Ice Formed By Contact Freezing: Pressure Matters, Not Just Temperature?

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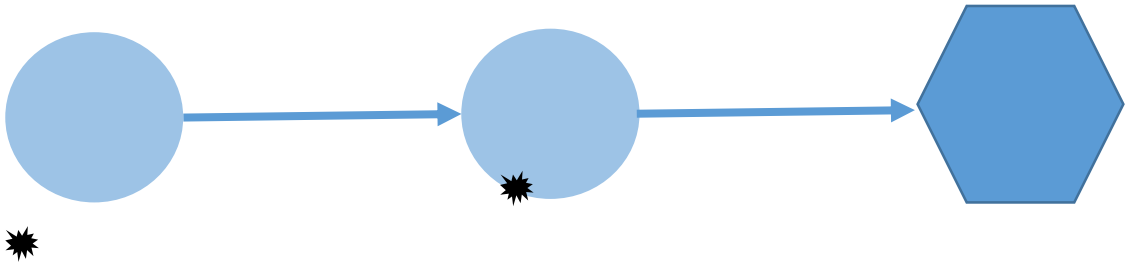

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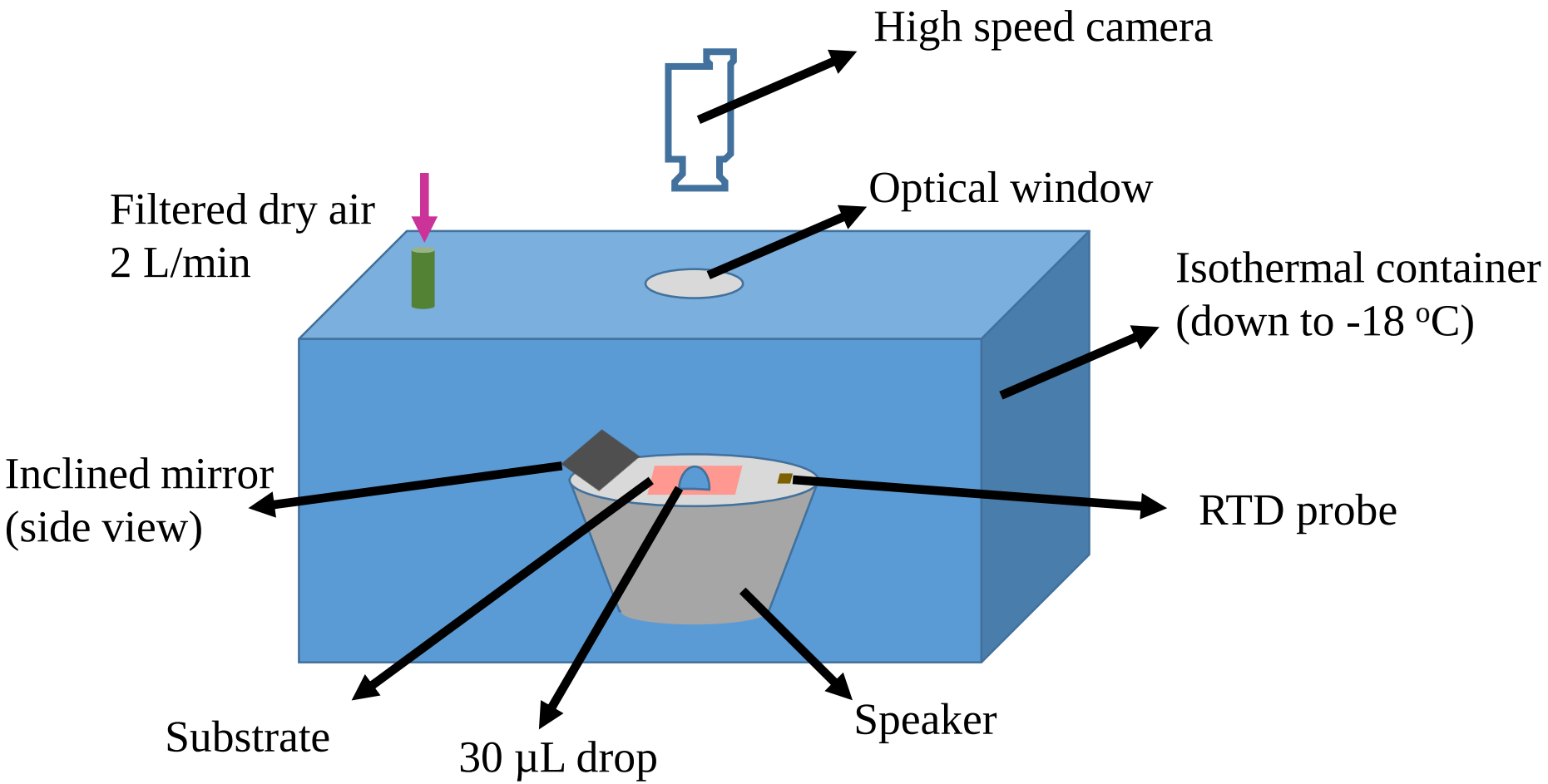
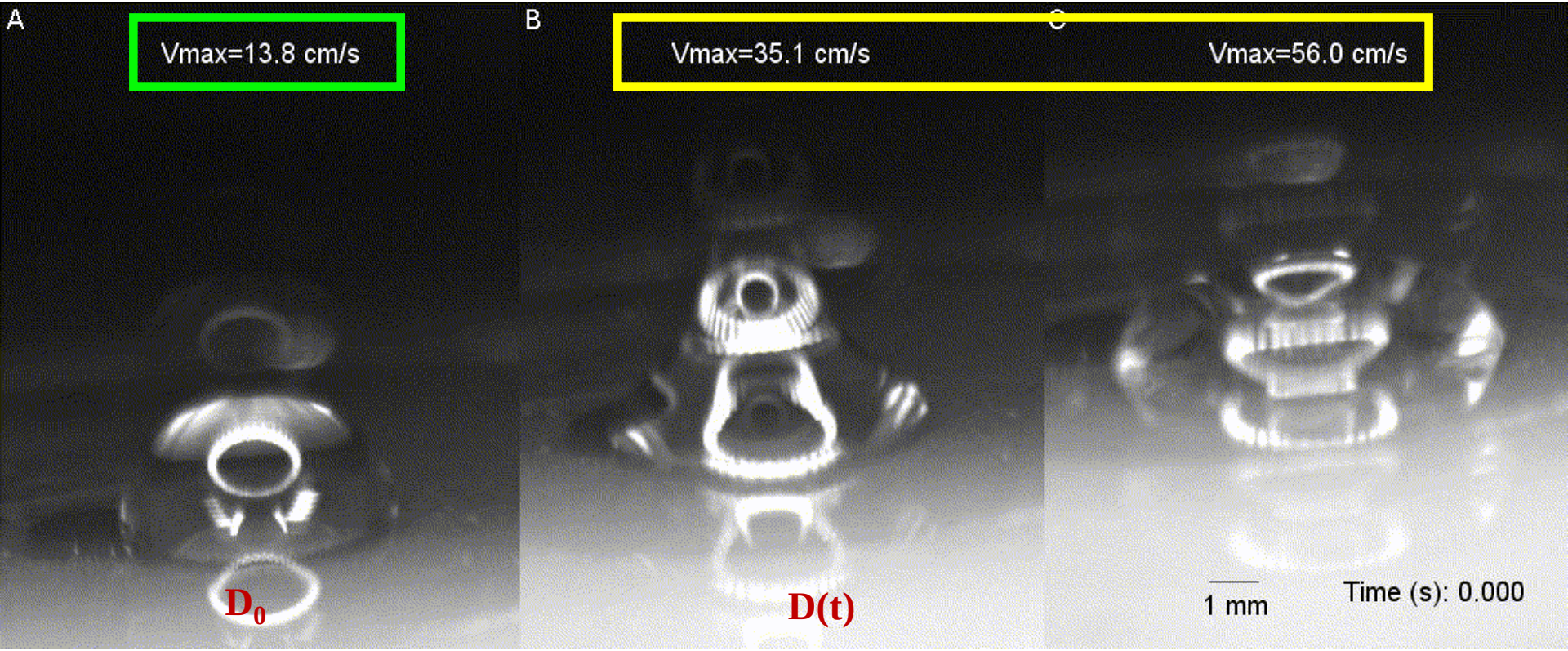
Experimental setup



Contact angle hysteresis

Contact angle oscillation

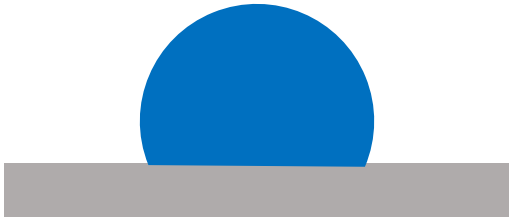
Moving contact line



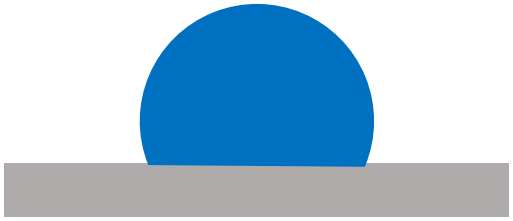
Frequency: 30 Hz

Relative spreading distance: $(D(t)-D_0)/D_0$

Experiments



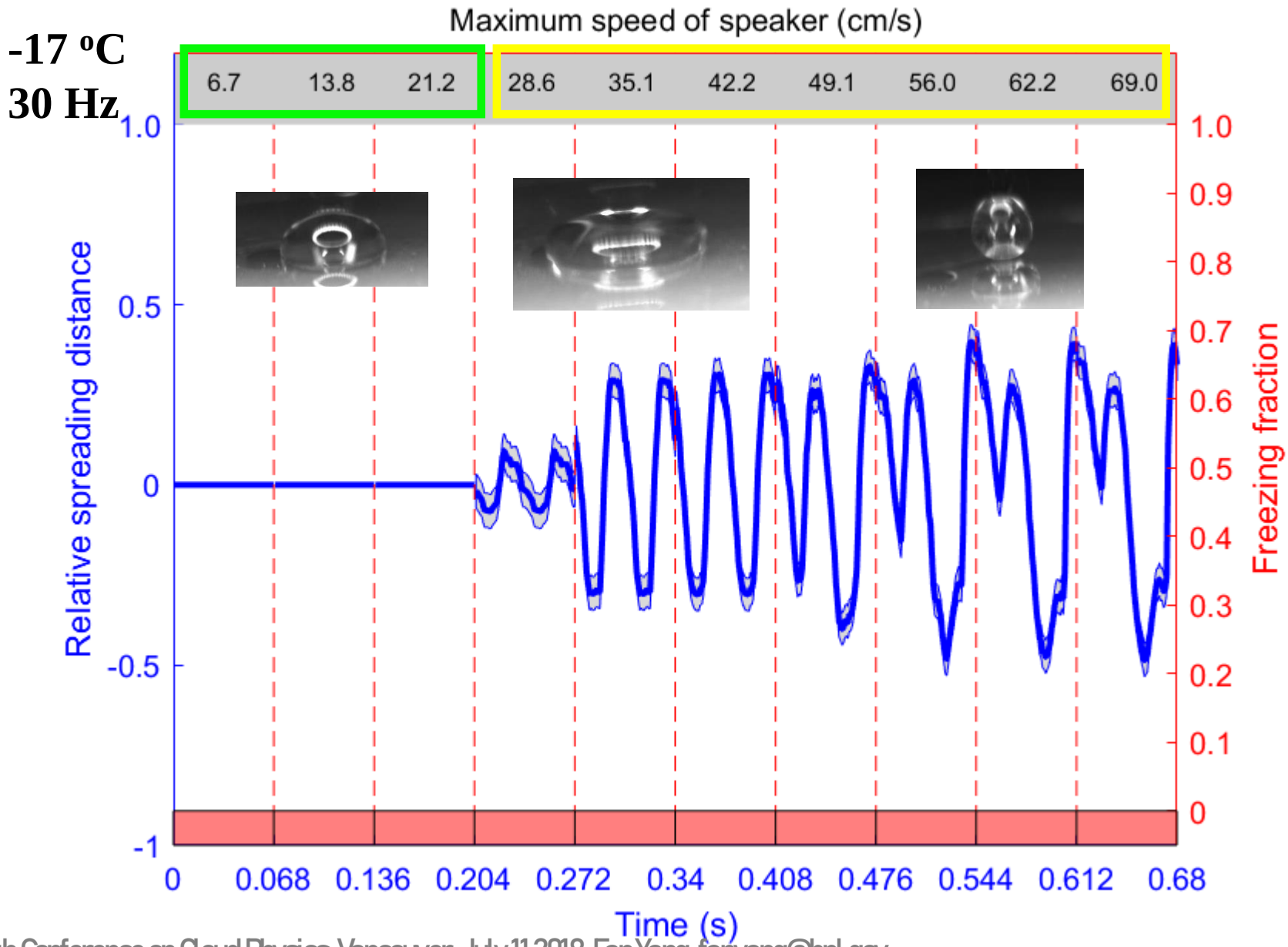
Pure water on
silicon glass substrate



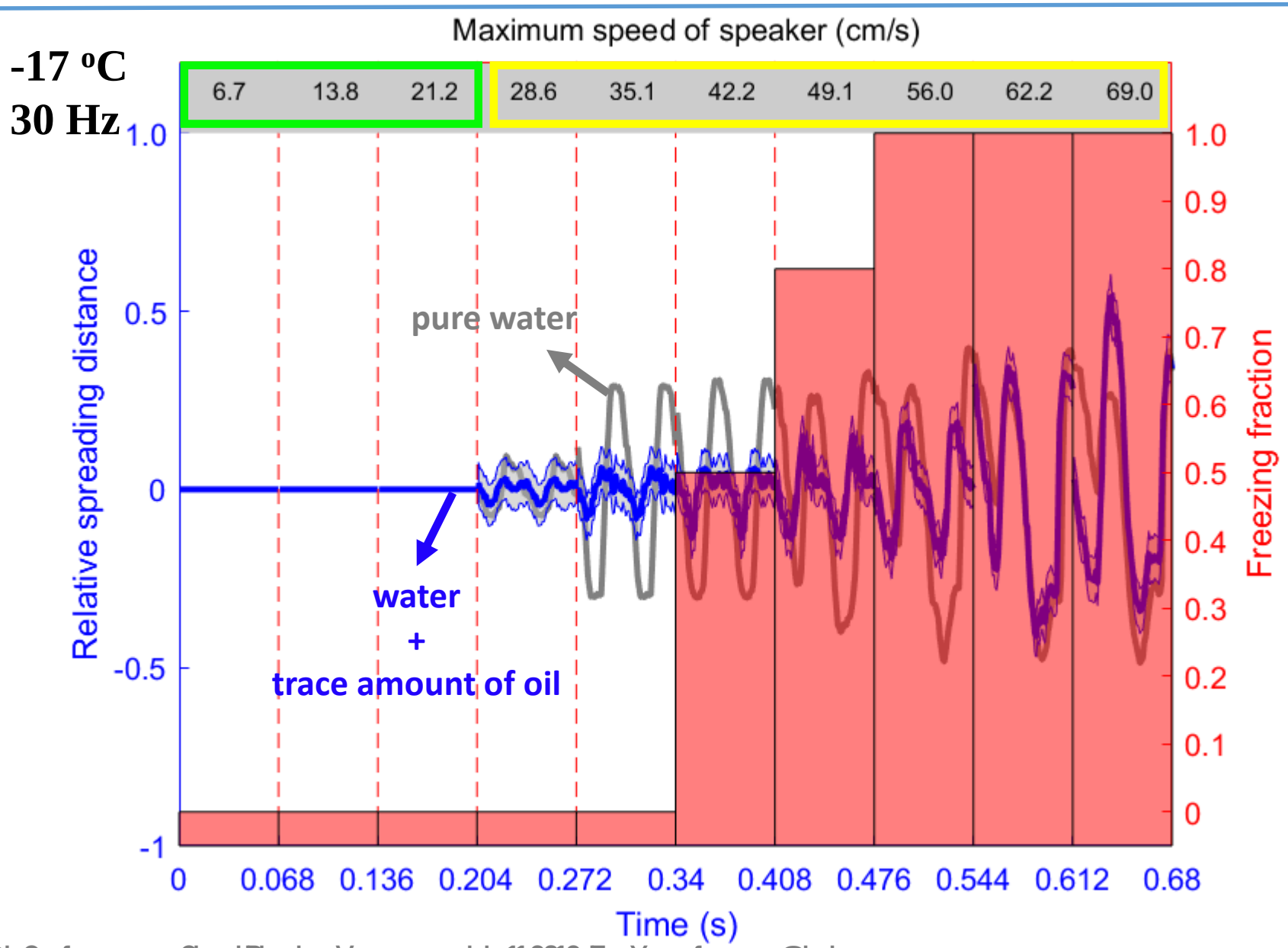
Water
with trace amount of oil
on silicon glass substrate

Look identical
Smell similar?
Taste similar?

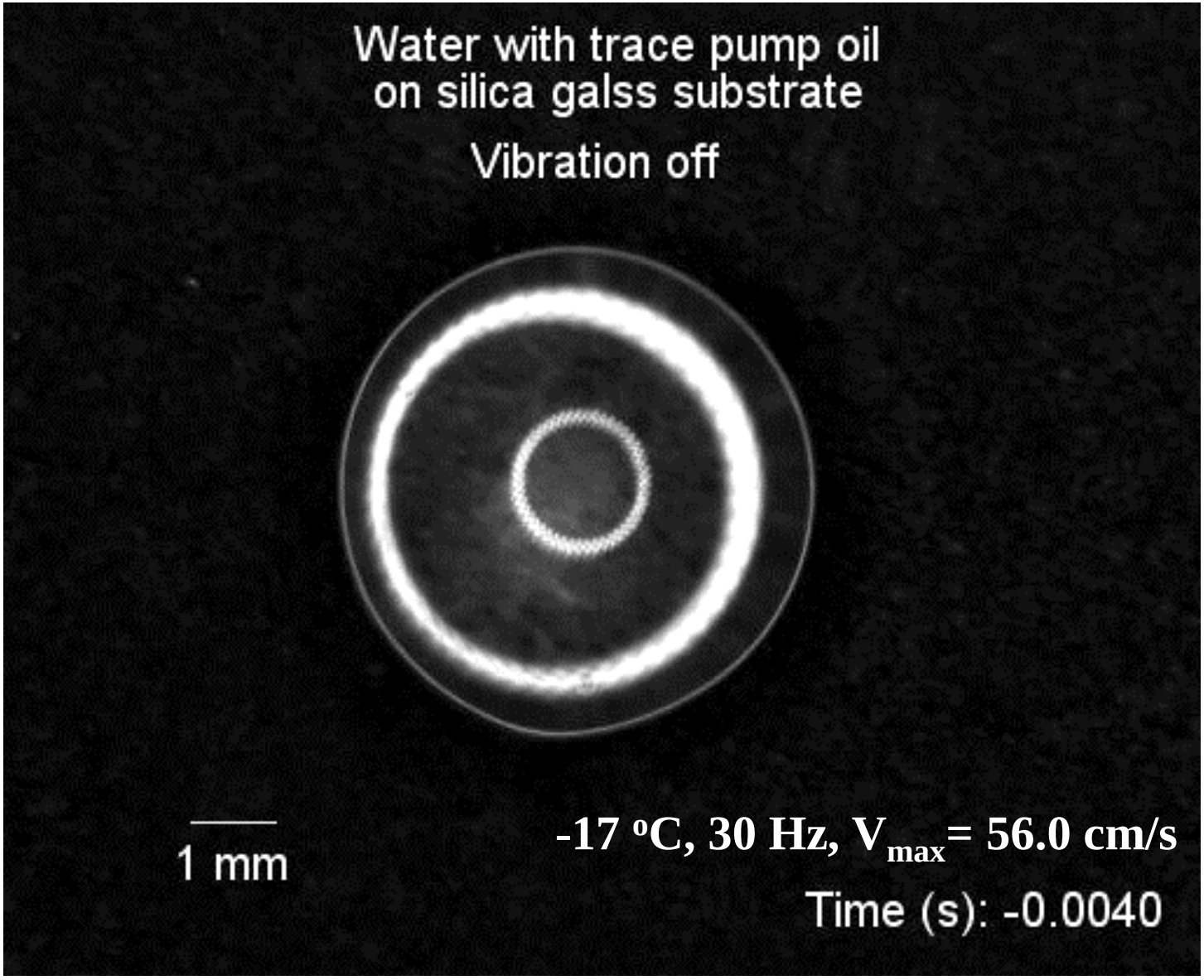
Pure water on silica glass



Water with 10 mg/mL pump oil



Water with 10 mg/mL pump oil



Possible mechanisms

☒ Temperature

Temperature for vibration experiment
-17 ± 0.5 °C

☒ Chemical property of oil or substrate

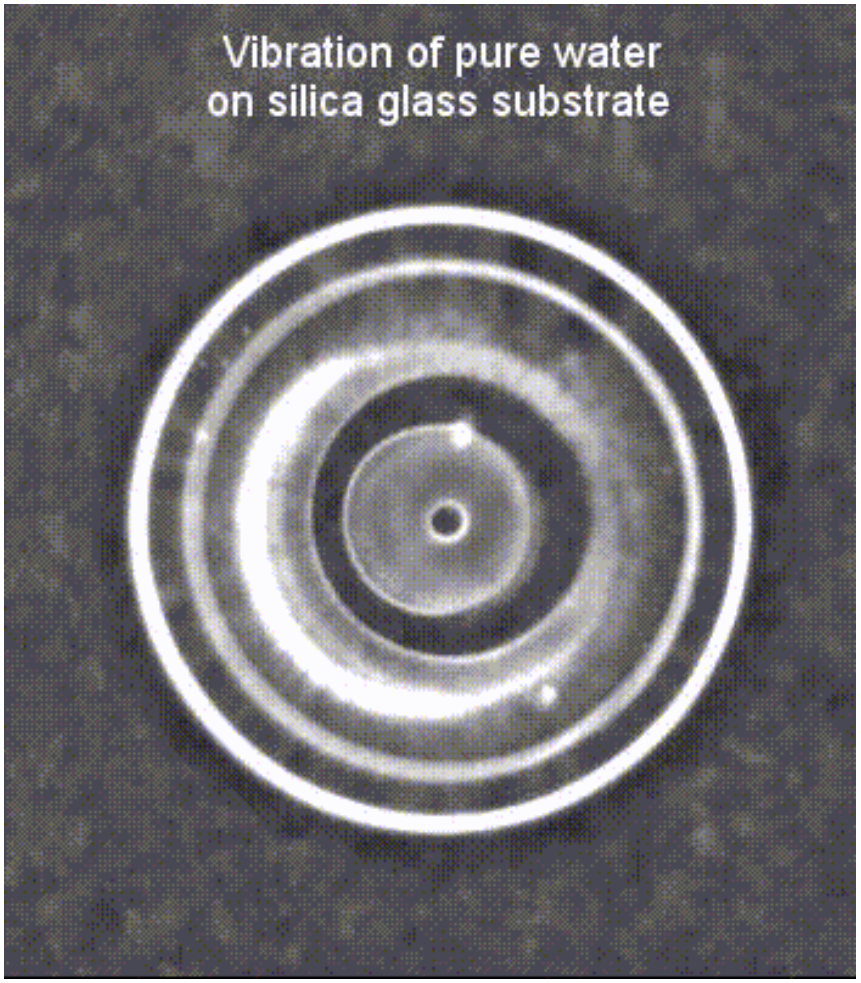
Experiments	Natural freezing temperature
30 µL pure water on silica substrate	-25.6 ± 0.6 °C
30 µL water with pump oil (10mg/mL)	-25.8 ± 0.6 °C
30 µL water with mineral oil (10mg/mL)	-26.5 ± 0.4 °C

☒ Nearby frost or active site

- No freezing at small amplitude
- Multiple point freezing
- No freezing for pure water

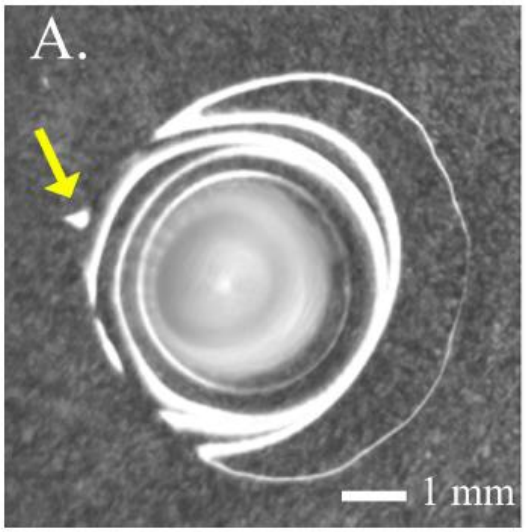
NOT temperature!
Whatever the mechanism is, it must be related to **drop oscillation and the moving contact line**, rather than
temperature, chemistry of oil, or defects on the substrate.

Top view of oscillating drop before freezing



Pressure as a cause of ice nucleation

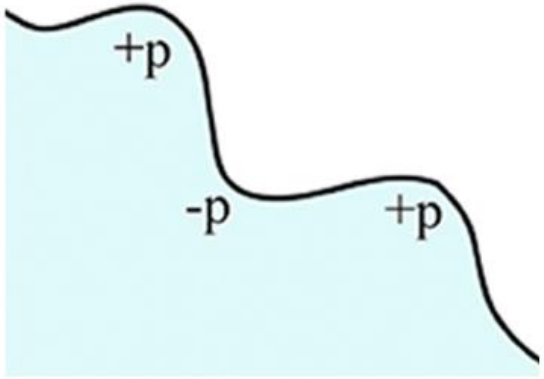
Oil-water mixture on glass substrate



Laplace pressure

B.

$$\Delta p = \frac{2\sigma}{r}$$



Locally curved contact line is related to ice nucleation, which suggests pressure, rather than temperature, as a cause of ice nucleation.

Chemical potential change for ice nucleation

Driven force for ice nucleation $\Delta\mu = \mu_L - \mu_S = l_f \frac{\Delta T_s}{T_0}$

Li et al. Nature Comm. 2013

Given the water density anomaly, Δv is negative,

- If $\Delta p > 0$, $\Delta\mu$ would be smaller, thus bad for ice nucleation.
  - Example: nanodroplet can exist much below -40 °C.

  positive Laplace pressure due to small radius of curvature.
- If $\Delta p < 0$, $\Delta\mu$ would be larger, thus good for ice nucleation.

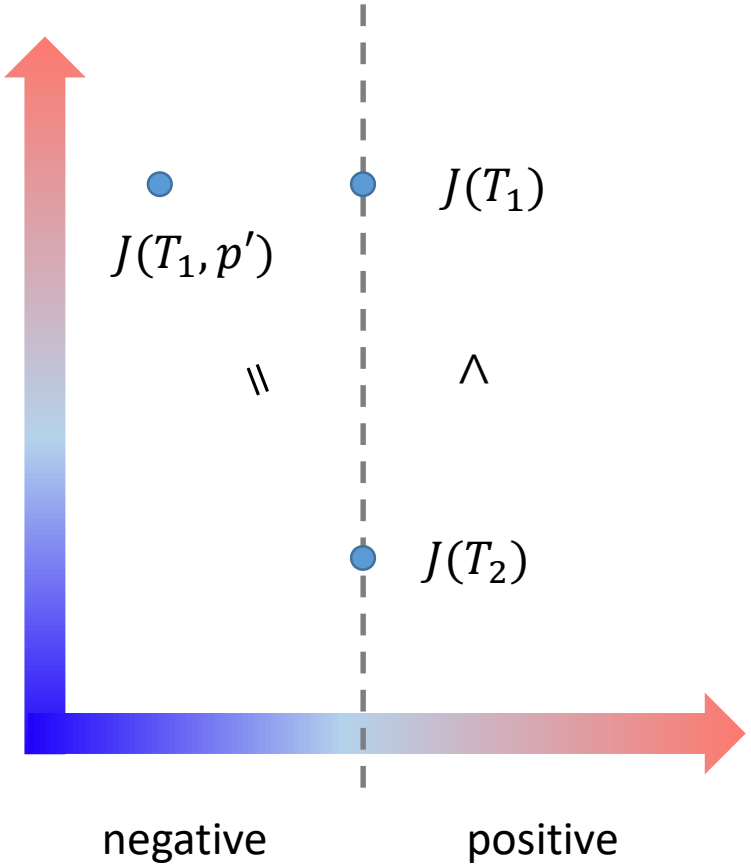
$$J_i(T) \propto exp\left(-\frac{\Delta G_i^*}{k_B T}\right)$$

Ice nucleation rate

$$\Delta G_i^* = \frac{16\pi\sigma_{IL}^3}{3\left(n_i \Delta\mu\right)^2} f_{het}$$

Energy barrier

Pressure perturbation—equivalent temperature



$$T_e = T - \frac{T_0 \Delta v}{l_f} p'$$

Explain previous experimental results

- Contact nucleation is more efficient.

  water droplet and ice nuclei *Fukuta et al. JAS, 1975*

  two supercooled droplets *Alkezweeny JAM, 1969*

  water droplet and salt particle *Niehaus and Cantrell JPCL, 2015*

- Enhanced ice nucleation rate at contact line region

  Locally curved contact line due to strong pinning

  Durant and Shaw GRL, 2005

  Gurganus et al. PRL, 2014

- Electrofreezing due to transient electrowetting field

  Yang et al. APL, 2015

Take home message

$$T_e = T - \frac{T_0 \Delta v}{l_f} p'$$

- Mechanical agitation induces freezing of supercooled water drops at distorted contact lines, pointing to **negative pressure**, rather than temperature as a cause.
- Pressure induced ice nucleation provides a new context for interpreting the previous diverse observations related to contact nucleation.

More details in Yang, F., Cruikshank, O., He, W., Kostinski, A., and Shaw, R. A.
Nonthermal ice nucleation observed at distorted contact lines of supercooled water drops.
Physical Review E, 2018