} effectively varies supersaturation



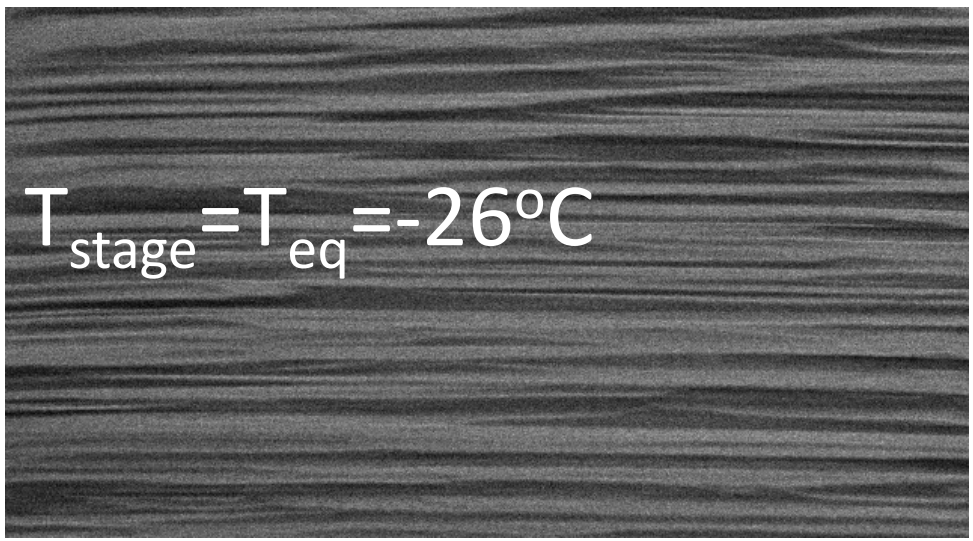
Fixing chamber conditions to T_{eq}=-26°C (P=100 Pa) while varying T_{stage} effectively varies supersaturation

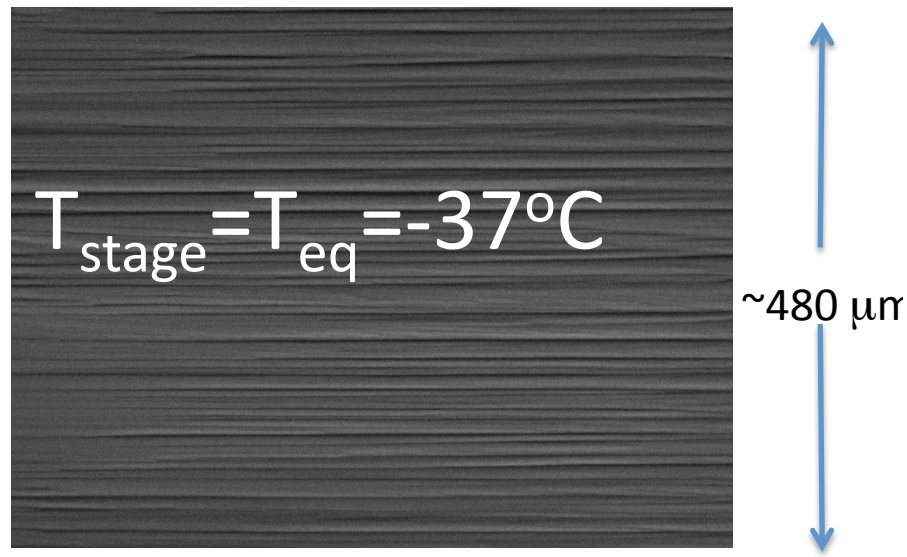


Fixing chamber conditions to T_{eq}=-26°C (P=100 Pa) while varying T_{stage} effectively varies supersaturation



If you set $T_{stage}=T_{eq}$, you can compare the effect of temperature on the equilibrium roughness

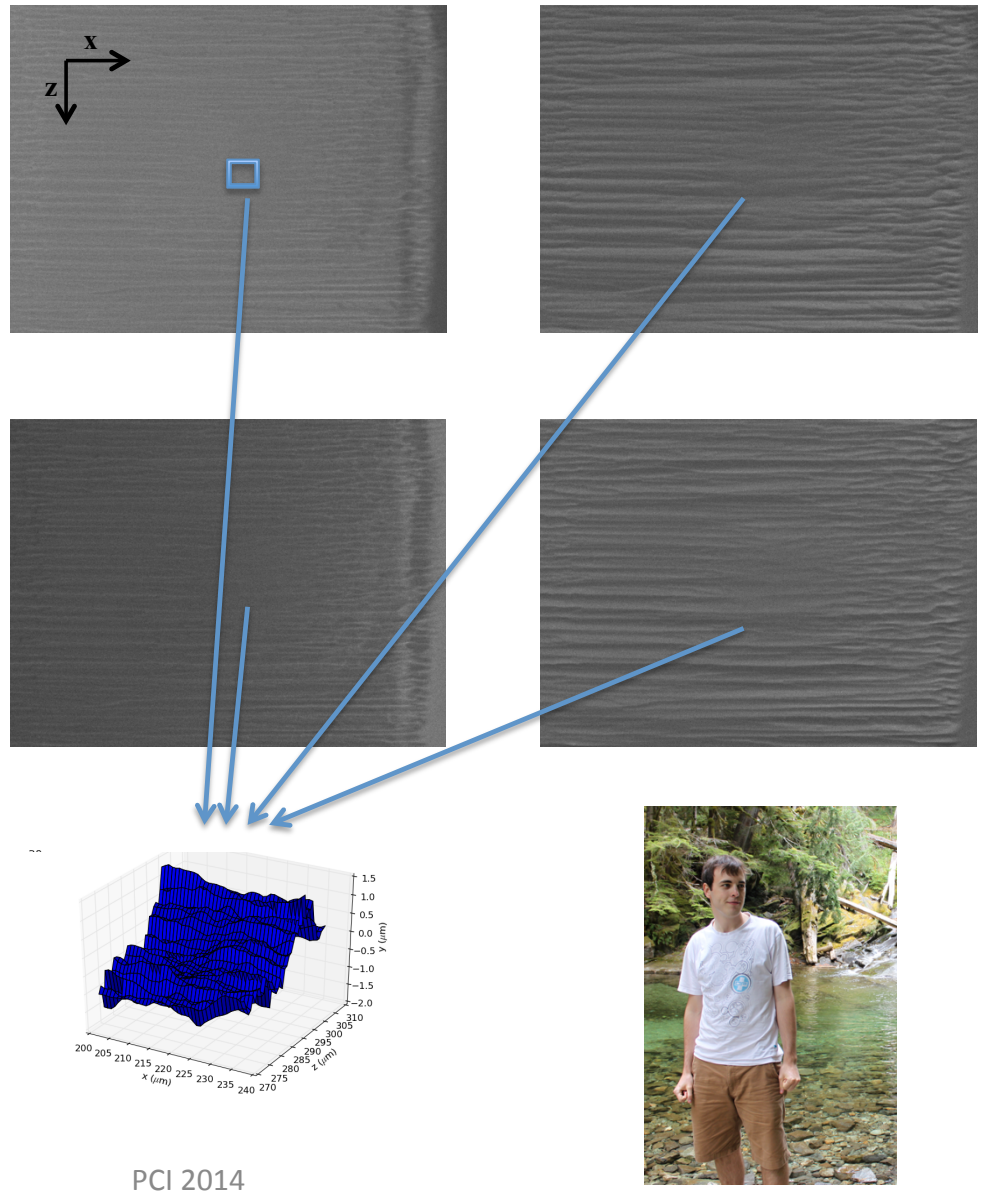




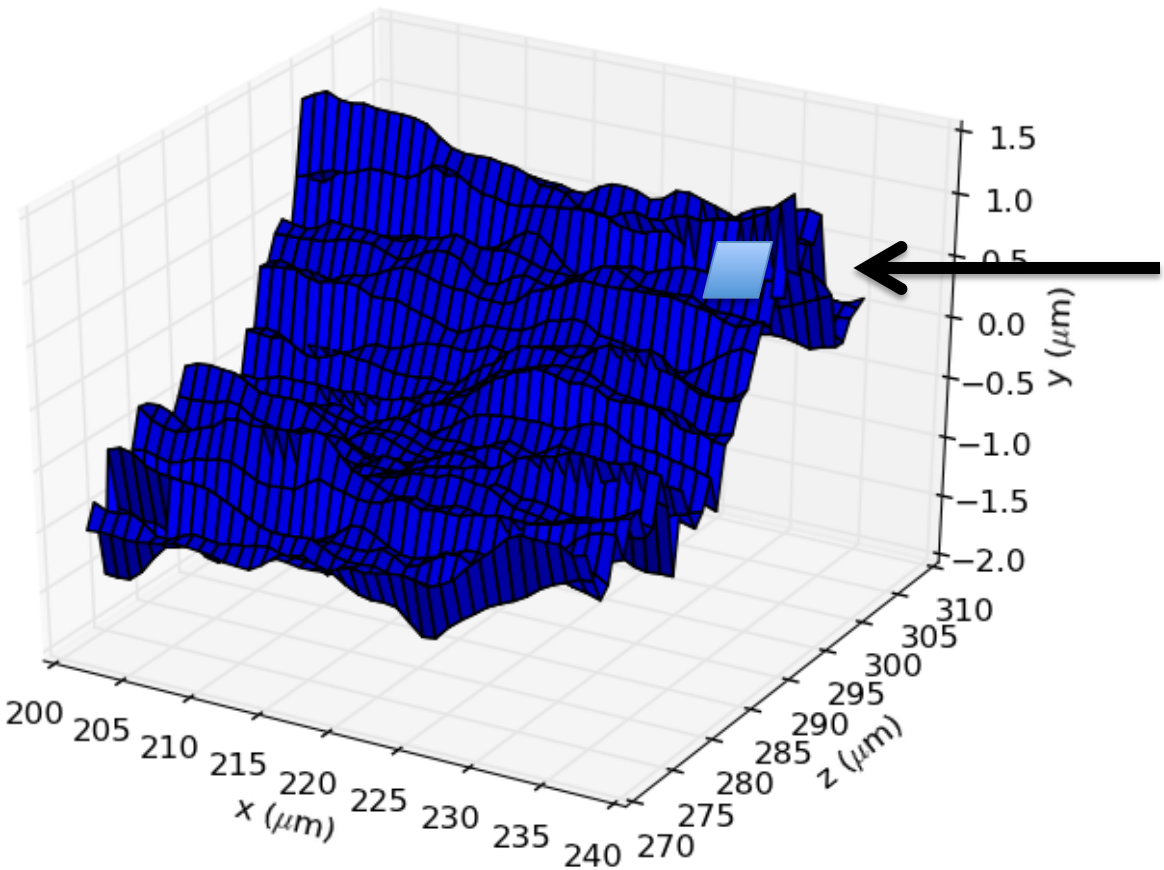
How do you quantify all this?

Parallax vision: Spiders have 8 eyes SEM has 4 detectors

Four near-simultaneous VPSEM micrographs

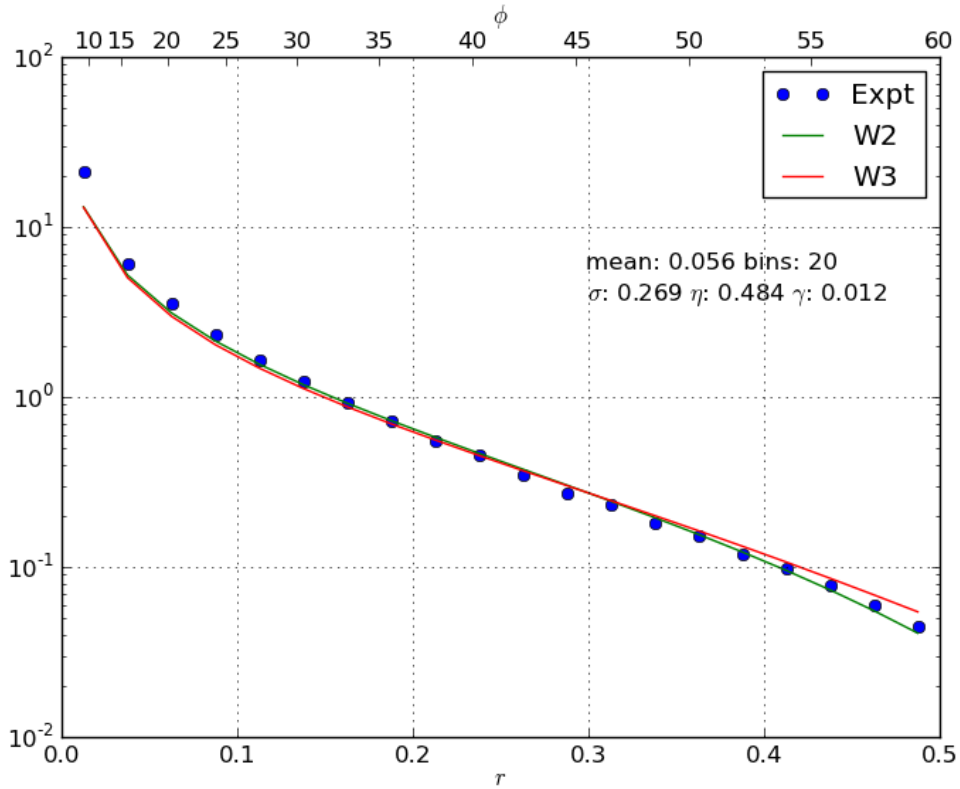


Calculation of a tilt angle for each of 640x480 pixels permits vigorous statistical analysis



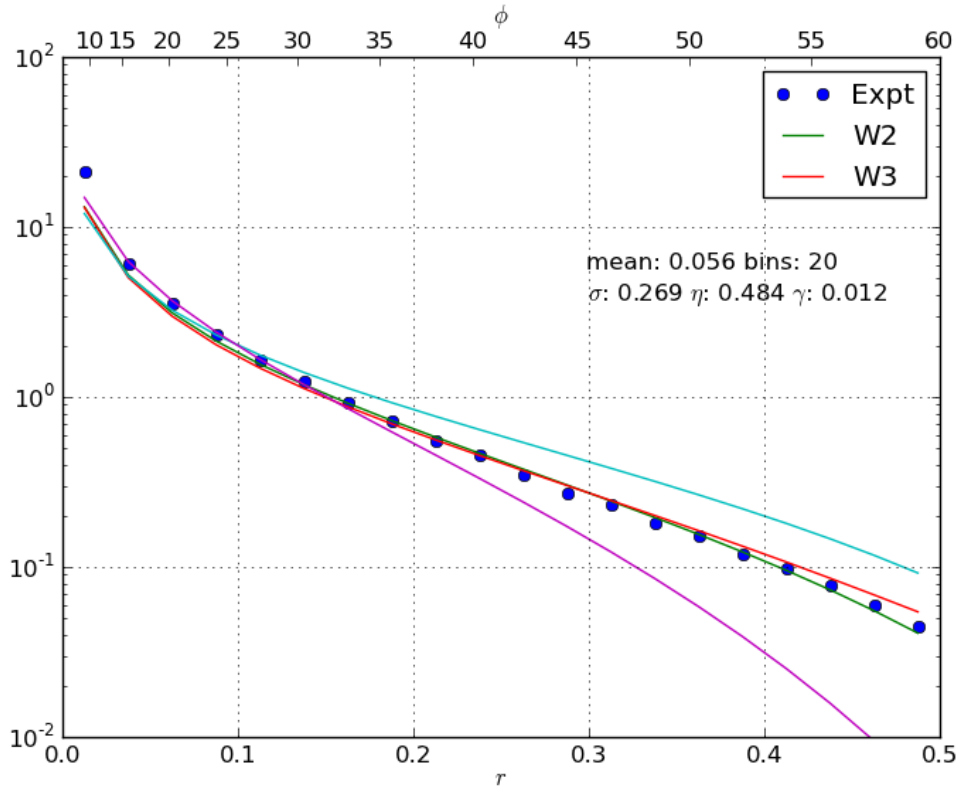
Each pixel has a tilt angle (ϕ) with respect to normal

Generate probability density functions of tilt angle in the z-direction, fit to Weibull distribution

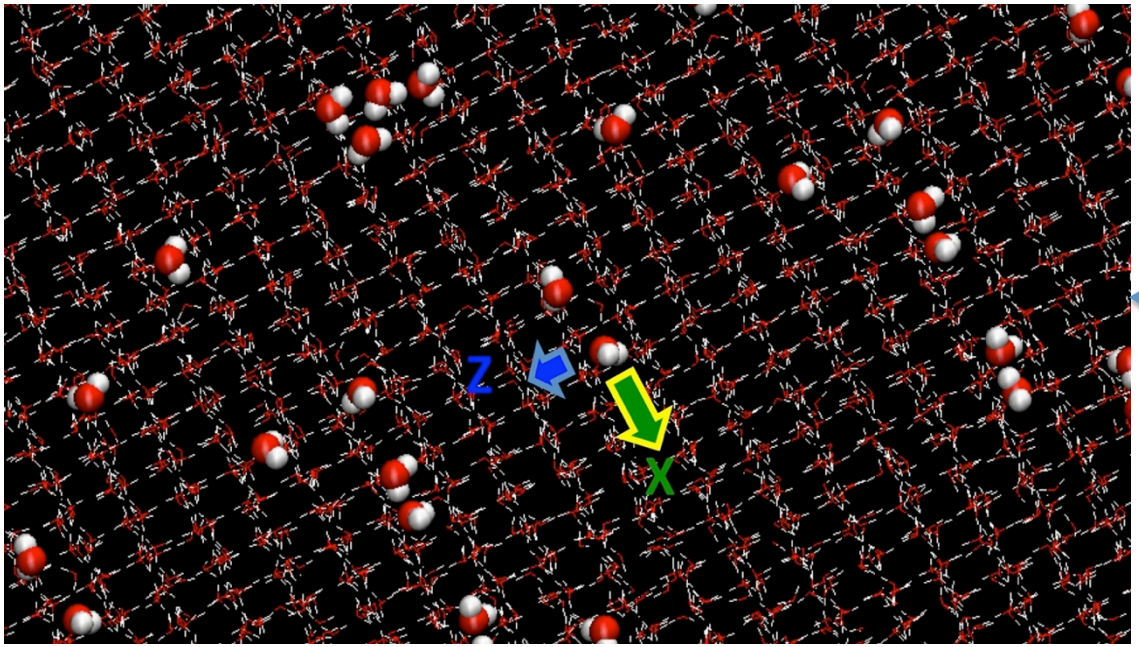


$$W_2 = \frac{2\eta}{\sigma^2\mu^3}\left(\frac{\mu^{-2}-1}{\sigma^2}\right) / \exp\left[\left(-\frac{\mu^{-2}-1}{\sigma^2}\right)^{\eta}\right] \quad \text{where} \quad \mu = 1 - r$$

When σ is 20% too big or η is 20% too big

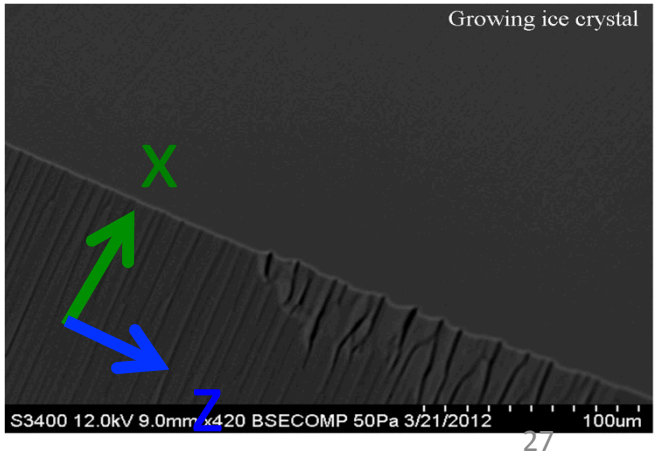


Classical molecular dynamics (MD) simulations predict anisotropic self-diffusion of admolecules on exposed prismatic facets

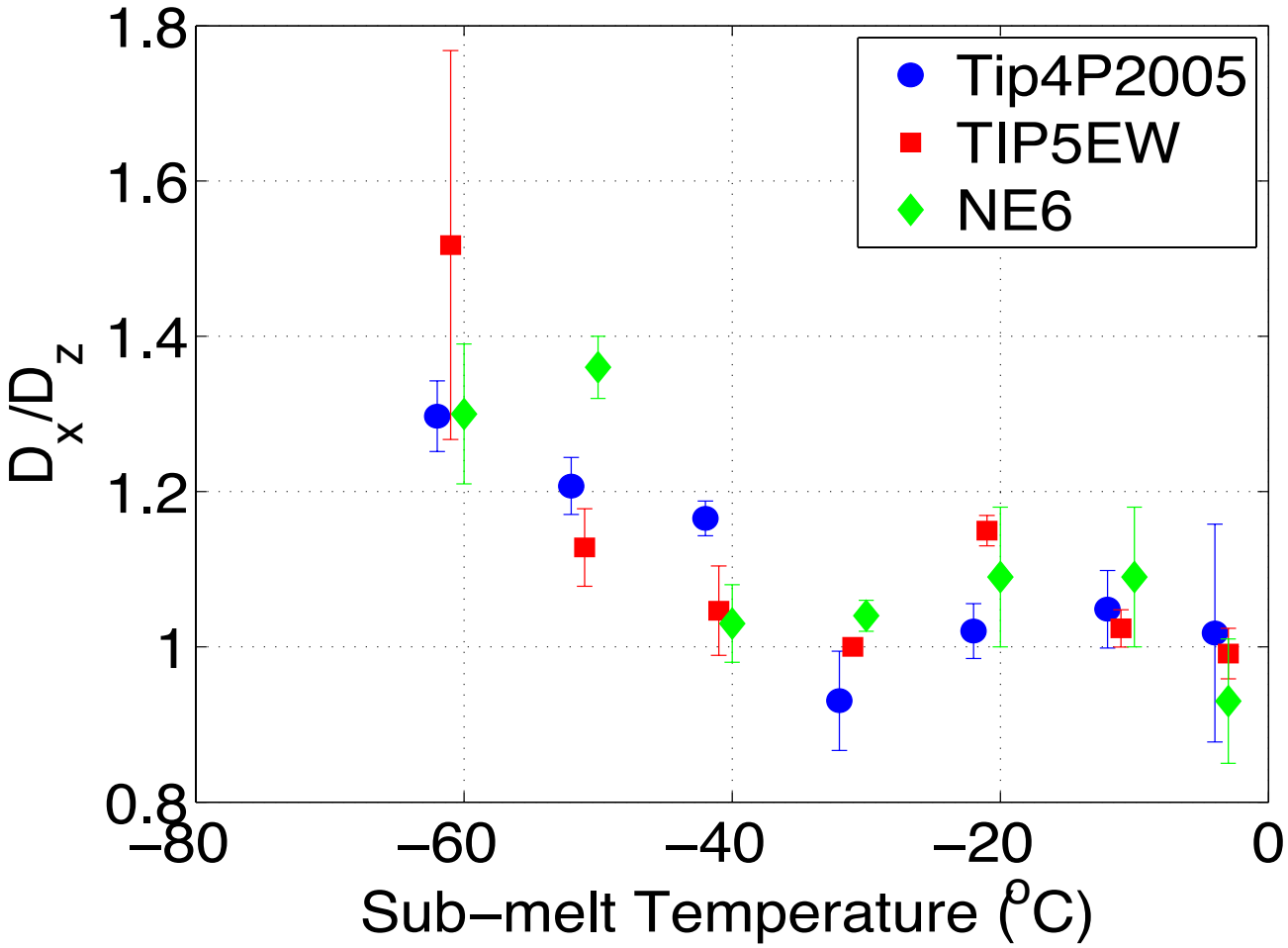


Plan view of the prismatic surface showing direction of diffusion anisotropy atop NE6 ice

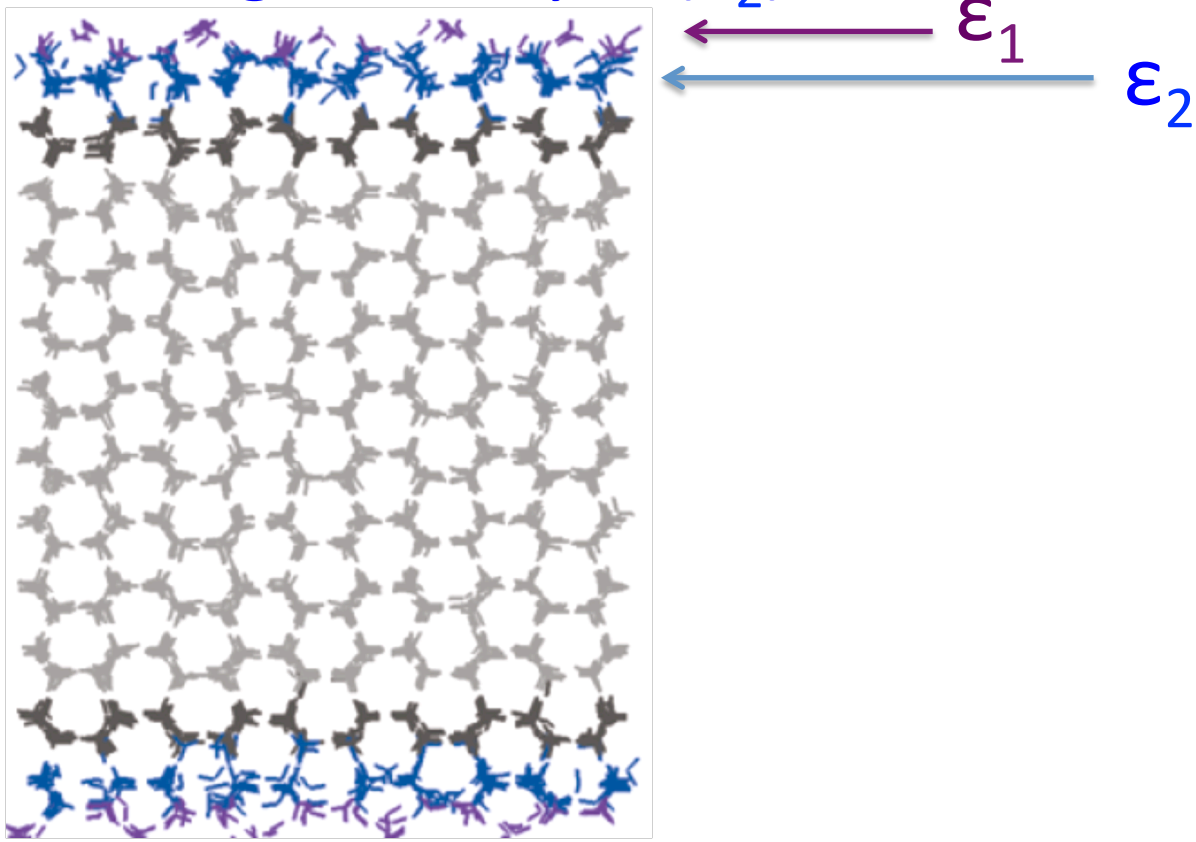
Which is the same orientation trans-prismatic strands seen in VPSEM



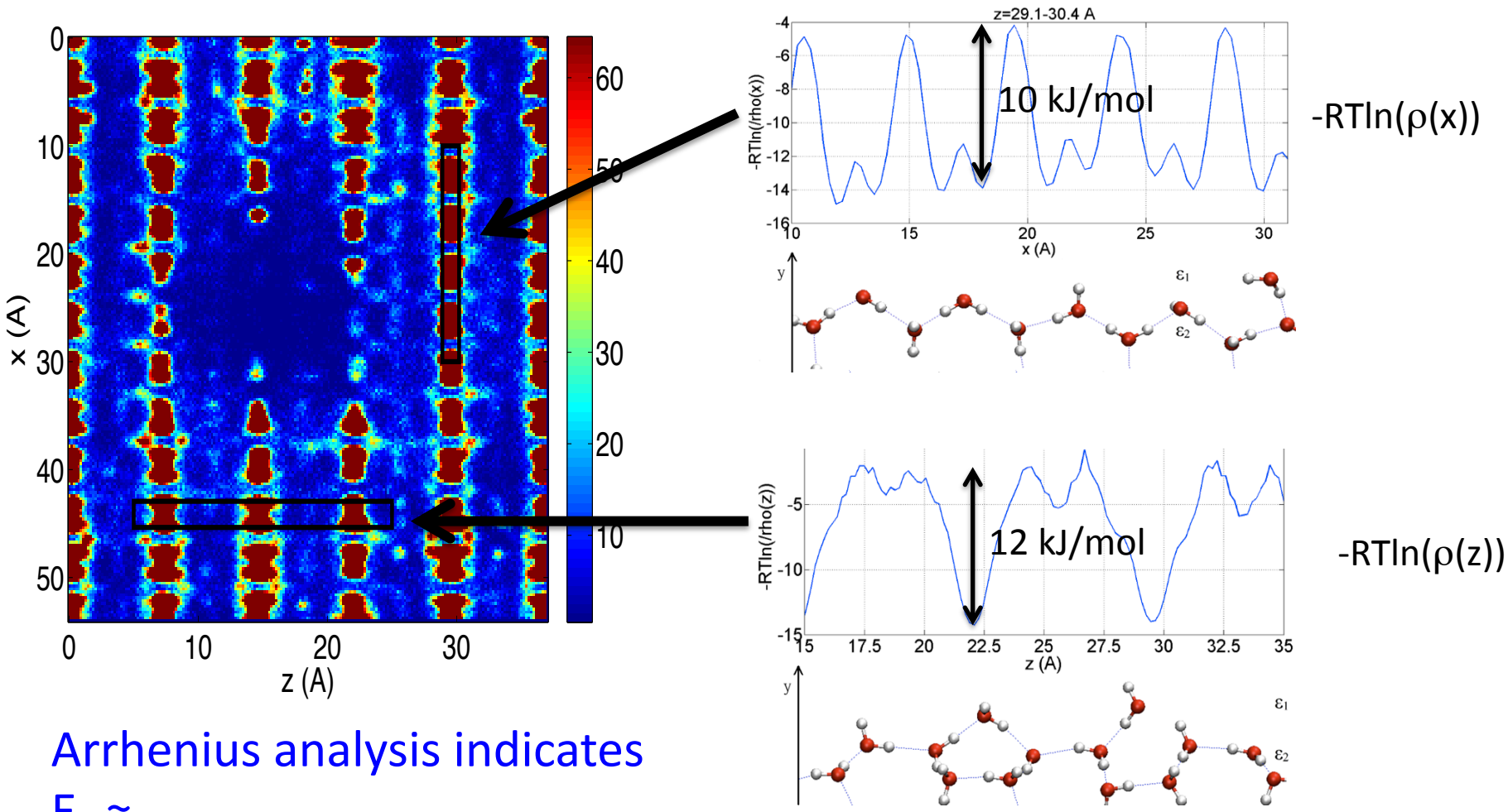
The diffusive anisotropy occurs at low temperature in three water models. In all three, a transition to isotropic diffusion occurs at about -40°C.



Why is diffusion anisotropic at low temperature? If the quasi-liquid layer (QLL) is not too thick, mobile admolecules (ε_1) may respond to the structure of the underlying, more rigid sublayer (ε_2).



Admolecules (ε_1) prefer interstitial sites atop ε_2



Arrhenius analysis indicates

$E_a \approx$

Conclusions

- Annular-symmetric roughening explains increased backscattered intensity and halo resilience (and possibly polarization remote sensing experiments)
- Roughening increases as supersaturation approaches zero (to a climatically significant degree)
- Cold roughening is more orderly (which could hold clues as to its origin)
- Faster diffusion in the x-direction is due to admolecule preference for interstitial positions atop a more ordered part of the QLL, and is entropically driven (which could point to an explanation for the transition to isotropic diffusion)

Acknowledgements

- NSF 1306366 & University of Puget Sound
- Roesel[1] and Harrison[2] (SEM)
- Oswald[2], Bowens[2], & Gladich[3] (MD)
- Roeselová[4], Burns[2], and Jaeger[2] (analysis)

[1]Czech Technical University in Prague, Faculty of Nuclear Sciences and Physical Engineering, Czech Republic

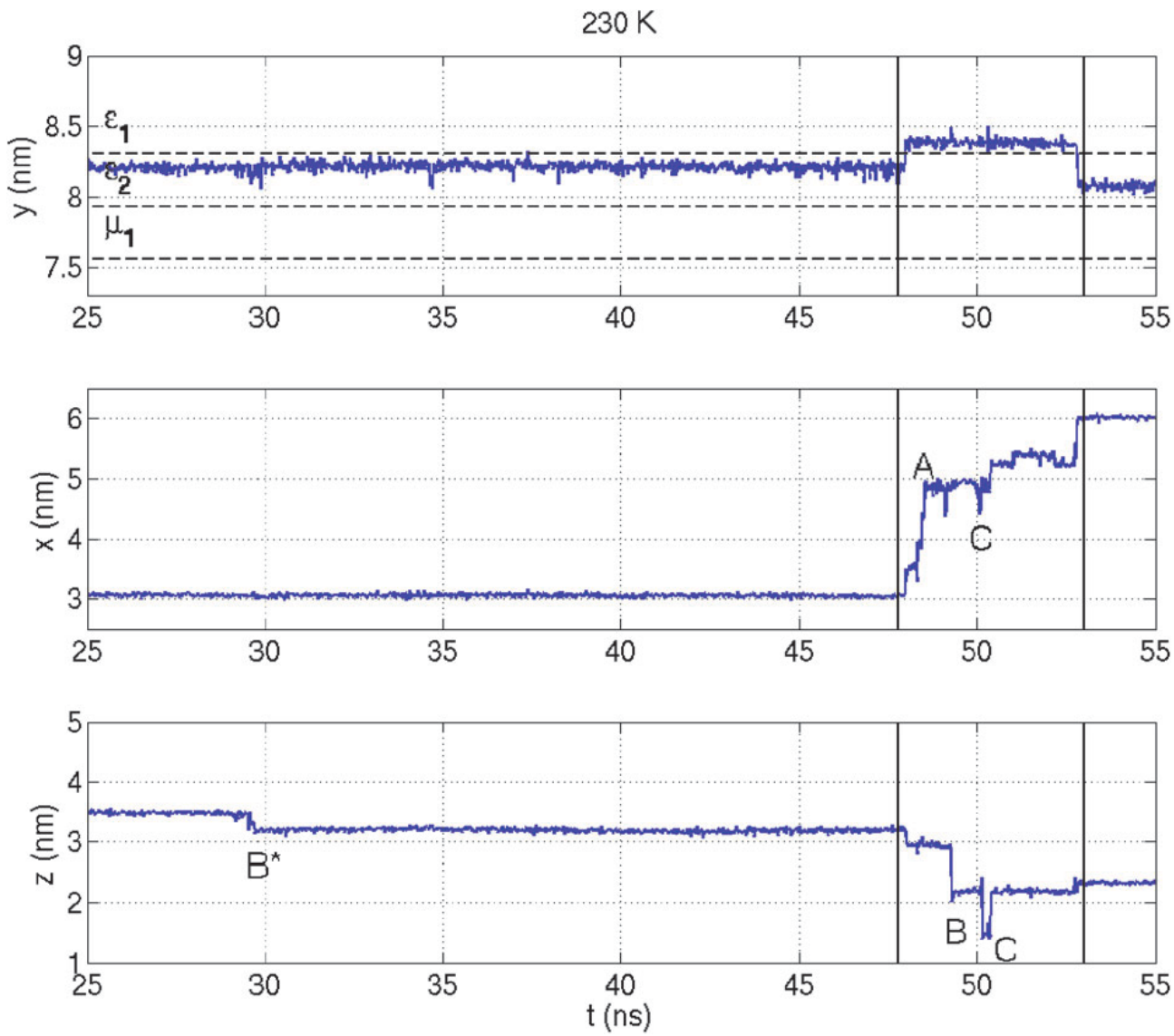
[2]University of Puget Sound, Tacoma, Washington, USA

[3]Ivan Gladich, International School for Advanced Studies (SISSA), Trieste, Italy

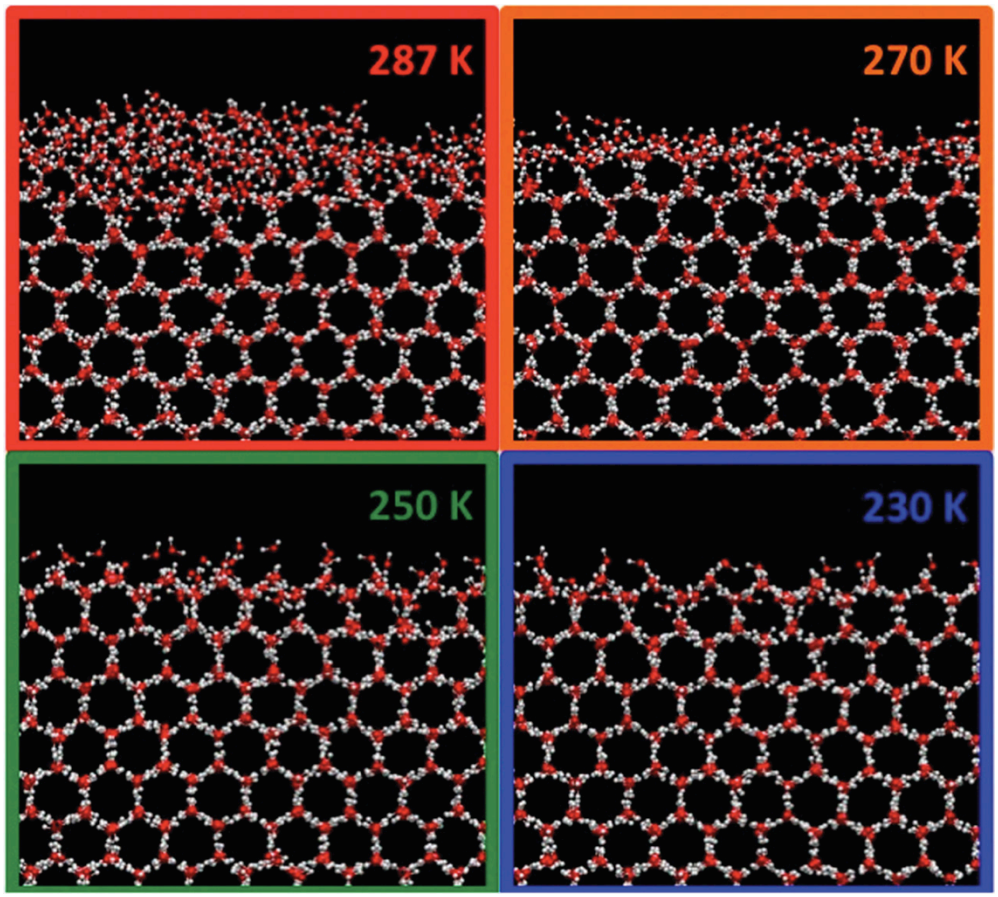
[4]Martina Roeselová, Institute of Organic Chemistry and Biochemistry, Academy of Sciences of the Czech Republic

Thank you for listening

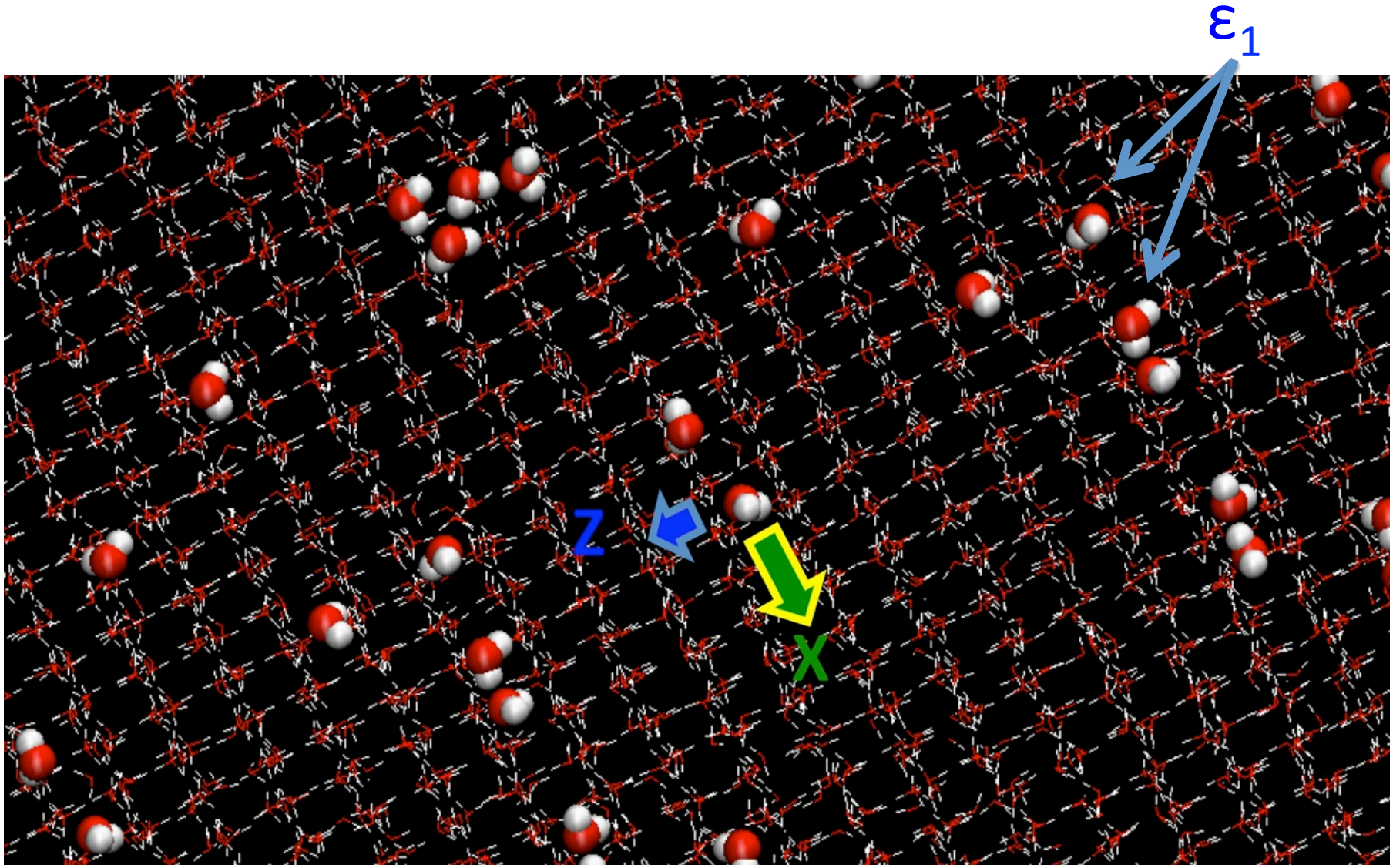
Diffusion below -50°C occurs as discrete jumps



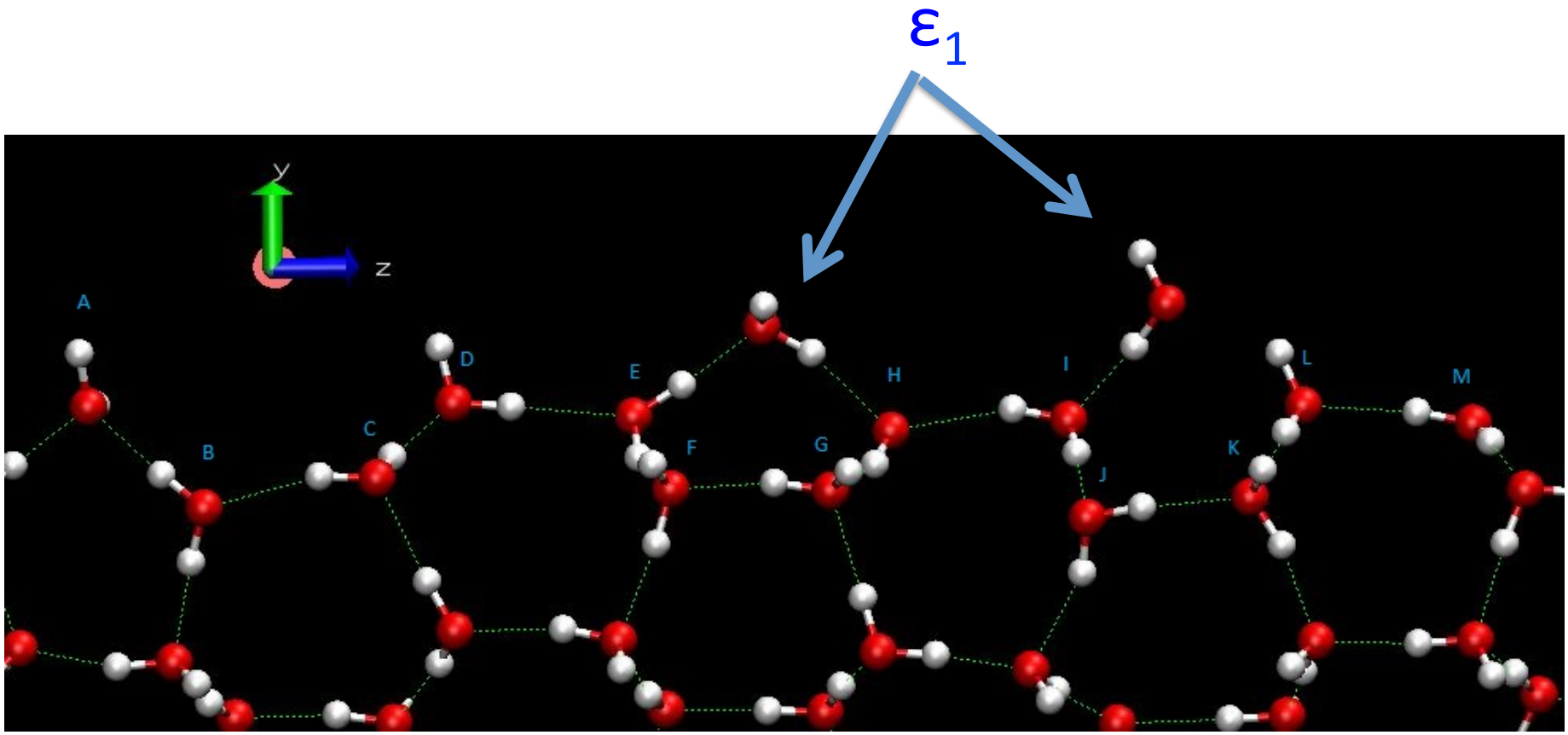
QLL gets too thick at higher temperature to support anisotropic diffusion

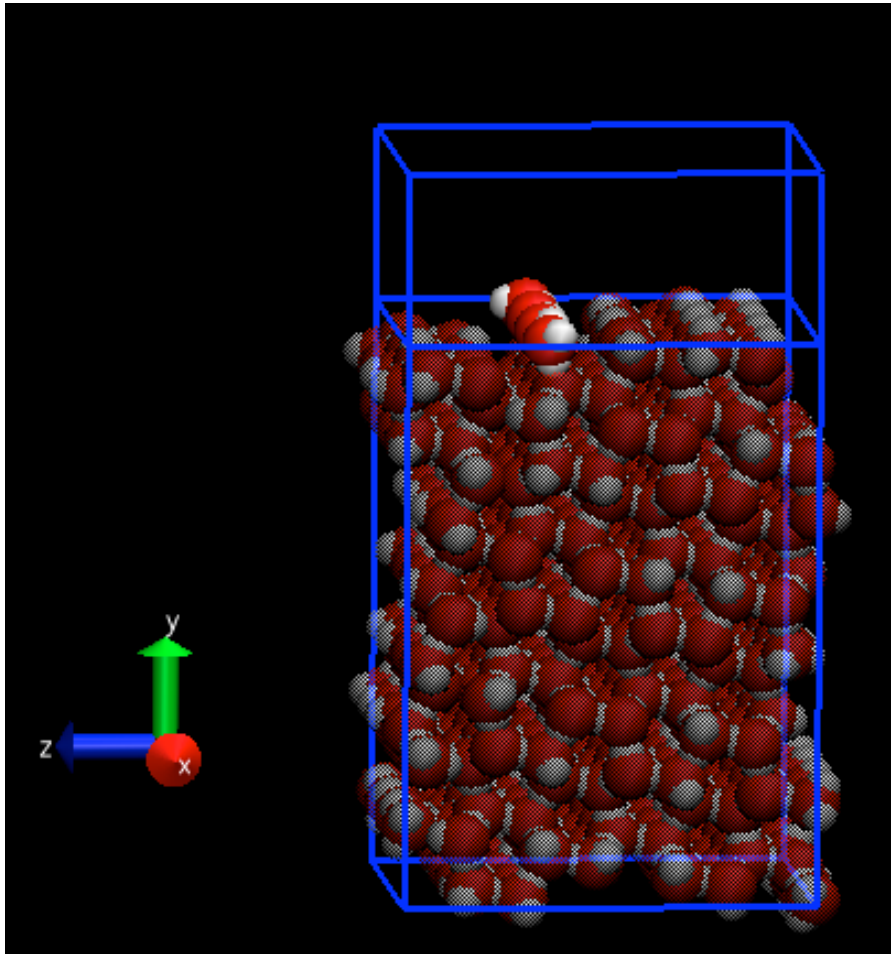


ε_1 admolecules prefer every other "row" by 70-30



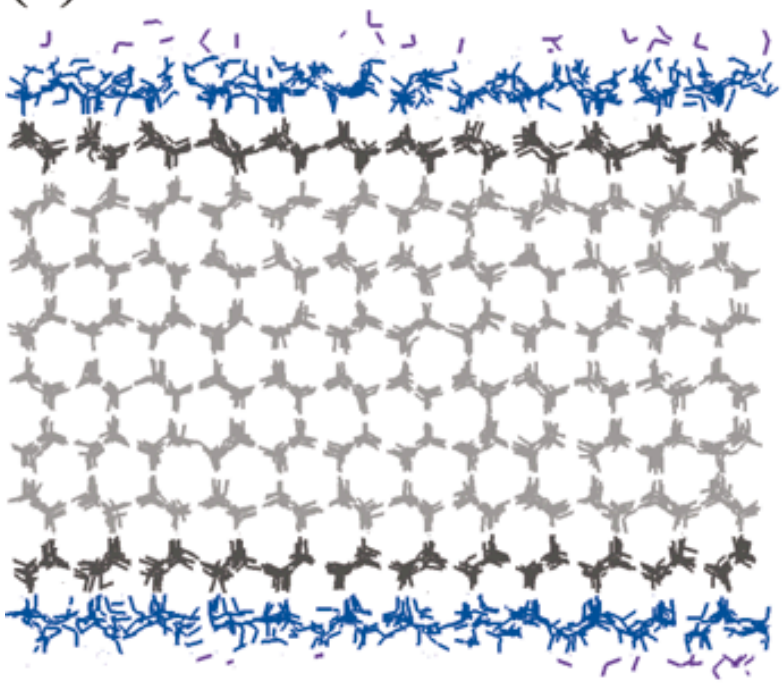
Possibly due to 5-membered ring completion in ε_2



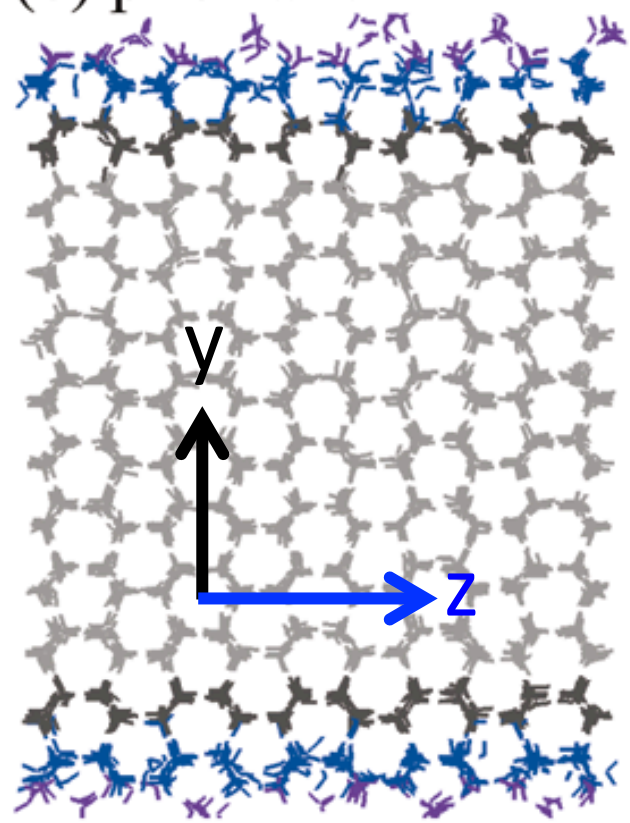

y
z
x

Molecular dynamics (MD) simulations in slab configuration, different facets exposed

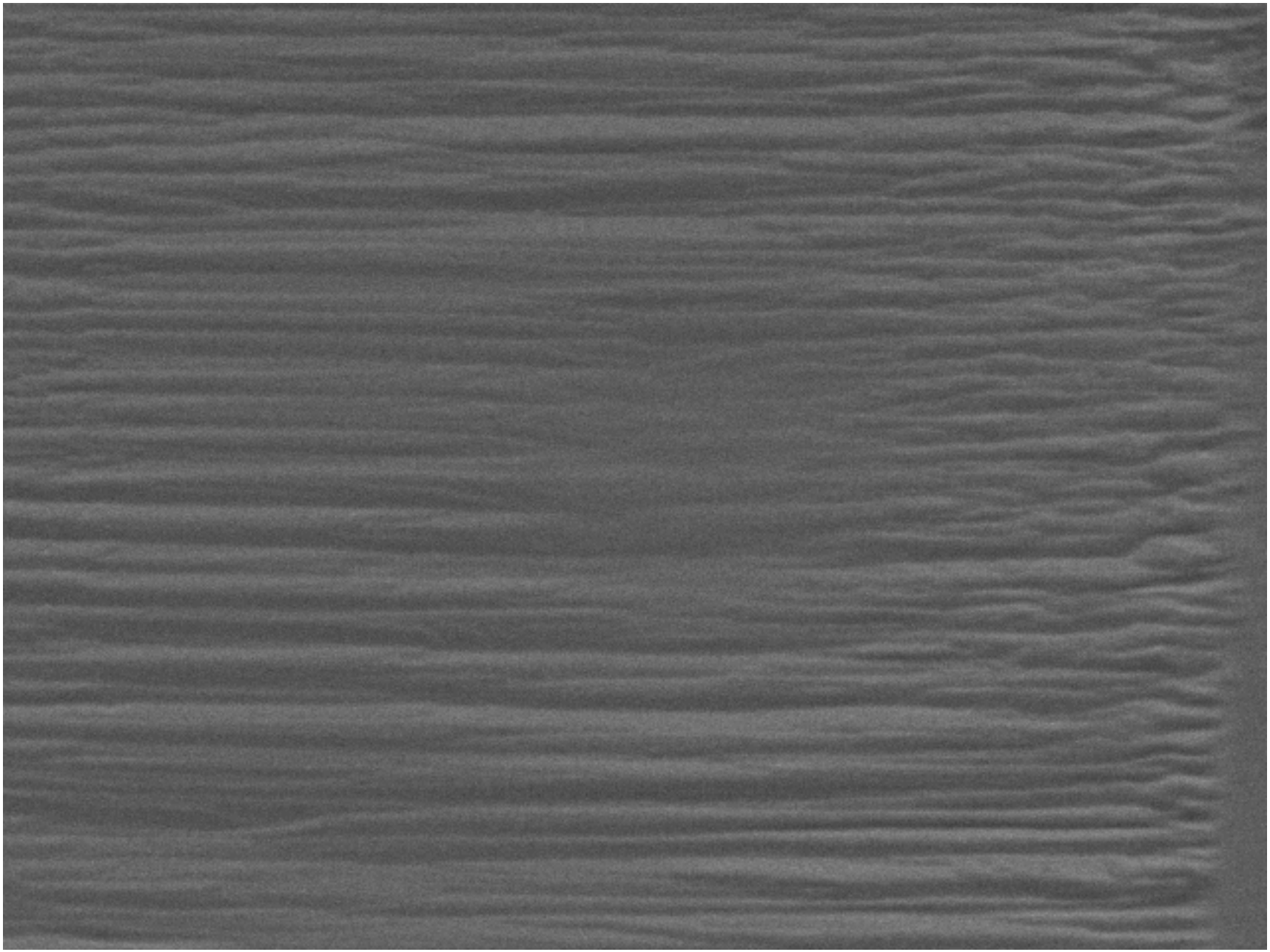
(a) basal

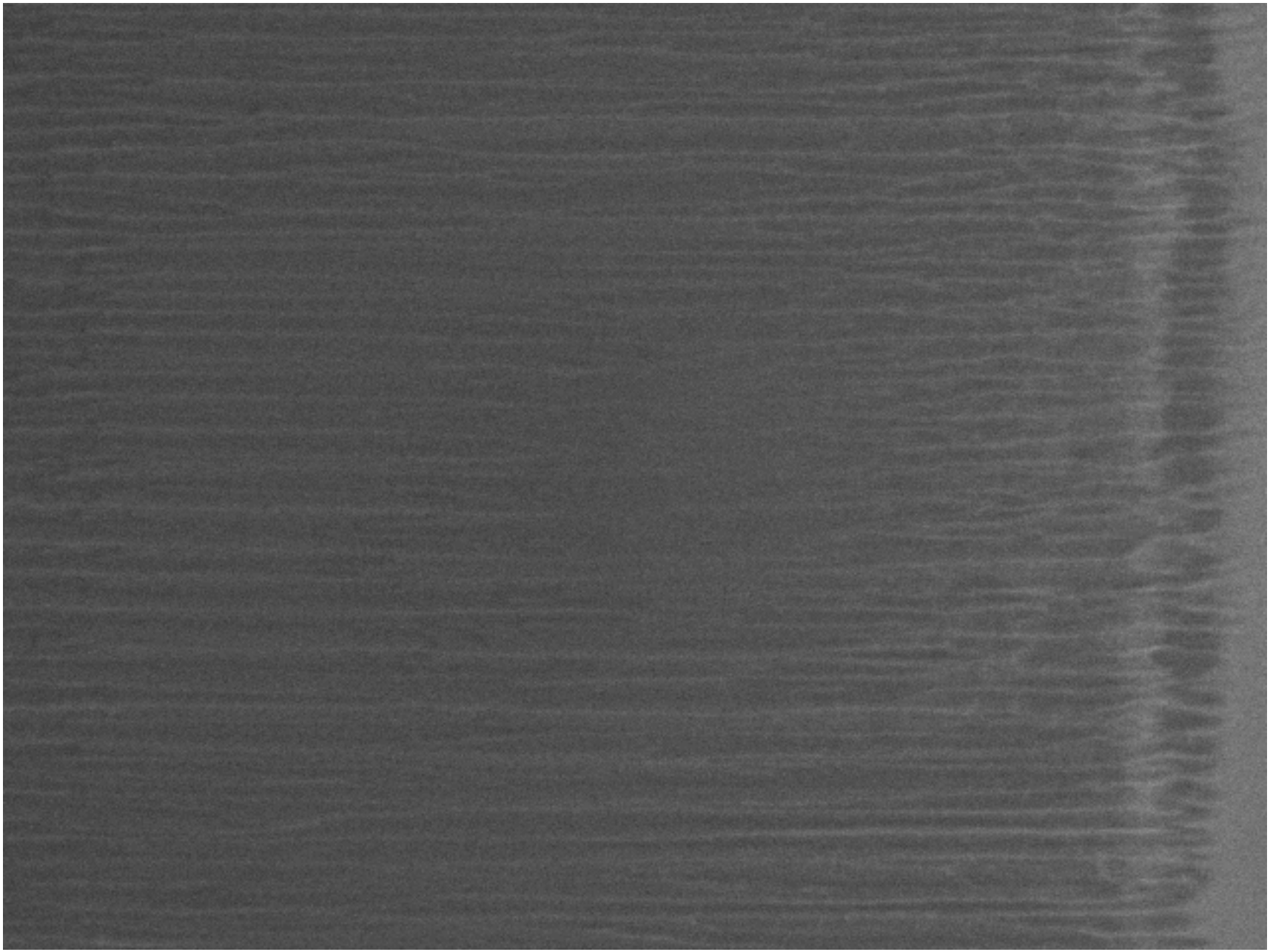


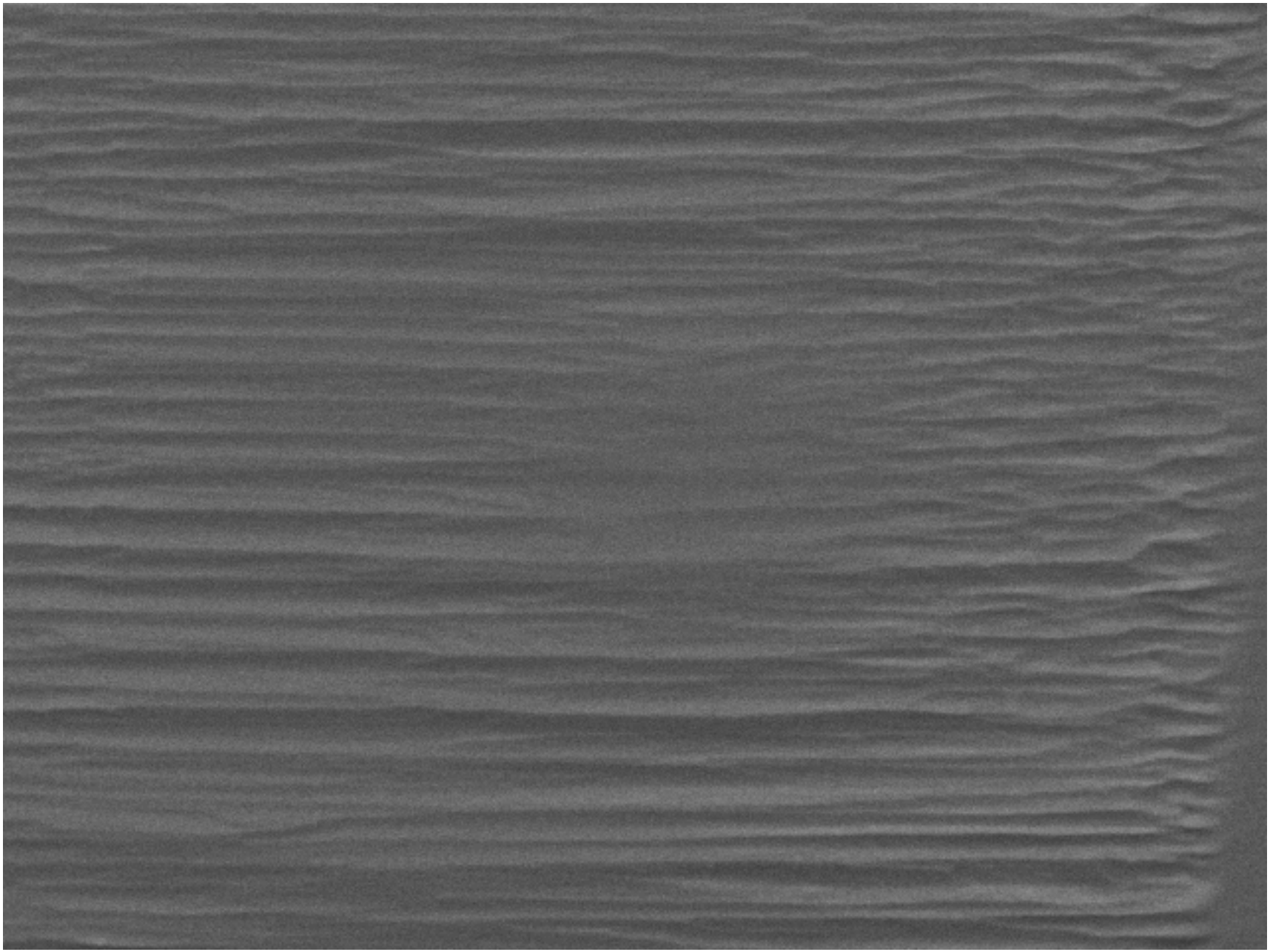
(b) prismatic



Pfalzgraff et al, JPC 2011, using NE6 potential of Nada & van der Eerden, JCP 2003







Initial method: extract roughness profile by digitizing a prismatic/prismatic edge