Cloud from a Chip

Developments in Microfluidic Ice Nucleation Experiments

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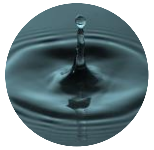


Atmospheric Physics

Prof. Ulrike Lohmann
Claudia Marcolli
Zamin Kanji
Michael Rösch

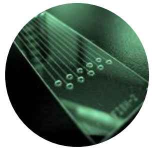
Separation Processes

Prof. Marco Mazzotti
Leif-Thore Deck
Imad El-Bakouri

Atmospheric Chemistry

Prof. Thomas Peter
Claudia Marcolli
Ulrich Krieger

Microfluidics



Prof. Andrew deMello
Stavros Stavrakis
Florin Isenrich

\+ Roland Walker,
Fredy Mettler,
& Benedikt Waser

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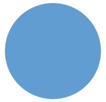
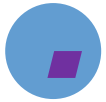
Motivation

- Presence or absence of ice in clouds affects
  - how much sunlight is absorbed, reflected, or transmitted through clouds
  - if and how much precipitation will fall

- To understand  and forecast , we need to first predict when ice forms
  - homogeneous and heterogeneous nucleation





How is ice formation investigated?

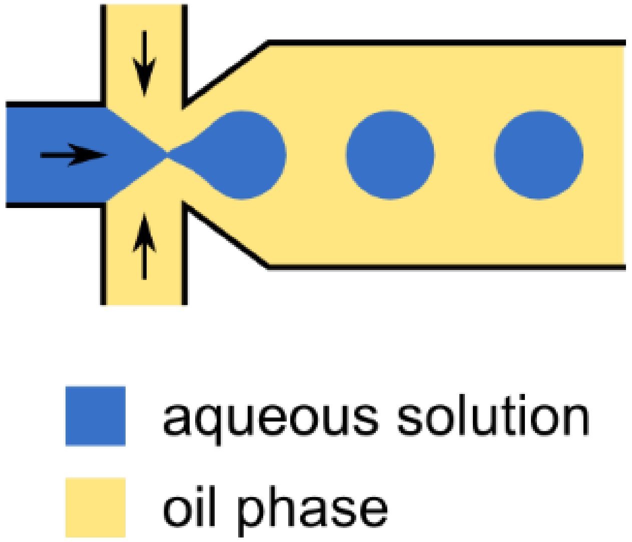
- Suspended droplets — short residence times
- Drop(s) on substrate — large drop volumes
- Emulsions — polydisperse droplet size

- Recently, droplets from **microfluidic instruments***
  - long residence times
  - small droplet volumes (nL to pL)
  - monodisperse droplet size

* Reicher et al. **2018** *AMT*, 11.
Brubaker et al. **2020** *Aerosol Sci Technol*, 54.
Tarn et al. **2021** *Micromachines*, 12(2).
Roy et al. **2021** *Micromachines*, 12(3).

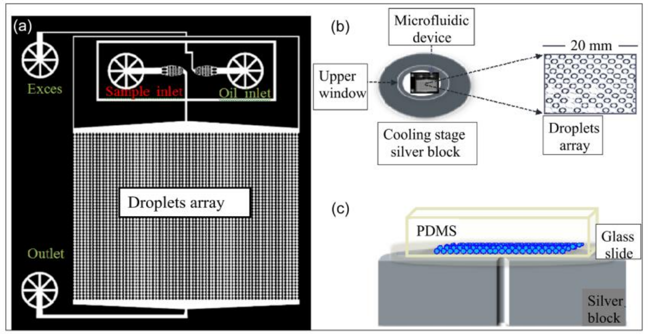
What is microfluidics?

- Manipulation and control of liquids at sub-microliter volumes



Microfluidic devices for studying ice nucleation

- **On-chip** droplet generation and cooling



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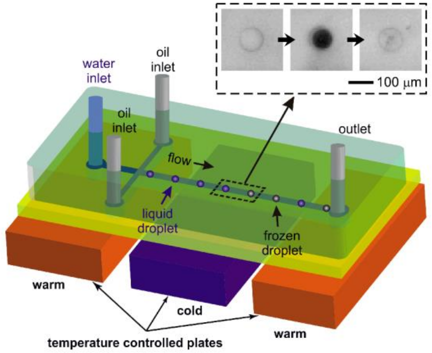

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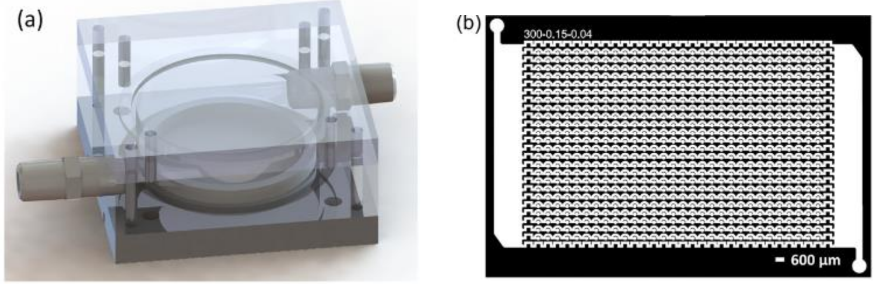

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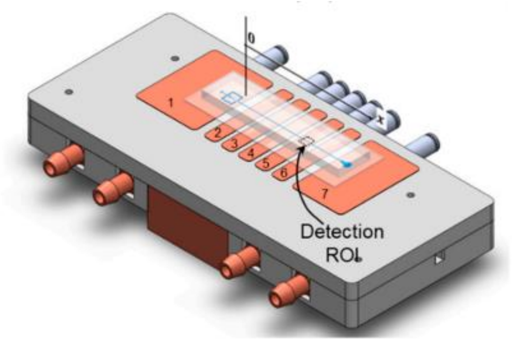


Roy et al. **2021** *Micromachines*, 12(3).

Microfluidic devices for studying ice nucleation

- **On-chip** droplet generation and cooling
    - droplets shrink in the polymer device
    - high temperature uncertainty

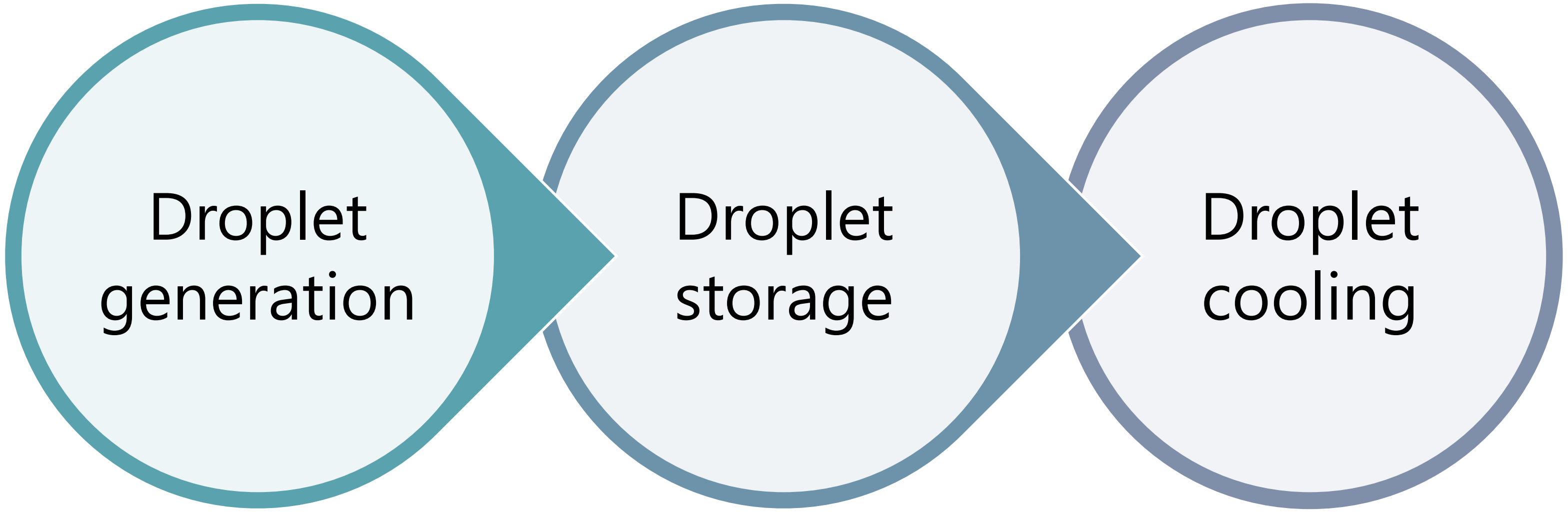
Can we rethink how we use microfluidics to study ice nucleation?

Outline

1. New microfluidics-based instrument

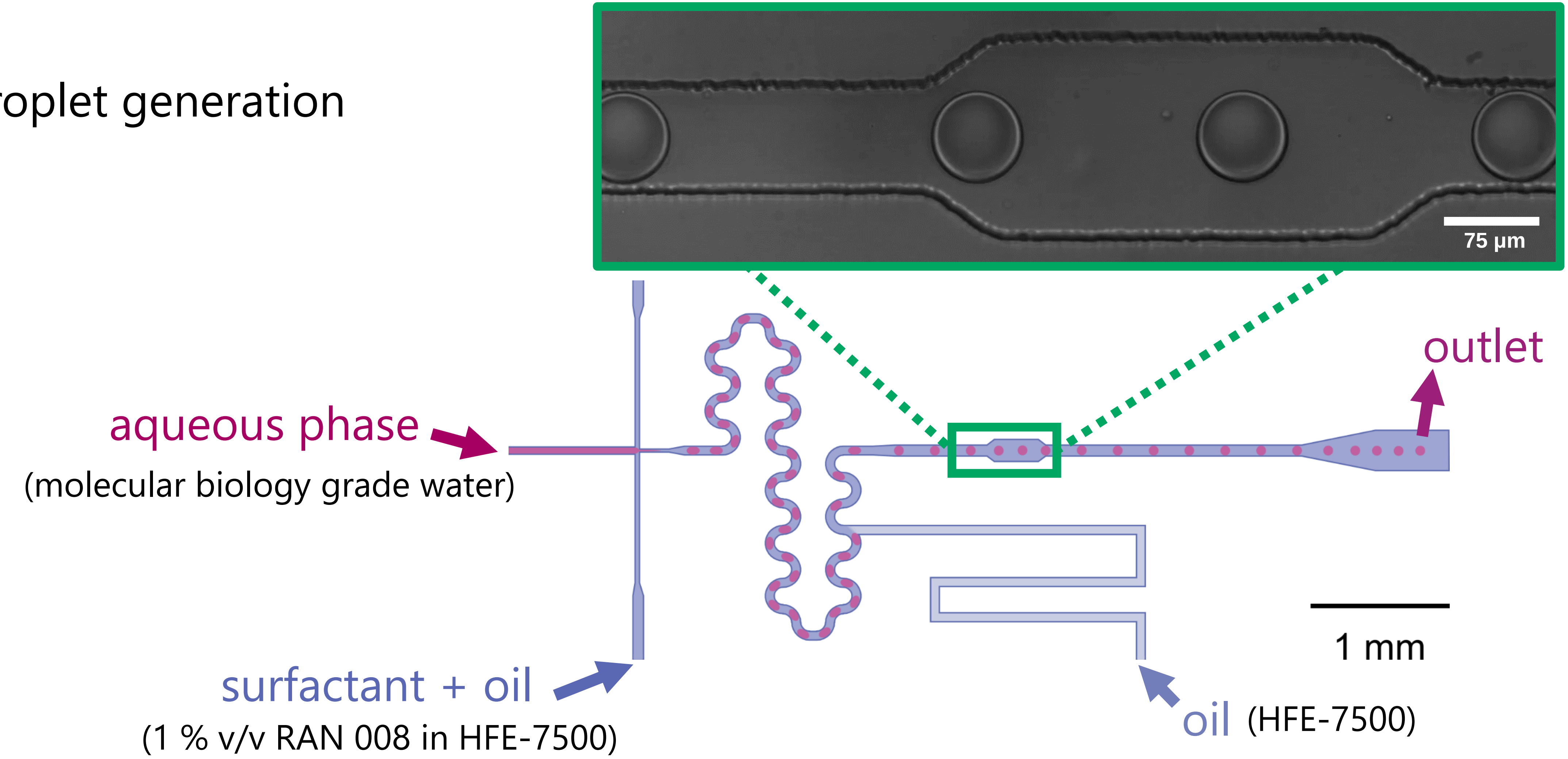
2. Homogeneous nucleation rate of water

New microfluidics-based instrument



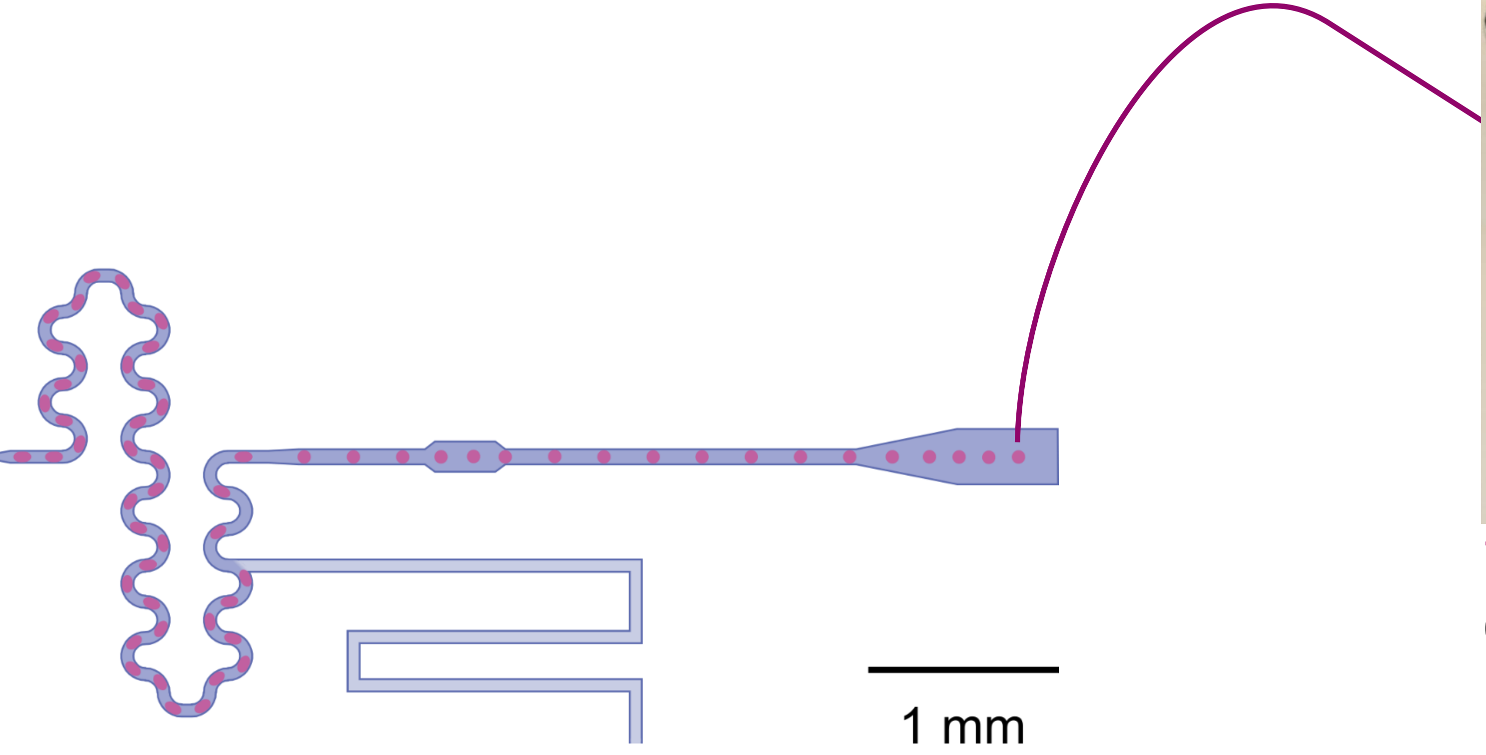
New microfluidics-based instrument

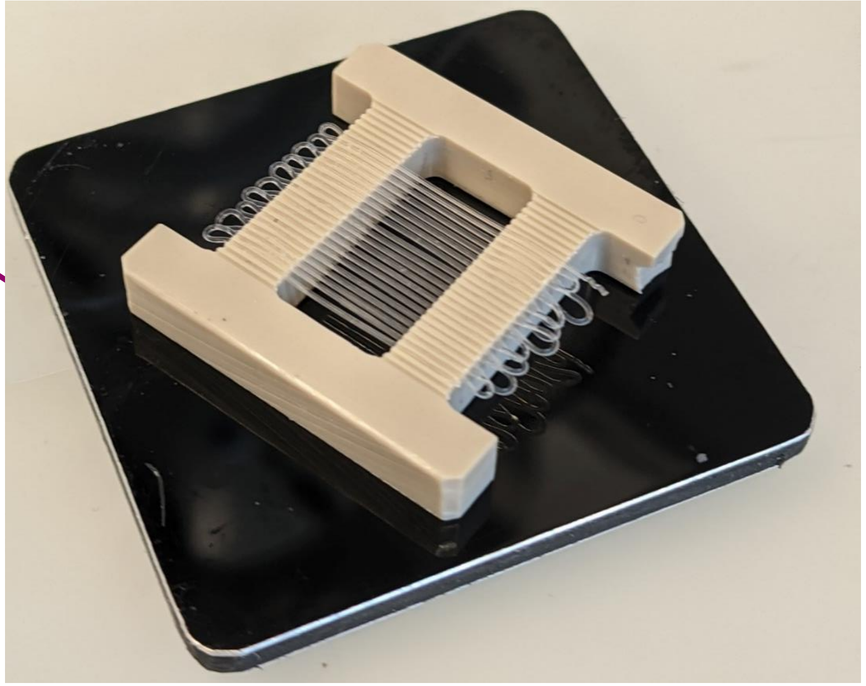
Droplet generation



New microfluidics-based instrument

Droplet storage

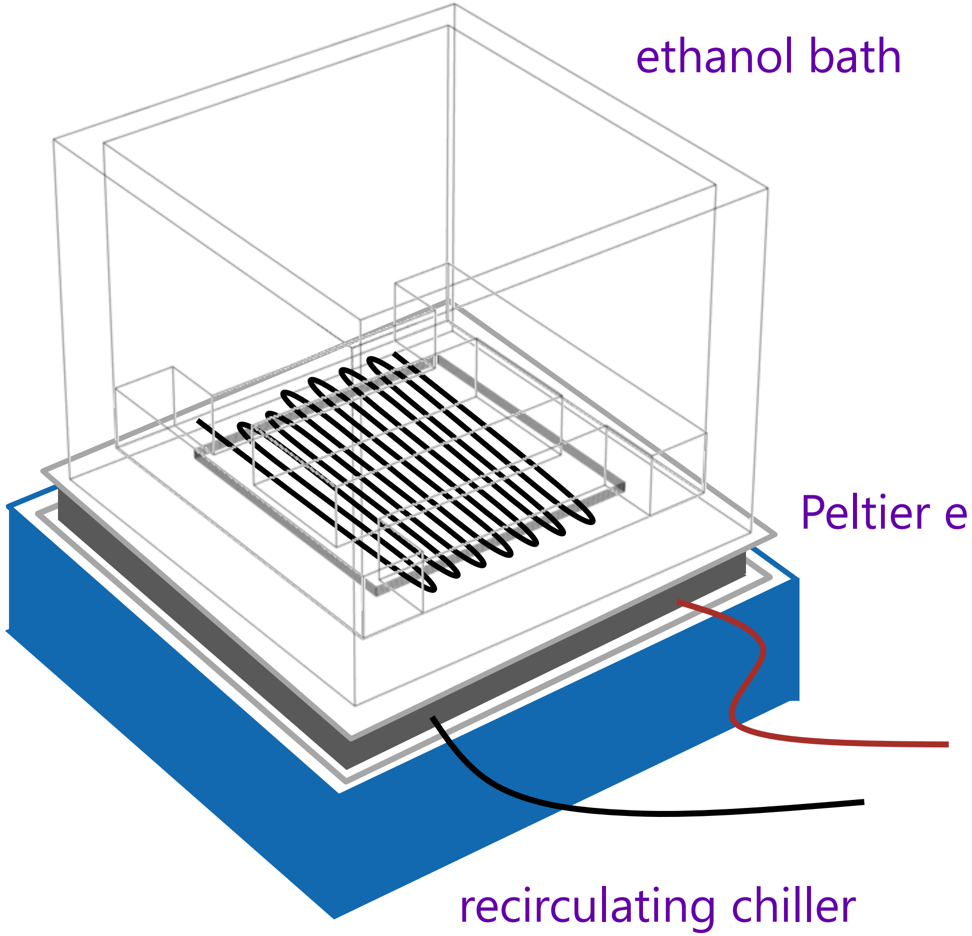




75 or 100 μm inner diameter tubing
(high-purity perfluoroalkoxy alkane)

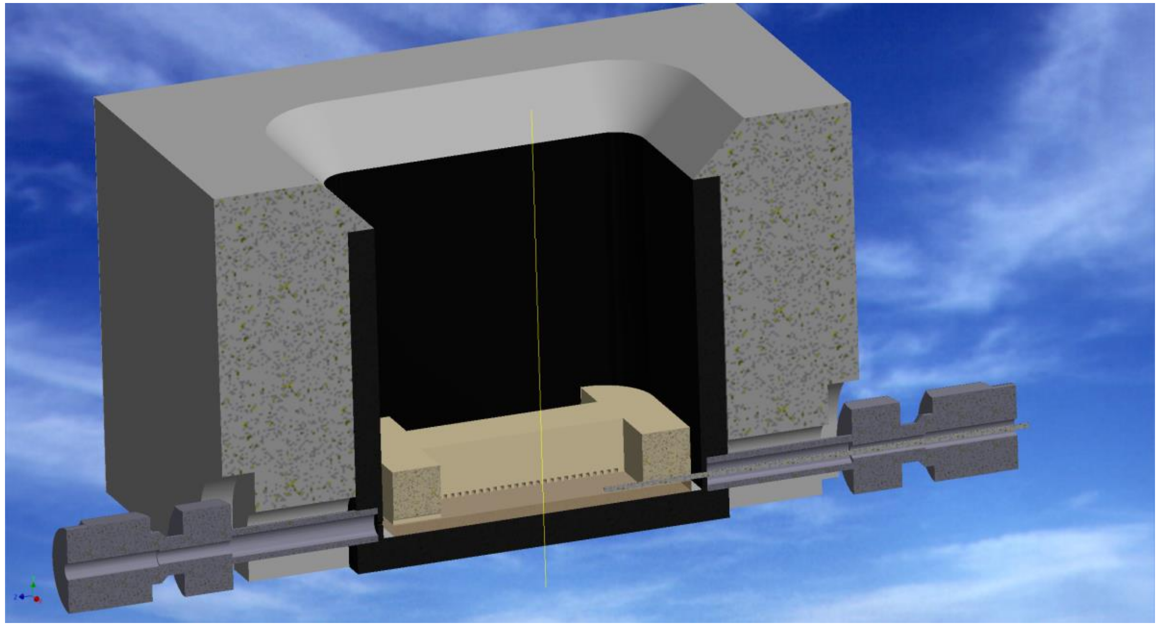
New microfluidics-based instrument

Droplet cooling



New microfluidics-based instrument

Droplet cooling



Courtesy of Roland Walker

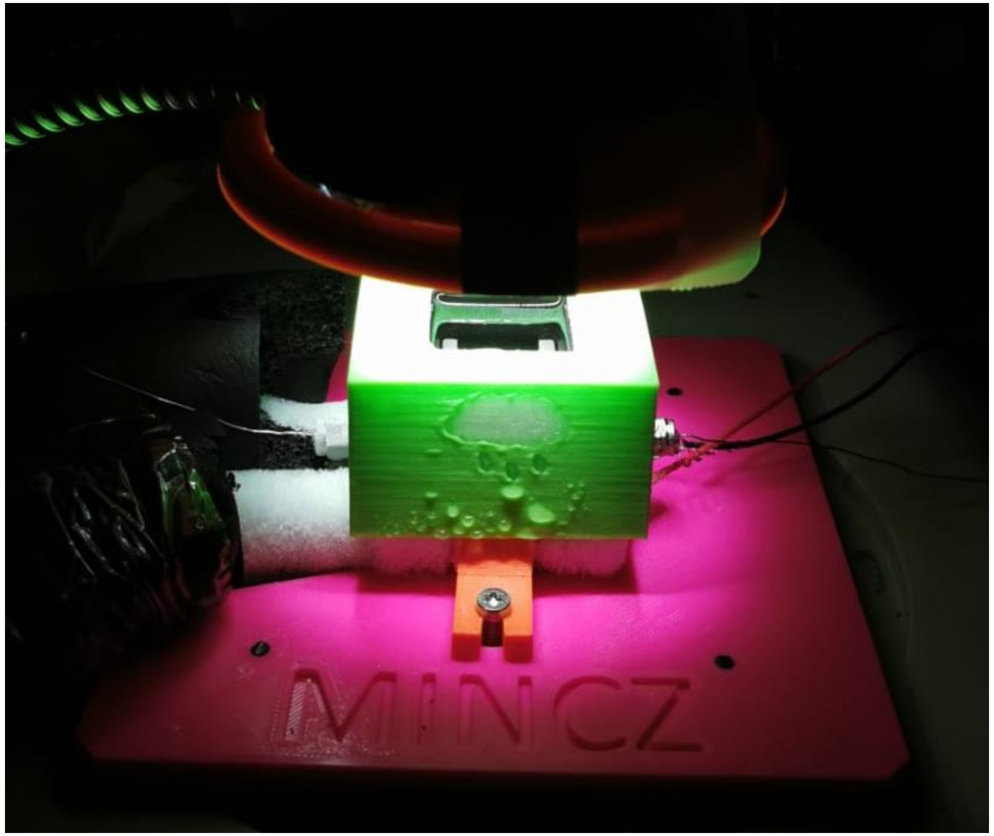
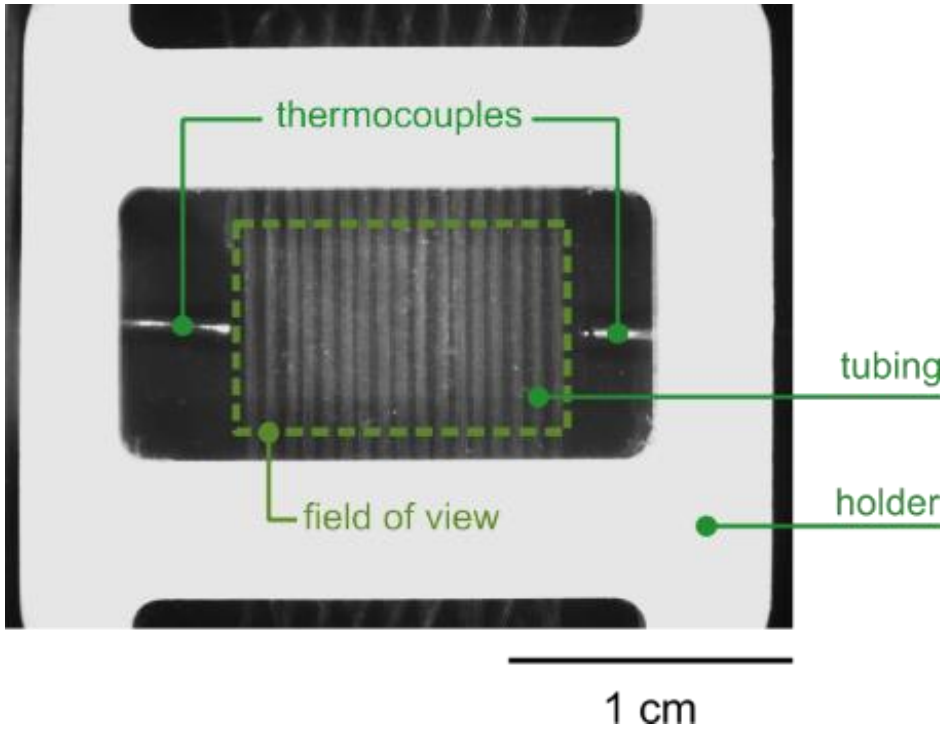


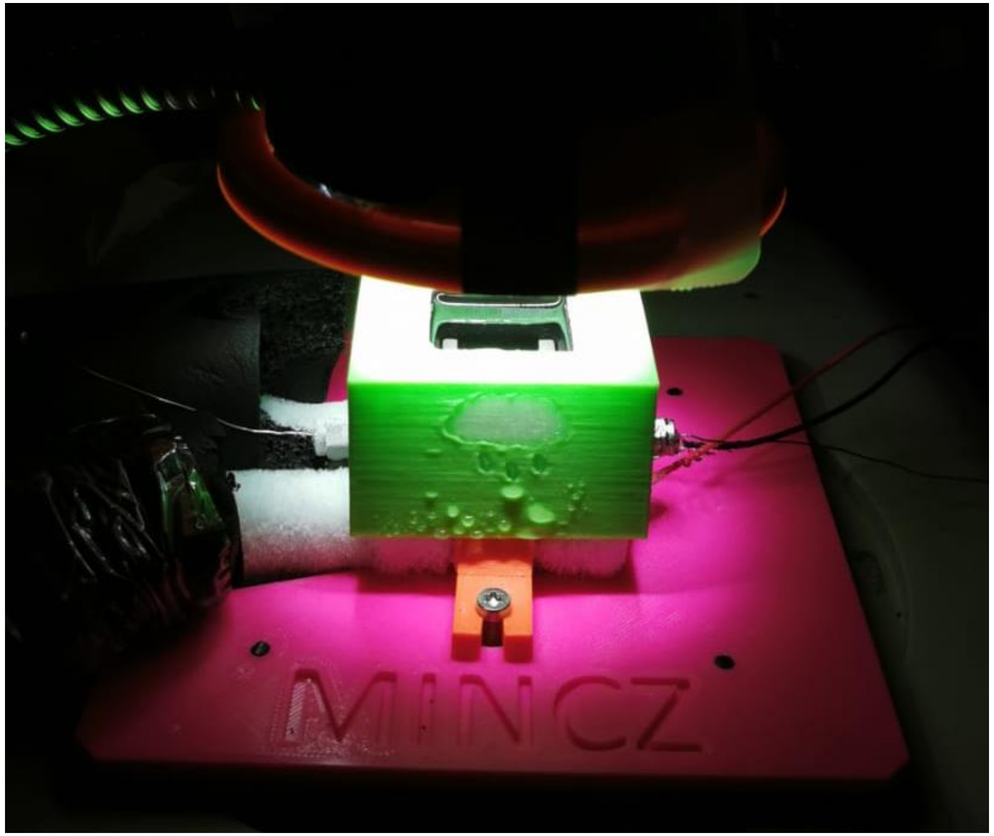
Photo by Imad El-Bakouri

New microfluidics-based instrument

Droplet cooling



New microfluidics-based instrument



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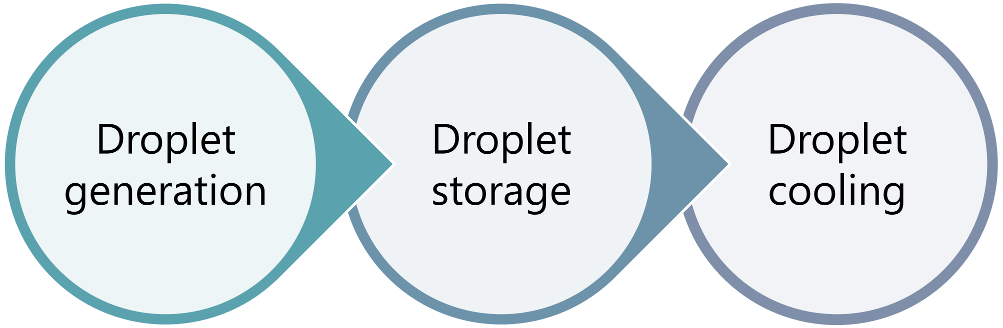

- By separating droplet generation and storage,
    - droplets are stable in storage
    - temperature accuracy is improved (± 0.2 K)

Outline

The Microfluidic Ice Nuclei Counter Zürich (MINCZ): a platform for homogeneous and heterogeneous ice nucleation

Florin N. Isenrich[1,★], Nadia Shardt[2,★], Michael Rösch[2], Julia Nette[1], Stavros Stavrakis[1], Claudia Marcolli[2], Zamin A. Kanji[2], Andrew J. deMello[1], and Ulrike Lohmann[2]

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[★]These authors contributed equally to this work.

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1. New microfluidics-based instrument

Isenrich et al. **2022** *AMT*, 15, 5367–5381.

2. Homogeneous nucleation rate of water

Outline

The Microfluidic Ice Nuclei Counter Zürich (MINCZ): a platform for homogeneous and heterogeneous ice nucleation

Florin N. Isenrich[1,★], Nadia Shardt[2,★], Michael Rösch[2], Julia Nette[1], Stavros Stavrakis[1], Claudia Marcolli[2], Zamin A. Kanji[2], Andrew J. deMello[1], and Ulrike Lohmann[2]
[1]Institute for Chemical and Bioengineering, ETH Zurich, Zürich, 8093, Switzerland
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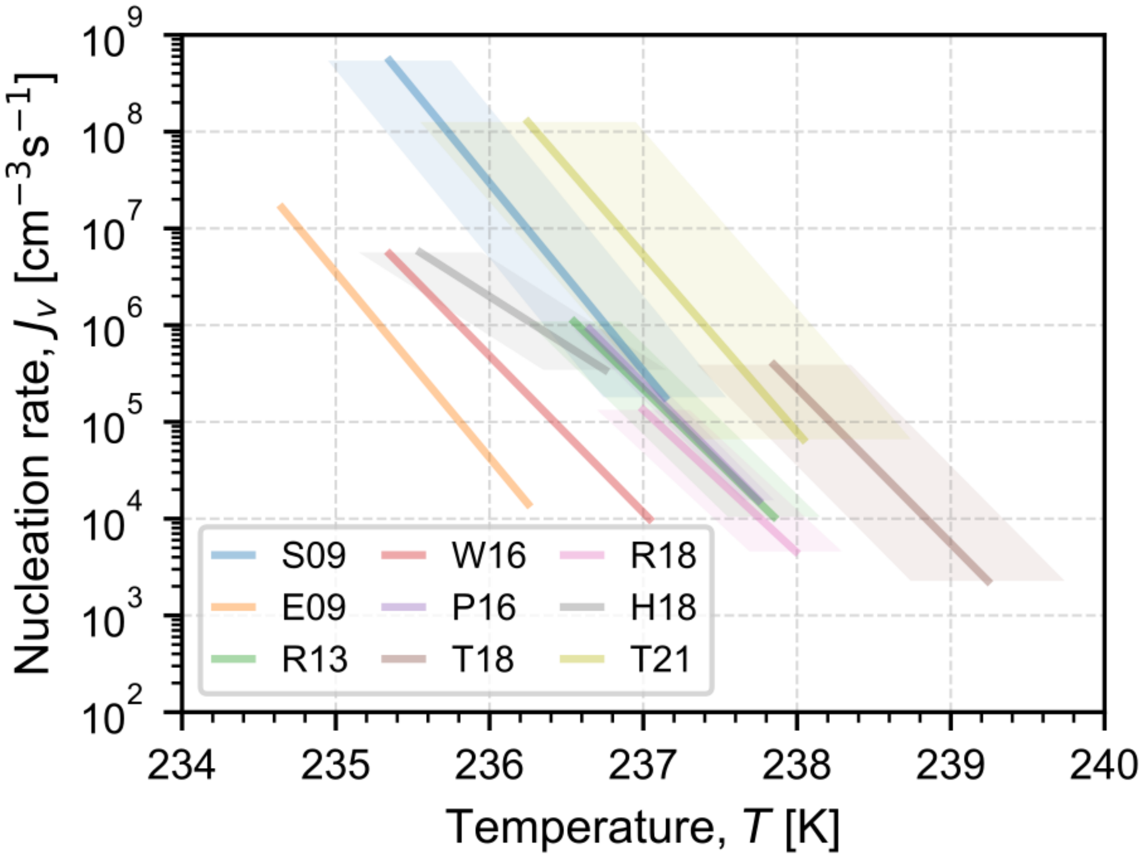
Nucleation rates from pL-droplet instruments

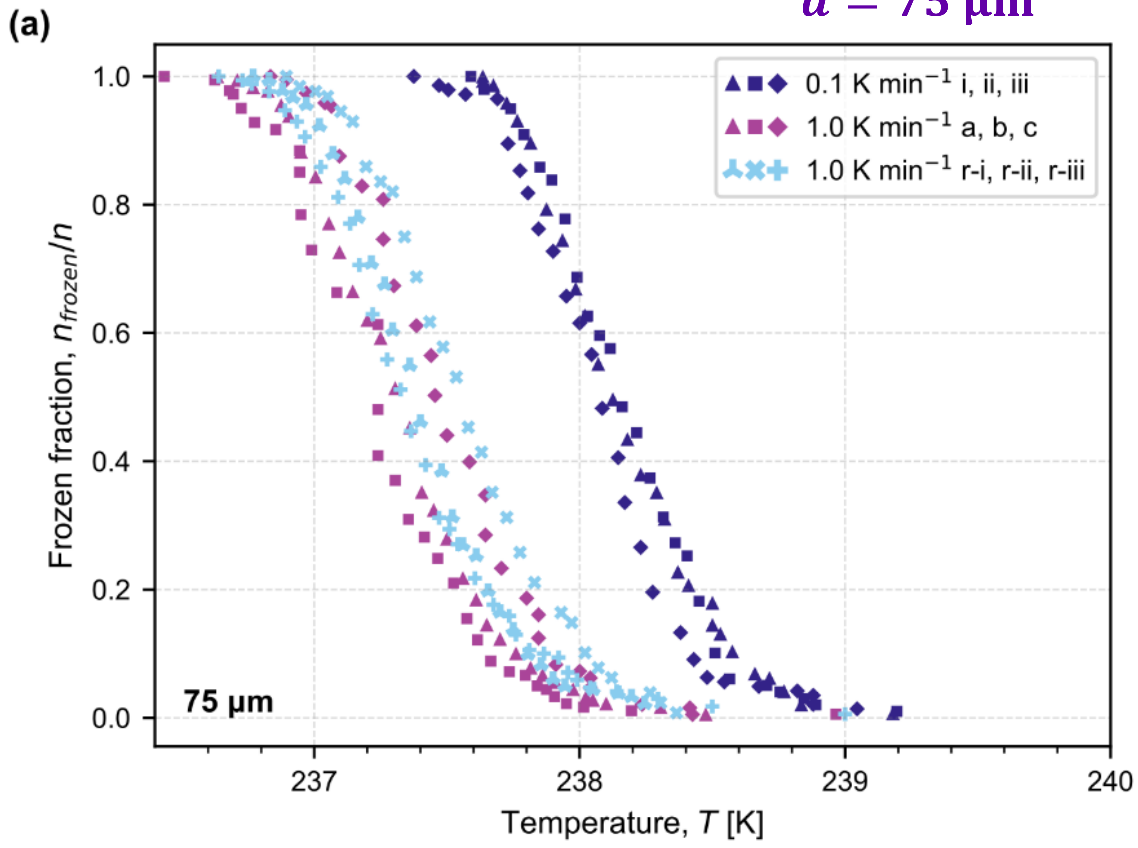


wide spread of nucleation rates across orders of magnitude…

large uncertainty

Fraction of droplets frozen vs. temperature

$d = 75\ \mu m$



Fraction of droplets frozen vs. temperature

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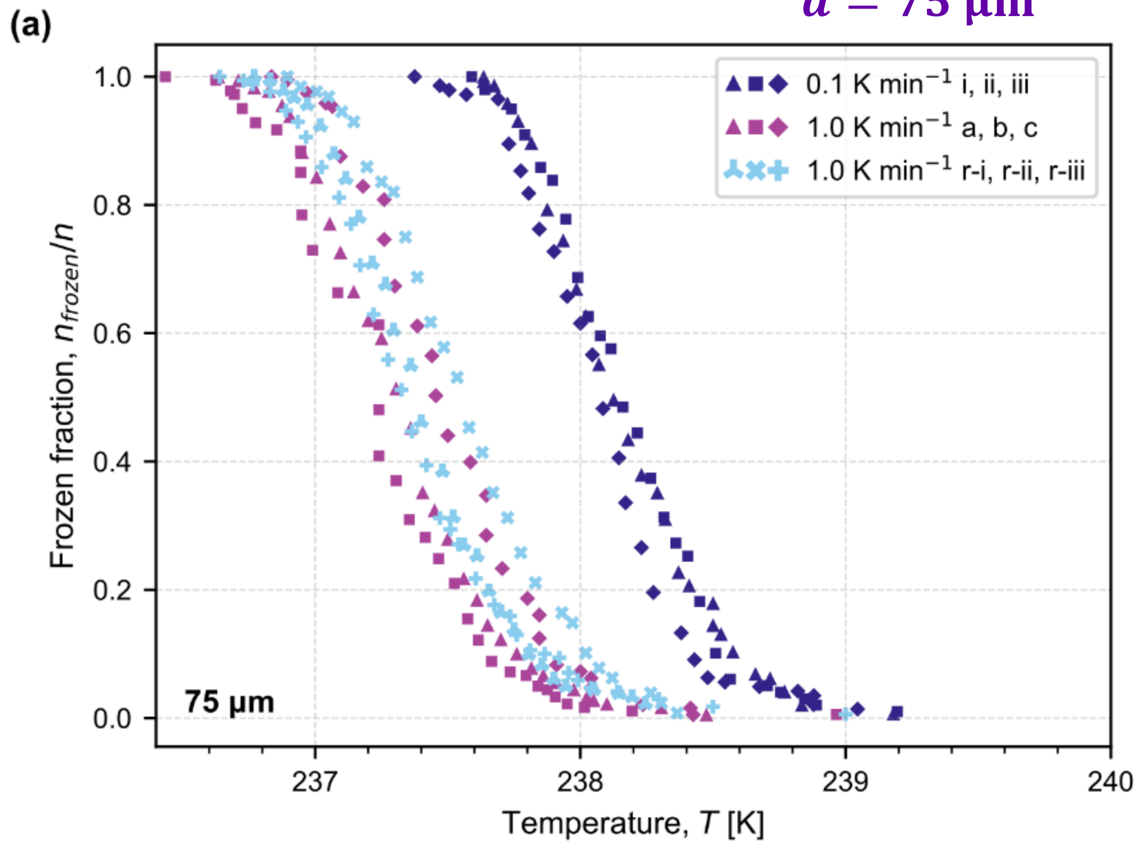
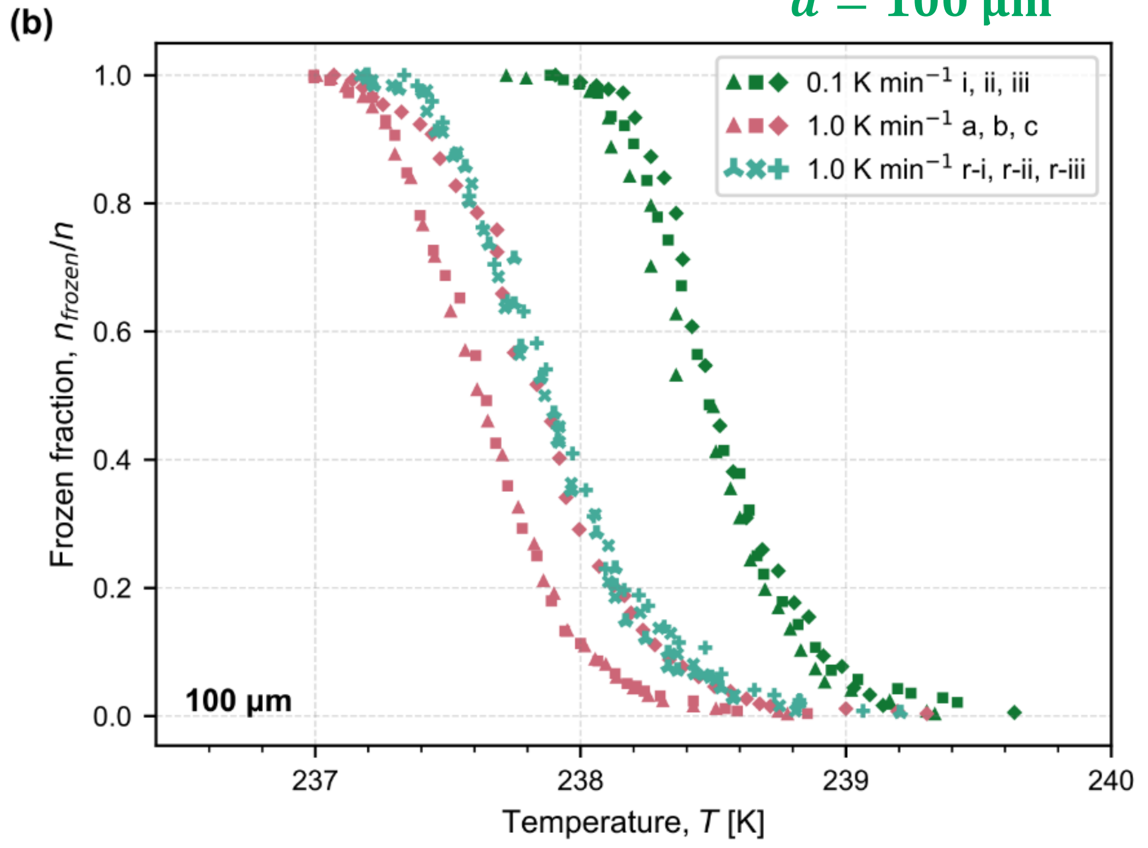


$d = 100\ \mu m$



Nucleation rate J_V from frozen fraction

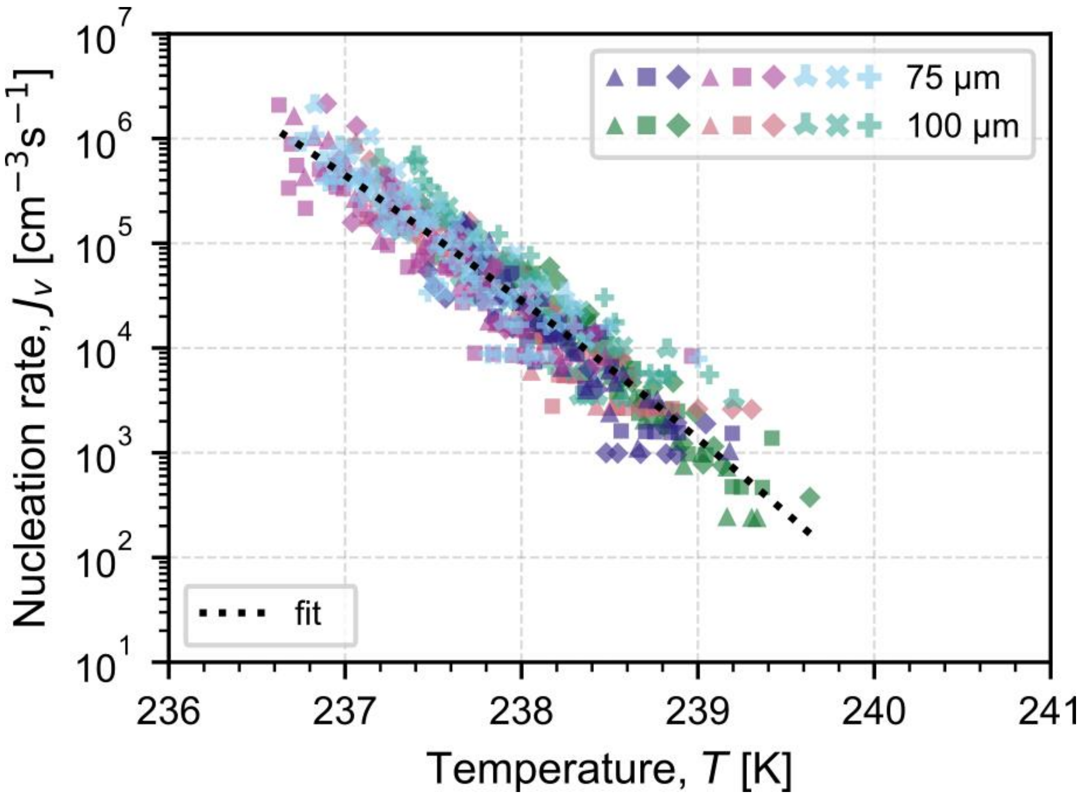
$$J_\mathrm{V} = -\frac{1}{V(t_2 - t_1)} \ln\left(\frac{1 - f_2}{1 - f_1}\right)$$

where V is droplet volume

$t_2 - t_1$ is an increment in time

f is the fraction of droplets that are frozen

Experimental homogeneous nucleation rate of water

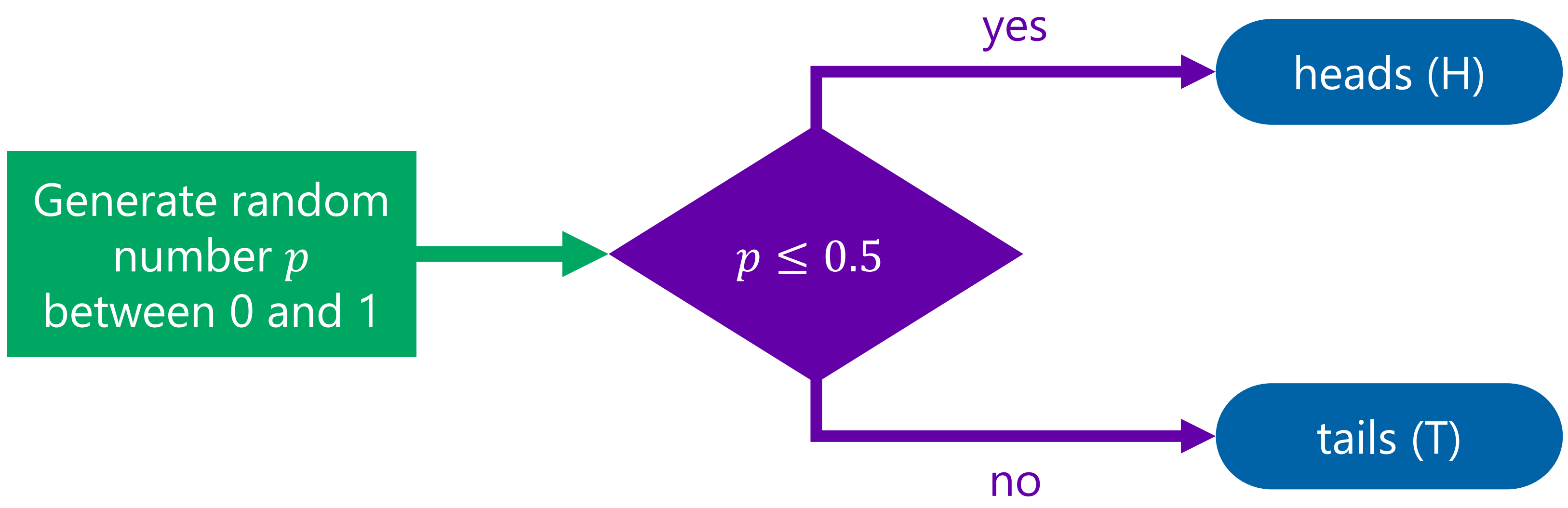


scatter from experimental error?
or scatter from randomness of nucleation?

use Monte Carlo simulation to determine
how much scatter comes from randomness

Monte Carlo simulation

example: coin toss



T T H T H H T H H T
T H H H H H T T H H
T H T H H H T T T H

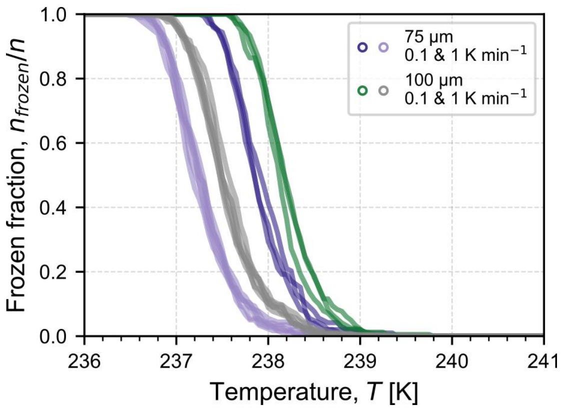
Monte Carlo simulation

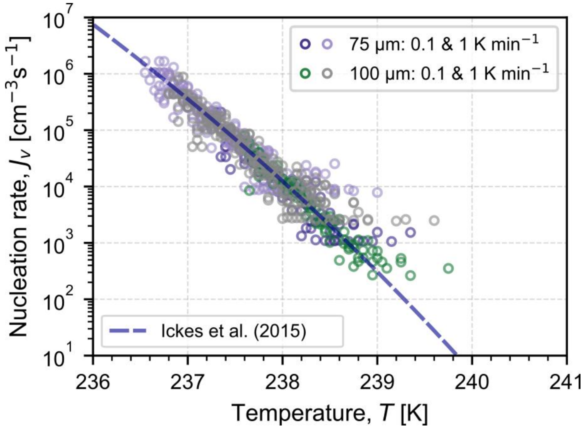
of a population of droplets being cooled

Generate a random number
for each droplet at every temperature

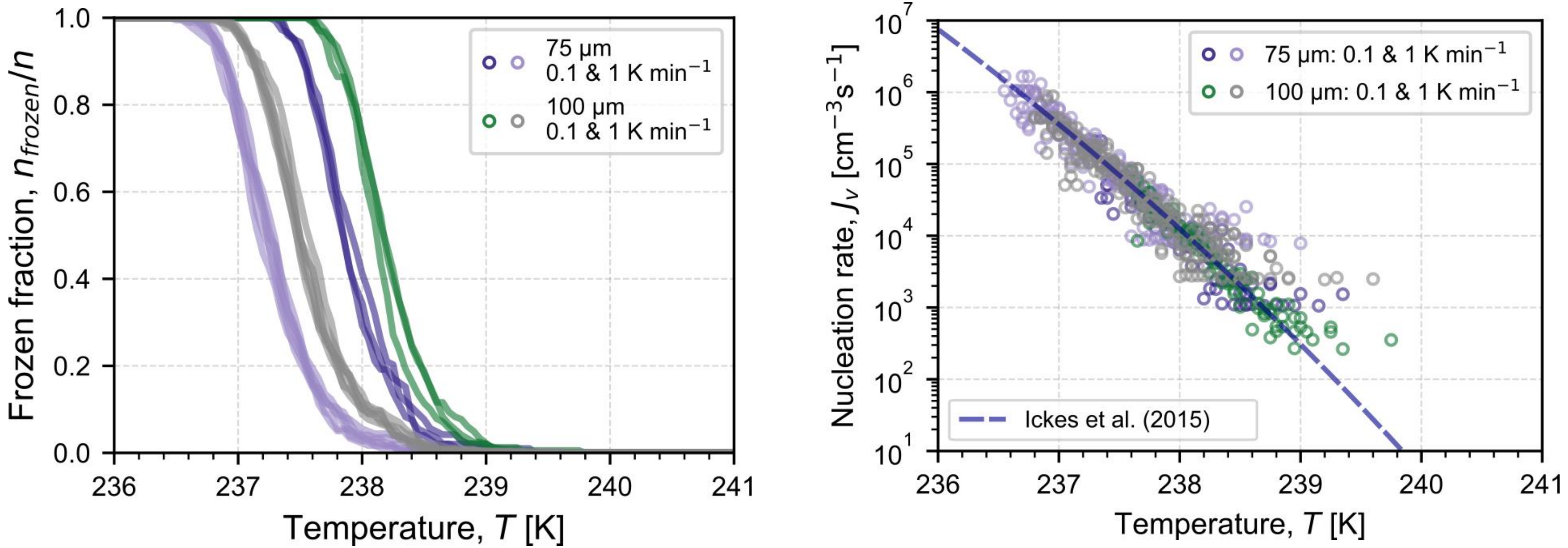
Compare random number
to probability of nucleation

Calculate nucleation rate
from simulated frozen fraction



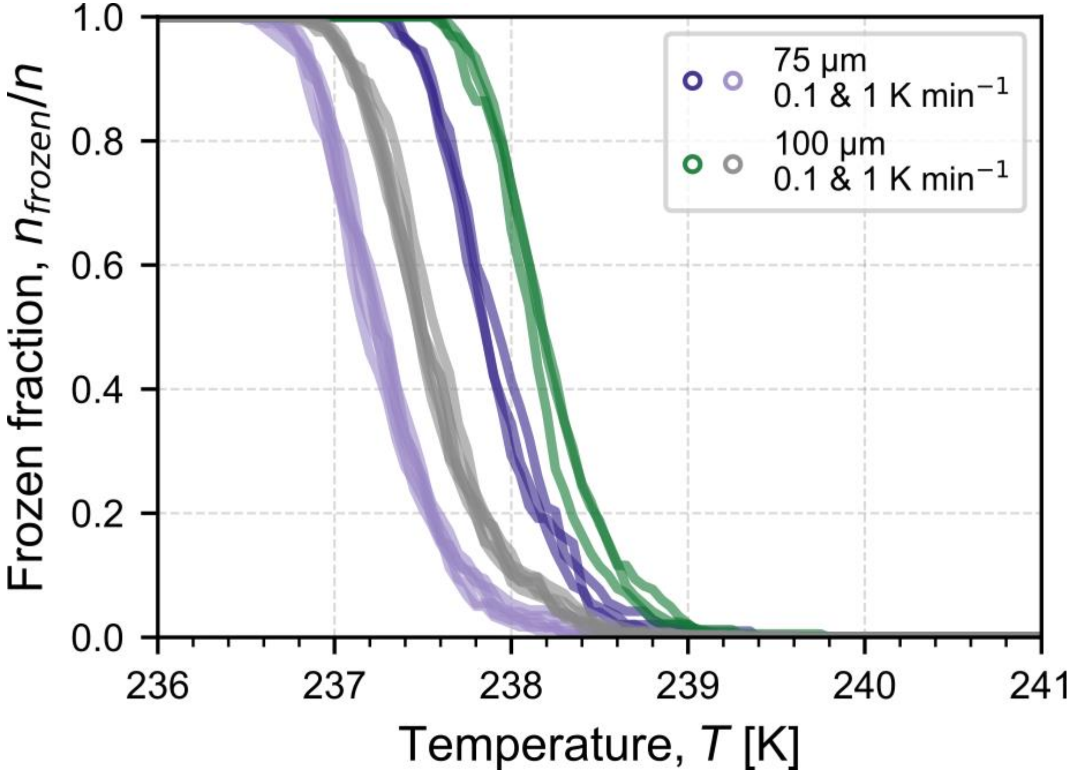


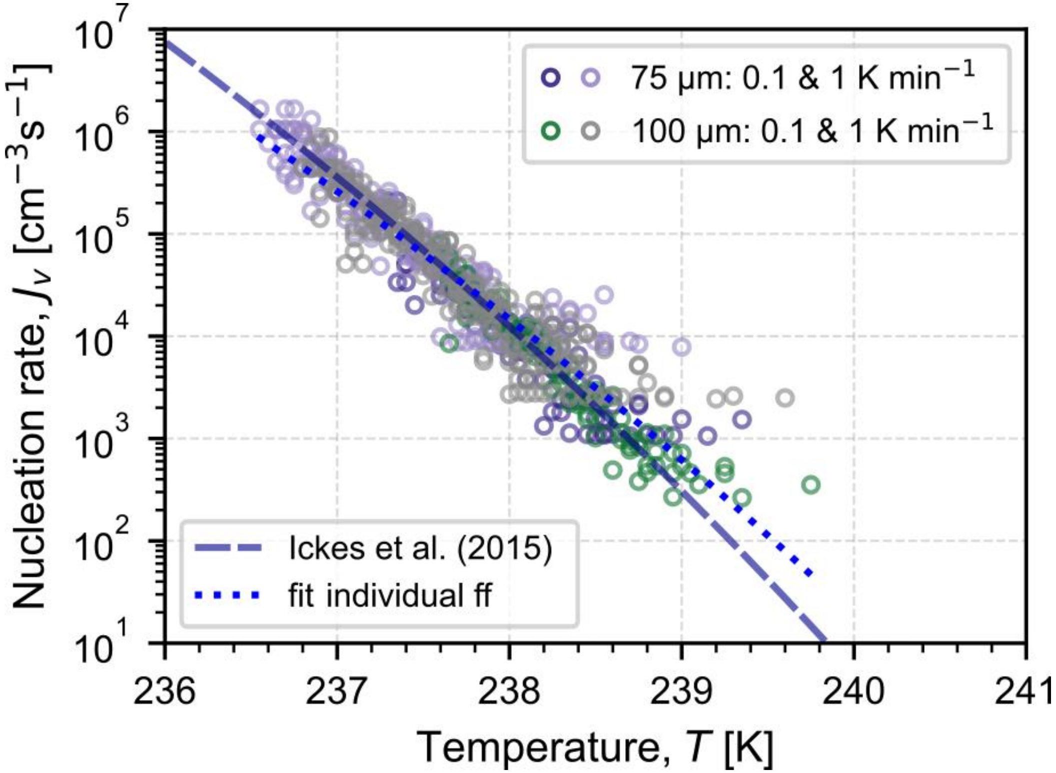
Monte Carlo simulation of experiments



∴ scatter from randomness of nucleation

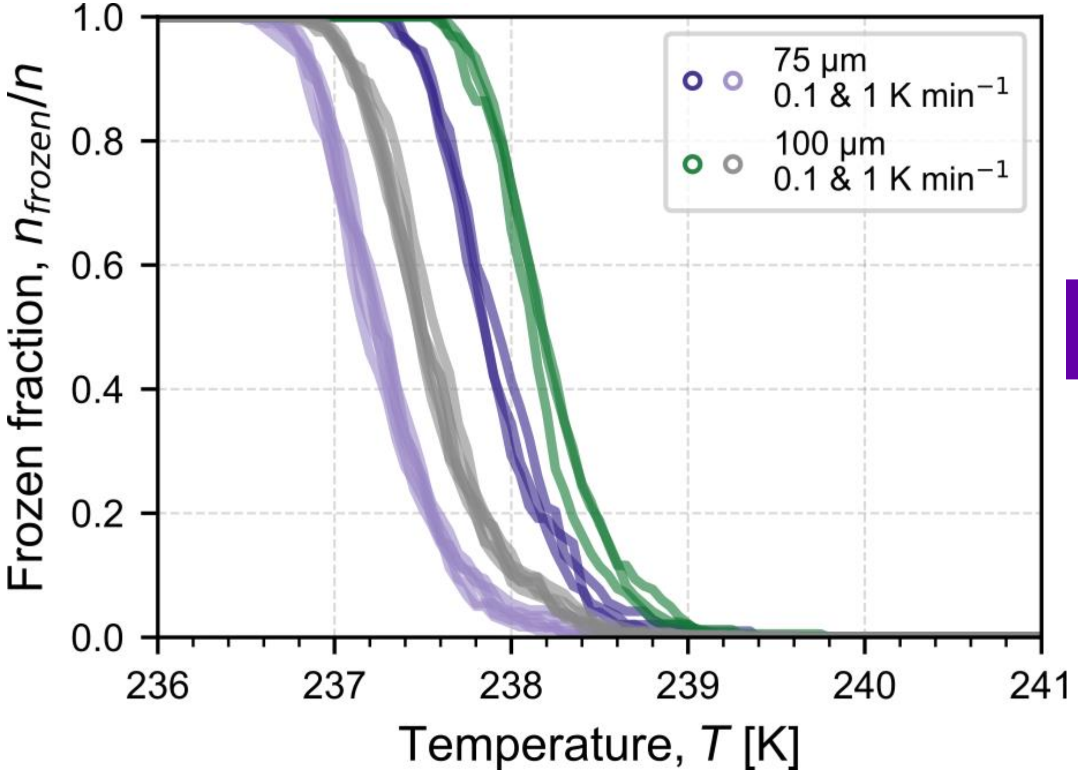
Monte Carlo simulation of experiments

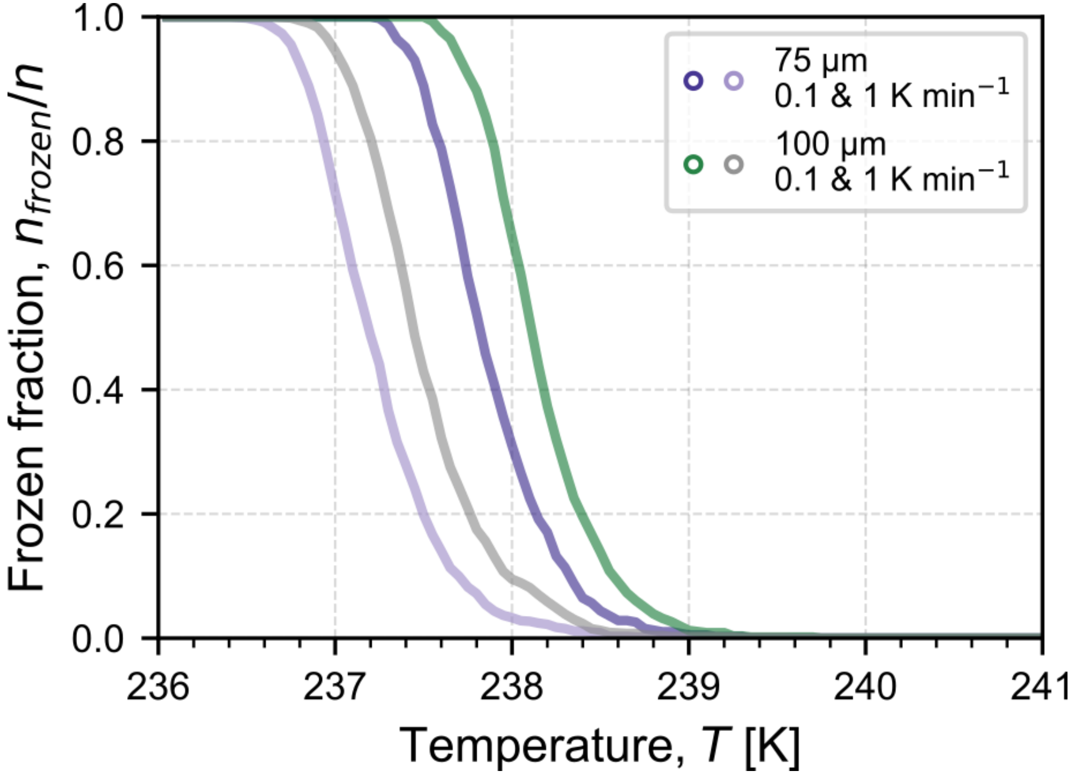




fitting to simulated nucleation rates does not recover underlying nucleation rate

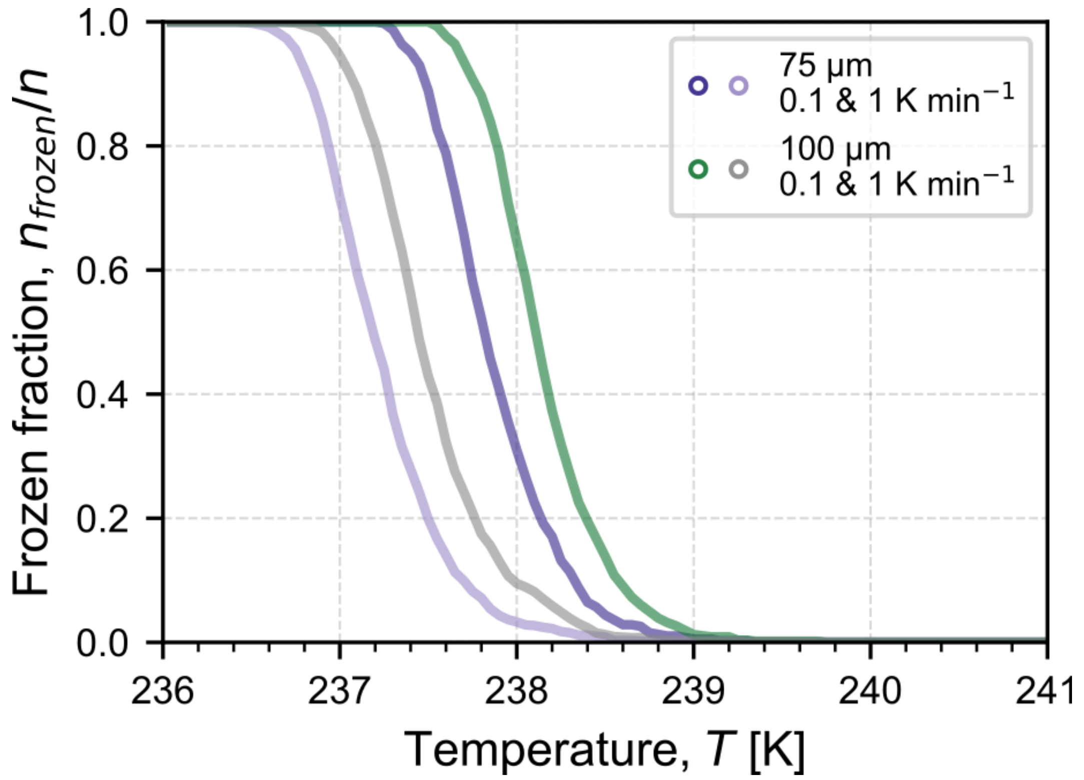
Monte Carlo simulation of experiments

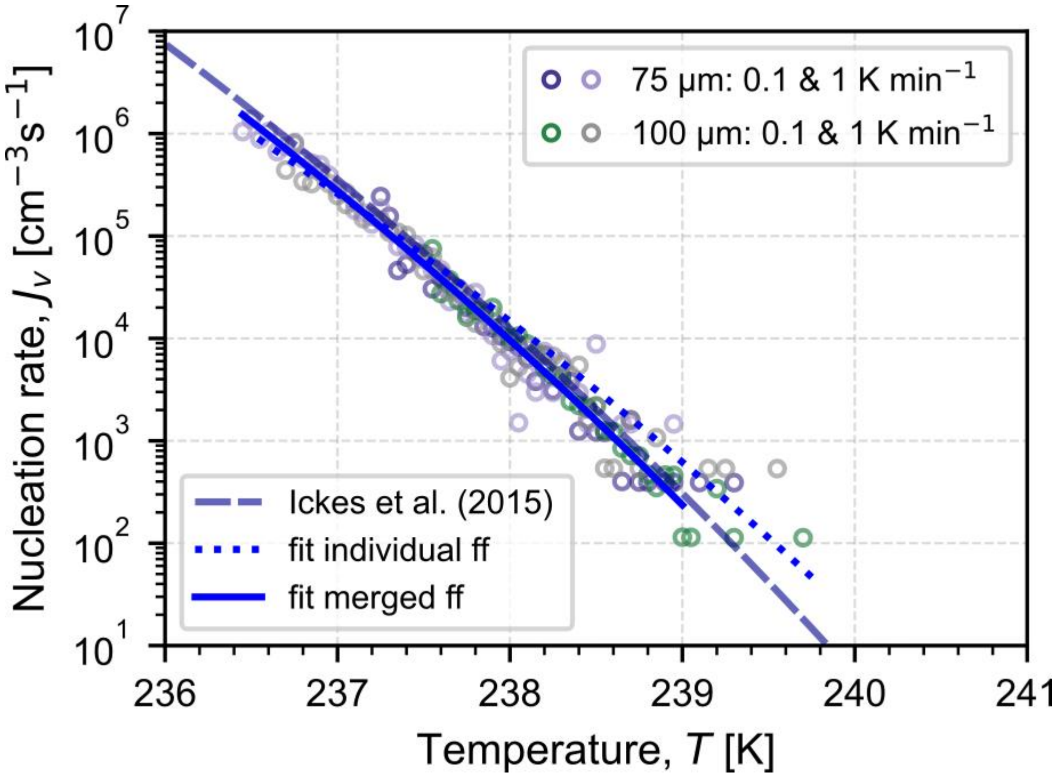




merge all droplets at same conditions to one frozen fraction curve

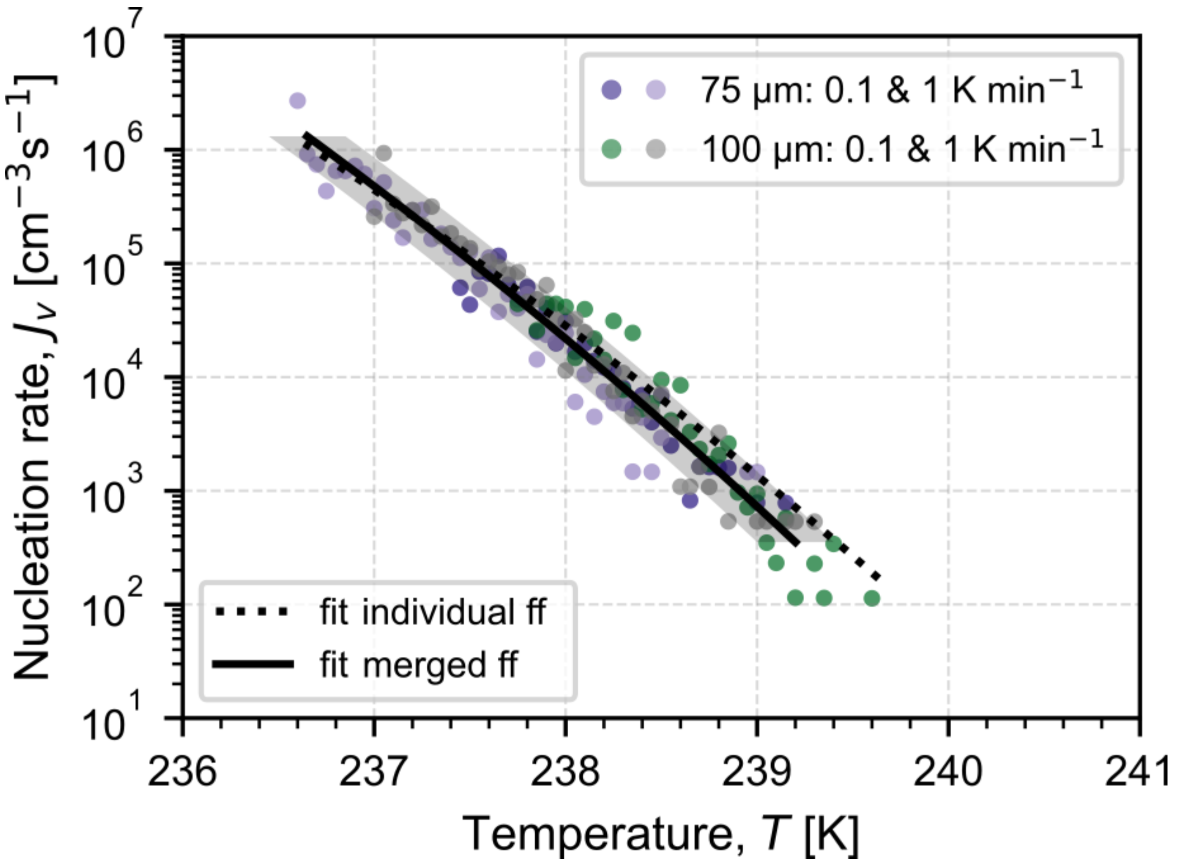
Monte Carlo simulation of experiments



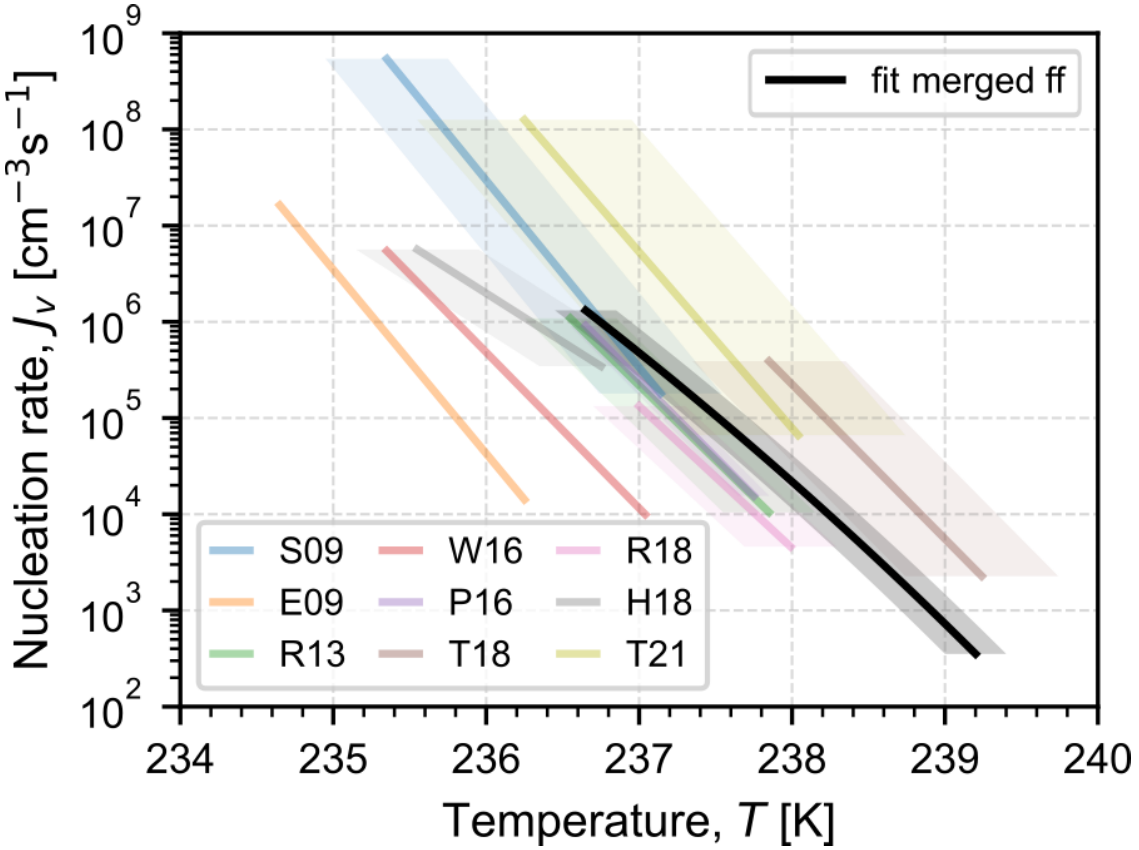


reduced scatter using merged frozen fraction
= can recover underlying rate

Experimental homogeneous nucleation rate of water



Comparison to other pL-droplet instruments



obtained new high-accuracy data over a wide temperature range

Outline

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Homogeneous freezing of water droplets for different volumes and cooling rates†

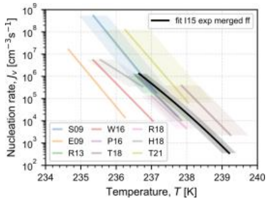
Nadia Shardt,*[a] Florin N. Isenrich,*[b] Benedikt Waser,[b] Claudia Marcolli,*[a] Zamin A. Kanji,[a] Andrew J. deMello[b] and Ulrike Lohmann[a]

1. New microfluidics-based instrument — Isenrich et al. **2022** *AMT*, 15, 5367–5381.

2. Homogeneous nucleation rate of water — Shardt et al. **2022** *PCCP*, 24, 28213.

Conclusions

1. Developed new microfluidics-based instrument Isenrich et al. **2022** *AMT*, 15, 5367–5381.



2. Precisely measured the nucleation rate of water Shardt et al. **2022** *PCCP*, 24, 28213.
   - Wide temperature range with 75 & 100 µm droplet diameters cooled at 0.1 and 1.0 K min^{-1}
   - Insight from Monte Carlo simulations for obtaining a parameterization of nucleation rate

Conclusions

1. Developed new microfluidics-based instrument Isenrich et al. **2022** *AMT,* 15, 5367–5381.

2. Precisely measured the nucleation rate of water Shardt et al. **2022** *PCCP,* 24, 28213.
   - Wide temperature range with 75 & 100 µm droplet diameters

Improved our understanding of and ability to predict ice nucleation, aiming towards improved predictions of  and