

Thermal imaging of a shattering freezing water droplet

Internal pressure can be derived from IR measurements of the droplet temperature

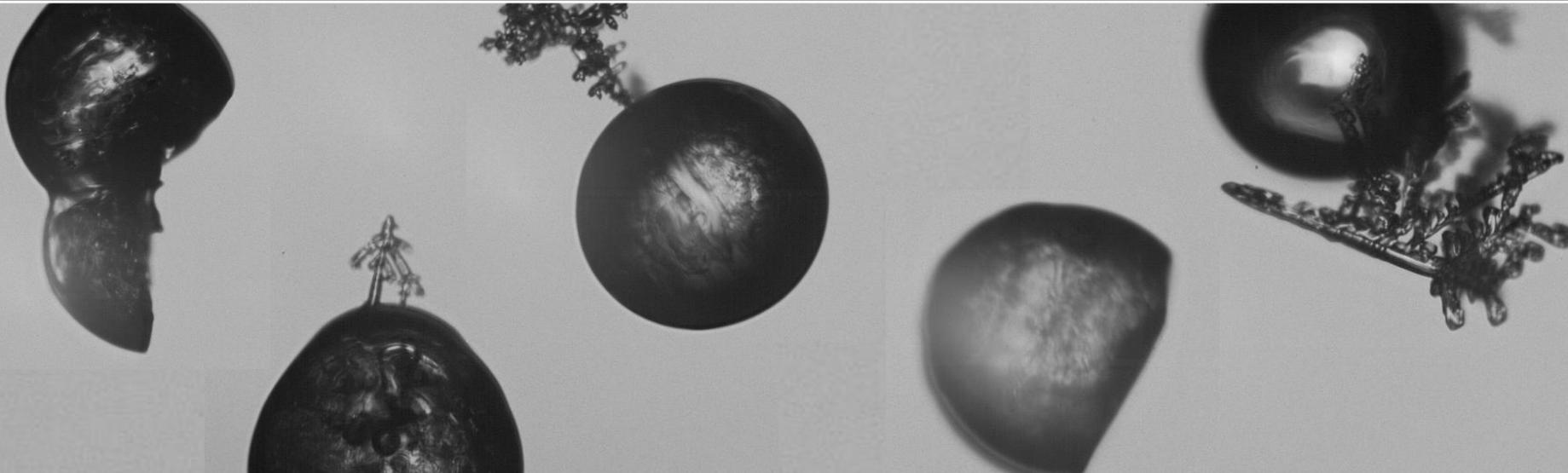
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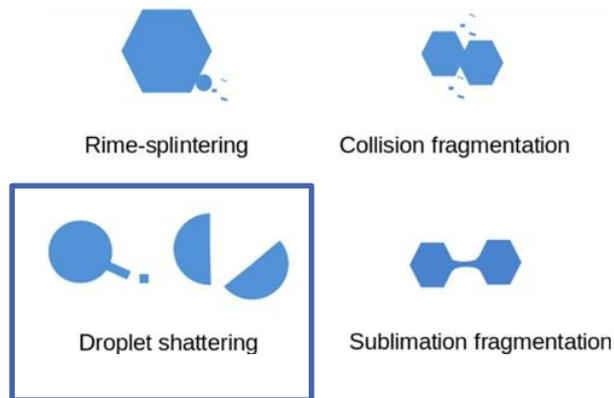
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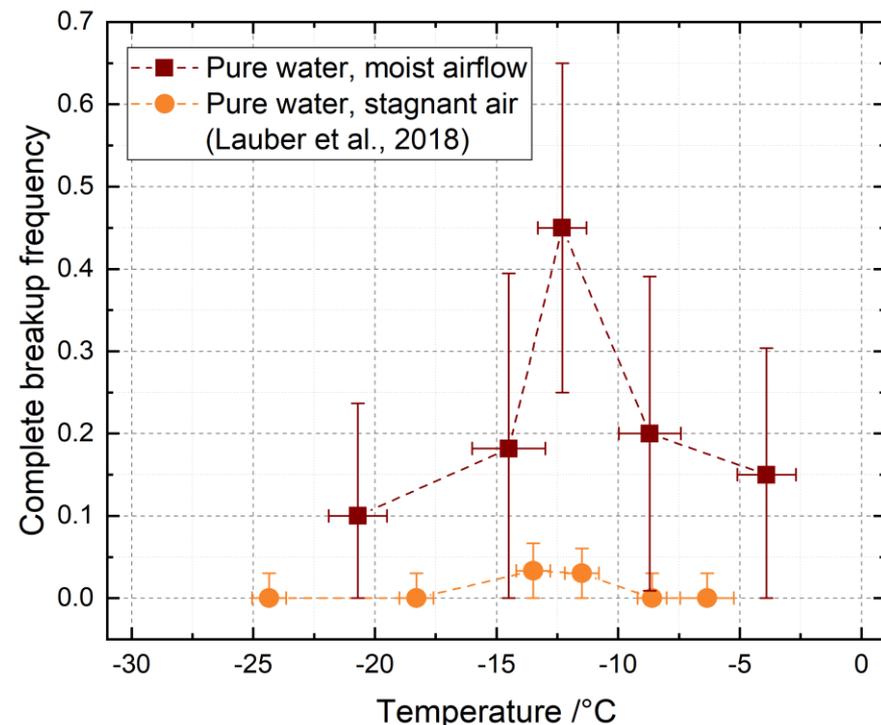
Secondary ice production upon freezing of water drops

In mixed-phase clouds, a strong discrepancy between the number concentration of ice particles and ice nucleating aerosol particles has been observed. Several secondary ice production (SIP) mechanisms are assumed to be responsible for this discrepancy. *This study focusses on the mechanism of droplet shattering upon freezing.*

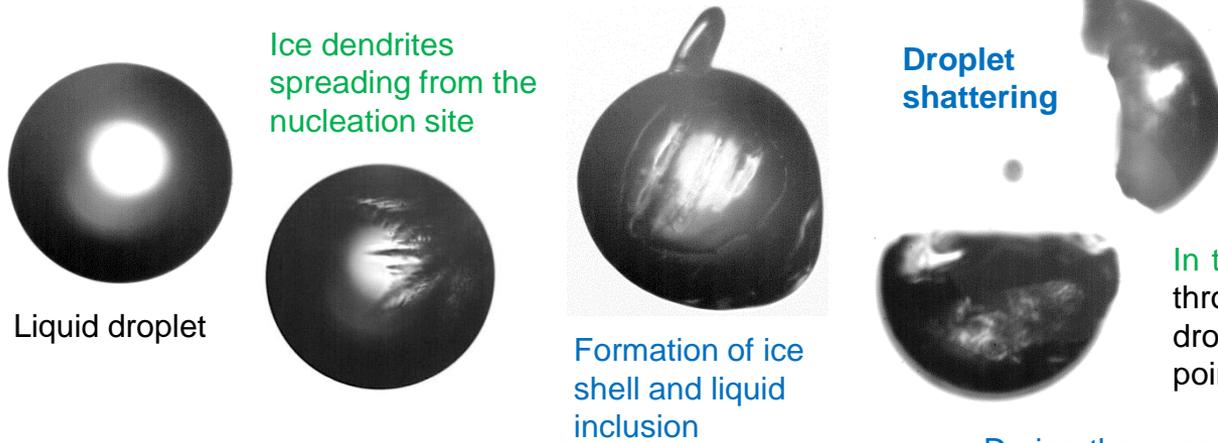


Secondary ice production (SIP) mechanisms described in Field et al. (2017)

In our previous study (Keinert et al., submitted to JAS), the complete breakup of freezing droplets was found to be more frequently happening in free fall. Moreover, the maximum frequency of droplet shattering was found at around -13°C (see the figure below). For details, see the presentation of Alice Keinert (IMK KIT) available at <https://doi.org/10.5194/egusphere-egu2020-7609>



Freezing of a free-falling water droplet



Liquid droplet

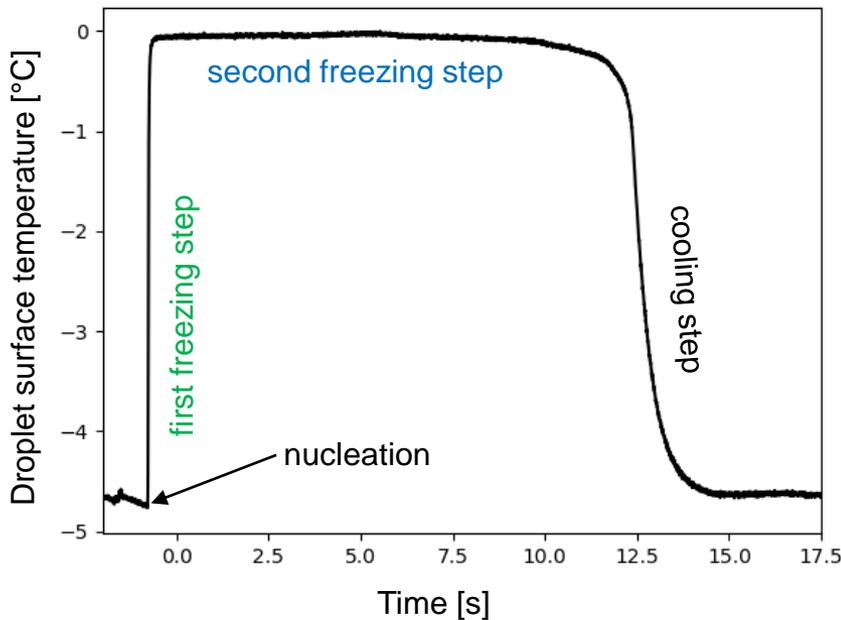
Ice dendrites spreading from the nucleation site

Formation of ice shell and liquid inclusion

Droplet shattering

Freezing of a free-falling water droplet proceeds in two steps:

In the **first freezing step**, ice dendrites spread through the droplet, releasing latent heat. The droplet acquires temperature of the melting point of ice.



Evolution of droplet temperature during freezing in stagnant air at $T = -5^{\circ}\text{C}$, droplet diameter $345\ \mu\text{m}$

During the **second freezing step**, the ice shell forms, which is slowly growing from outside in. The volume increase upon solidification of water leads to a **pressure rise** in the liquid phase and **stress in the ice shell**. As a result of the pressure rise, the droplet develops bulges and spikes, and also can crack, eject bubbles or jets, or even **shatter**. Such events are known to produce **secondary ice particles**.

To understand the shattering of droplets, the **mechanical processes** during freezing need to be quantified. A mechanical modelling requires a thorough description of **thermal processes** governing the freezing of water droplets.

This is achieved by means of:

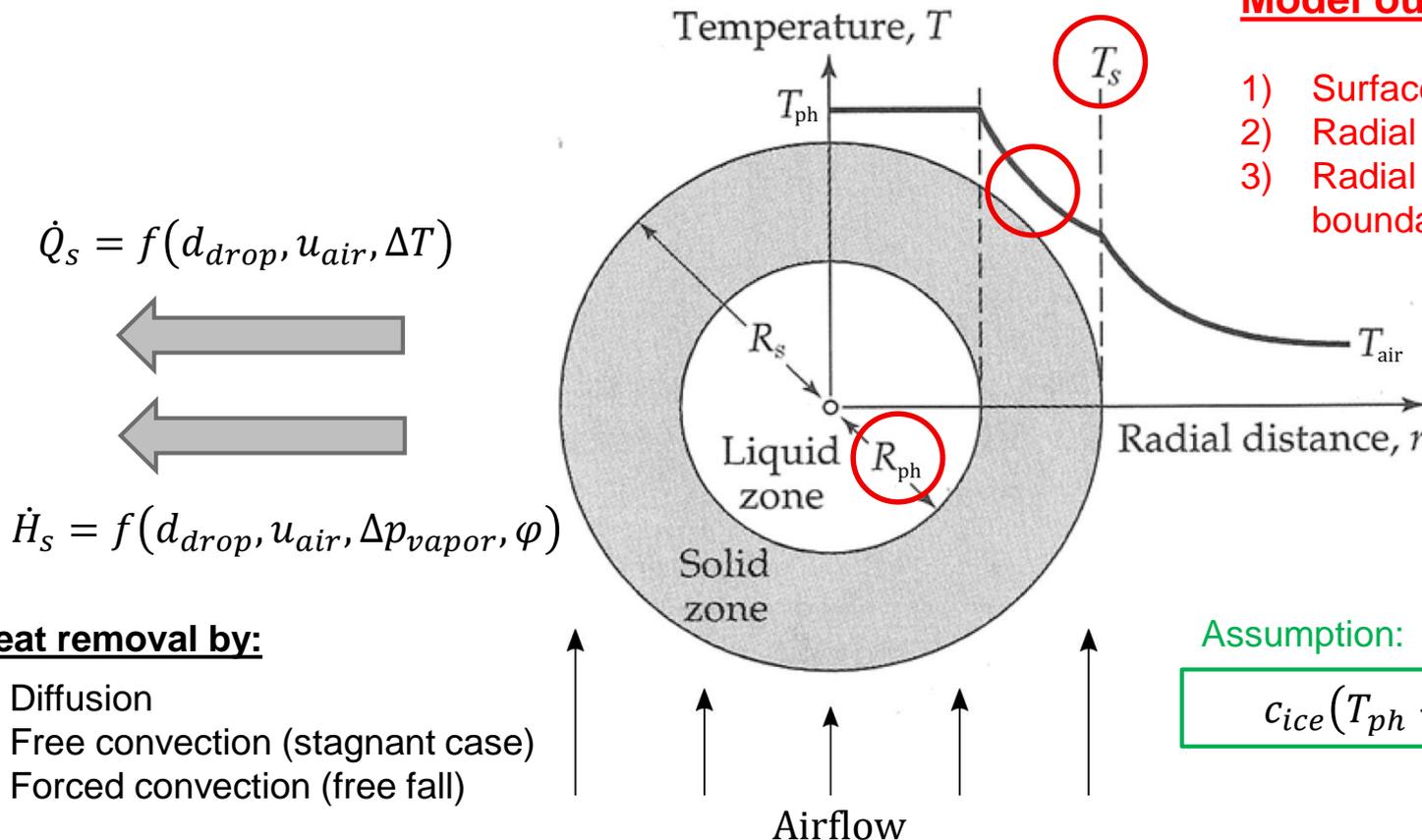
- 1) **Thermal modelling** and
- 2) **Infrared measurements** of drizzle-sized droplets freezing in free-fall conditions.

Thermal modelling of second freezing step

Second freezing step and cooling step were modelled separately. During freezing, the **enthalpy change of the ice shell is neglected (see Assumption)** to simplify the numeric solution algorithm. During the cooling stage, the cooling of the ice shell is explicitly taken into account.

Model output:

- 1) Surface temperature
- 2) Radial temperature profile
- 3) Radial position of phase boundary



Heat removal by:

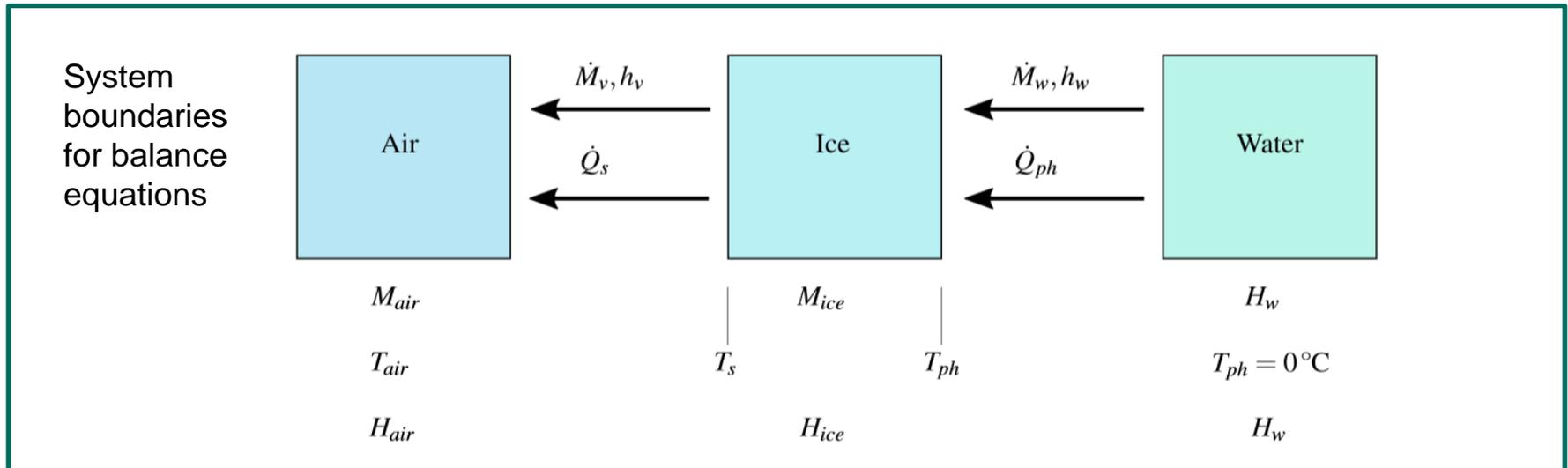
- 1) Diffusion
- 2) Free convection (stagnant case)
- 3) Forced convection (free fall)

Assumption:

$$c_{ice}(T_{ph} - T_s) \ll \Delta h_{s,l}$$

Thermal modelling of second freezing step

The solution was obtained by setting up the enthalpy and mass balance equations for the ice phase and for the water phase.



The resulting equations for the **movement of the phase boundary** and the **temperature profile** are interconnected and have been solved simultaneously using the *explicit time forward method*.

$$\frac{dR_{ph}^3}{dt} = - \frac{\dot{H}_s(T_s) + \dot{Q}_s(T_s)}{C}$$

$$T(\mathbf{R}) = T_{ph} + \frac{\dot{H}_s + \dot{Q}_s}{\lambda_{ice} 4\pi r_d} \left(\frac{R_{ph}(t) - R}{RR_{ph}(t)} \right)$$

$$T_s(\mathbf{t}) = T_{ph} + \frac{\dot{H}_s + \dot{Q}_s}{\lambda_{ice} 4\pi r_d} \left(\frac{R_{ph}(t) - 1}{R_{ph}(t)} \right)$$

Model output for the reference case

A model case study for the reference case: droplet freezing in free fall.

Reference case conditions:

Drop size: $d_d = 330 \mu m$

Ambient air temperature:

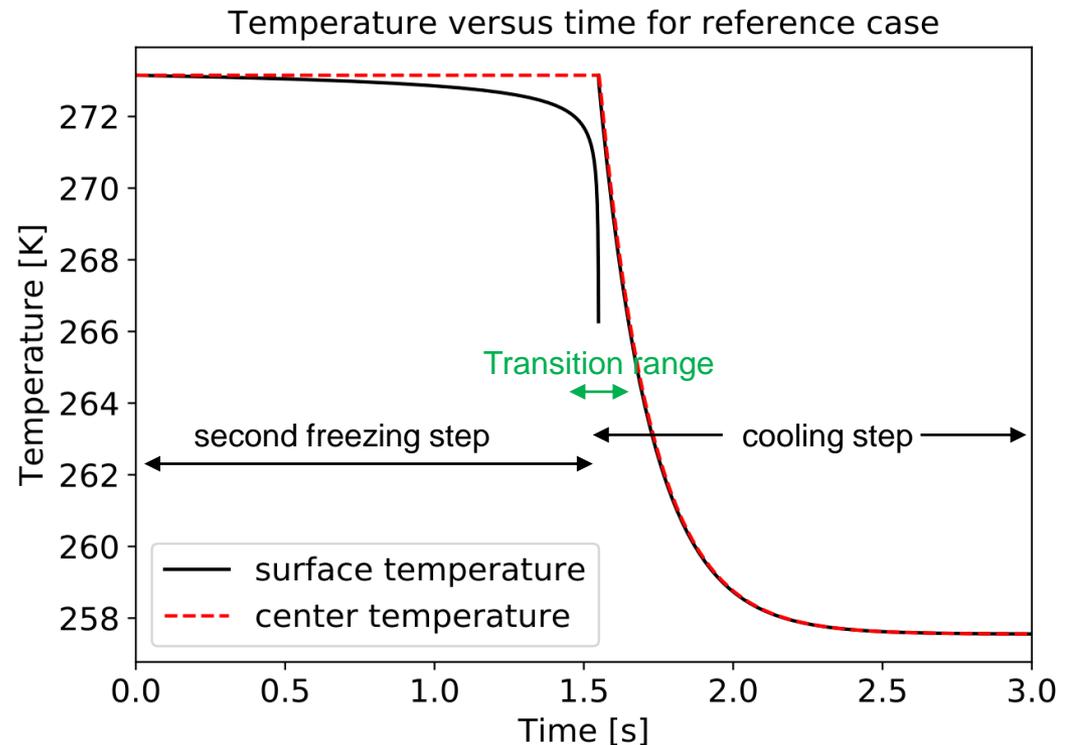
$$T_{air} = -15 \text{ }^\circ\text{C}$$

Airflow velocity $u_{air} = 1.3 \text{ m/s}$

Fully developed forced convection

Relative humidity wrt ice:

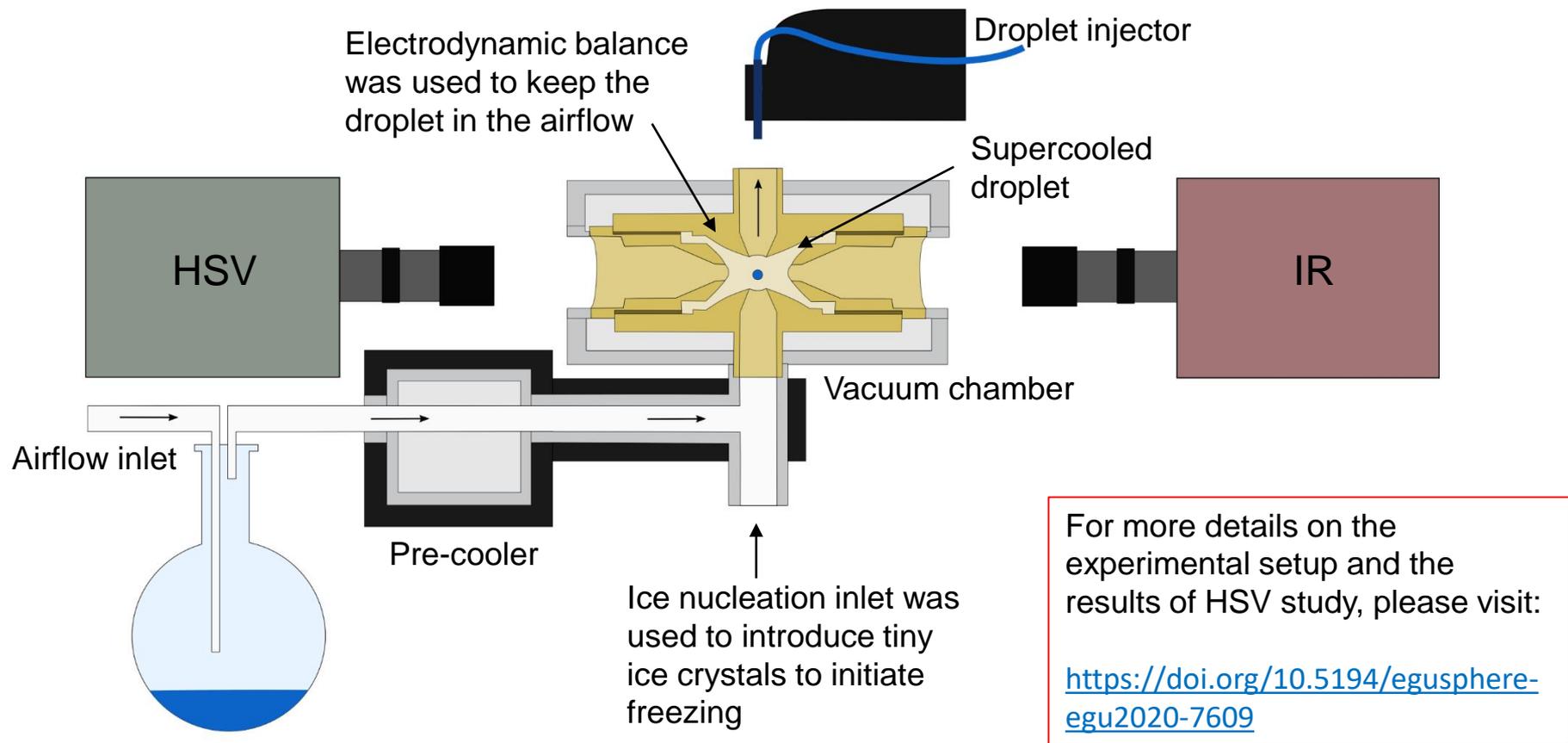
$$\varphi = 100 \%$$



With increasing thickness of the ice shell, the surface temperature decreases. In the **transition range** the modelled surface temperature shows unphysical behavior. This is the result of the **assumption** (neglection of the enthalpy change of ice, see slide 4).

Experimental setup

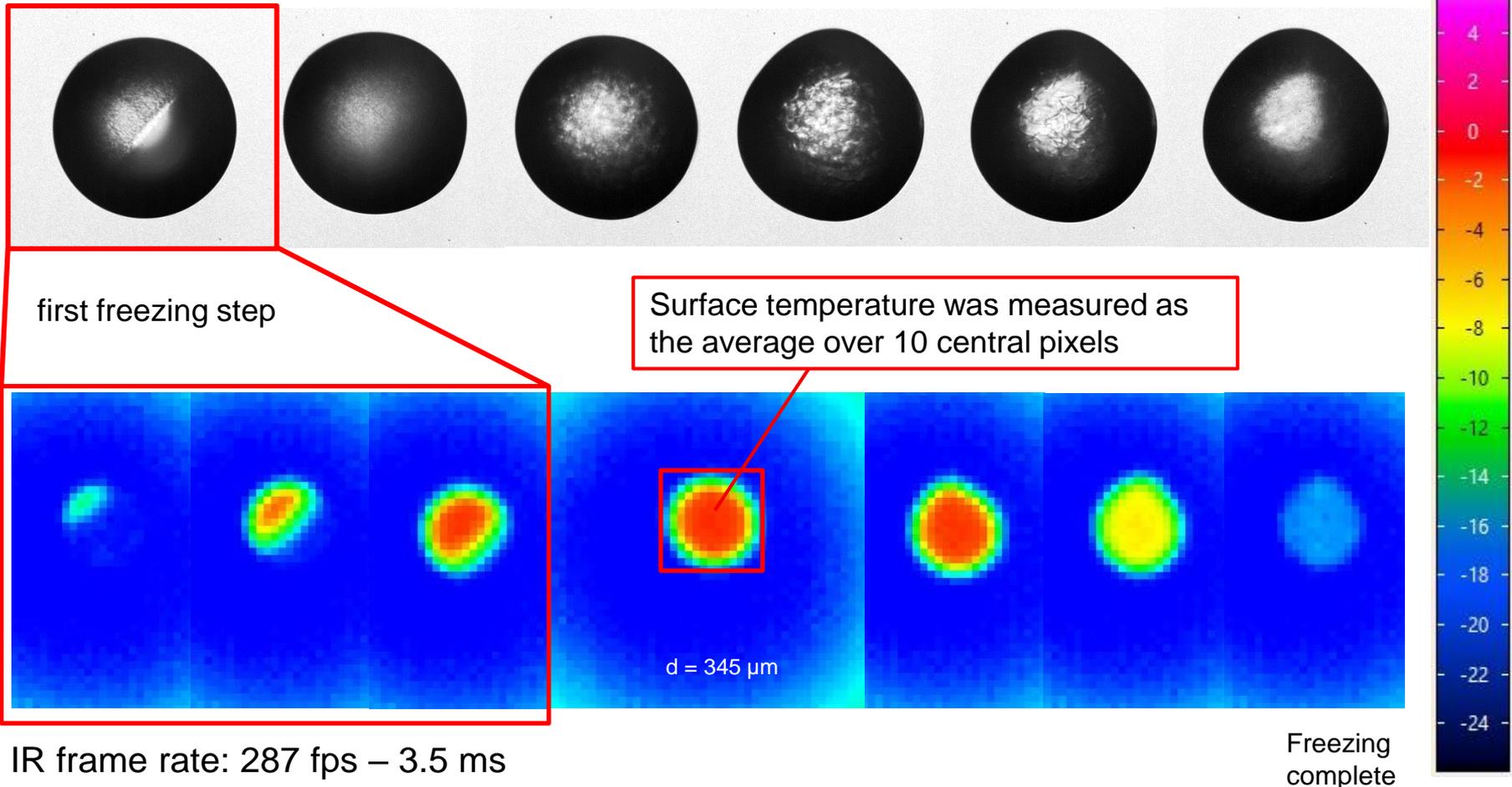
Droplets of (330 ± 10) μm diameter were levitated in the cooled electrodynamic balance (Paul trap). To simulate free fall, the droplets were exposed to an upstream humidified airflow. The freezing process was recorded with the high-speed video (HSV) camera (Phantom v710, *Vision Research*) and the infrared (IR) camera (*InfraTec*).



Results: synchronous HSV and IR records

HSV frame rate: 2000 to 9000 fps – 0.5 to 0.1 ms

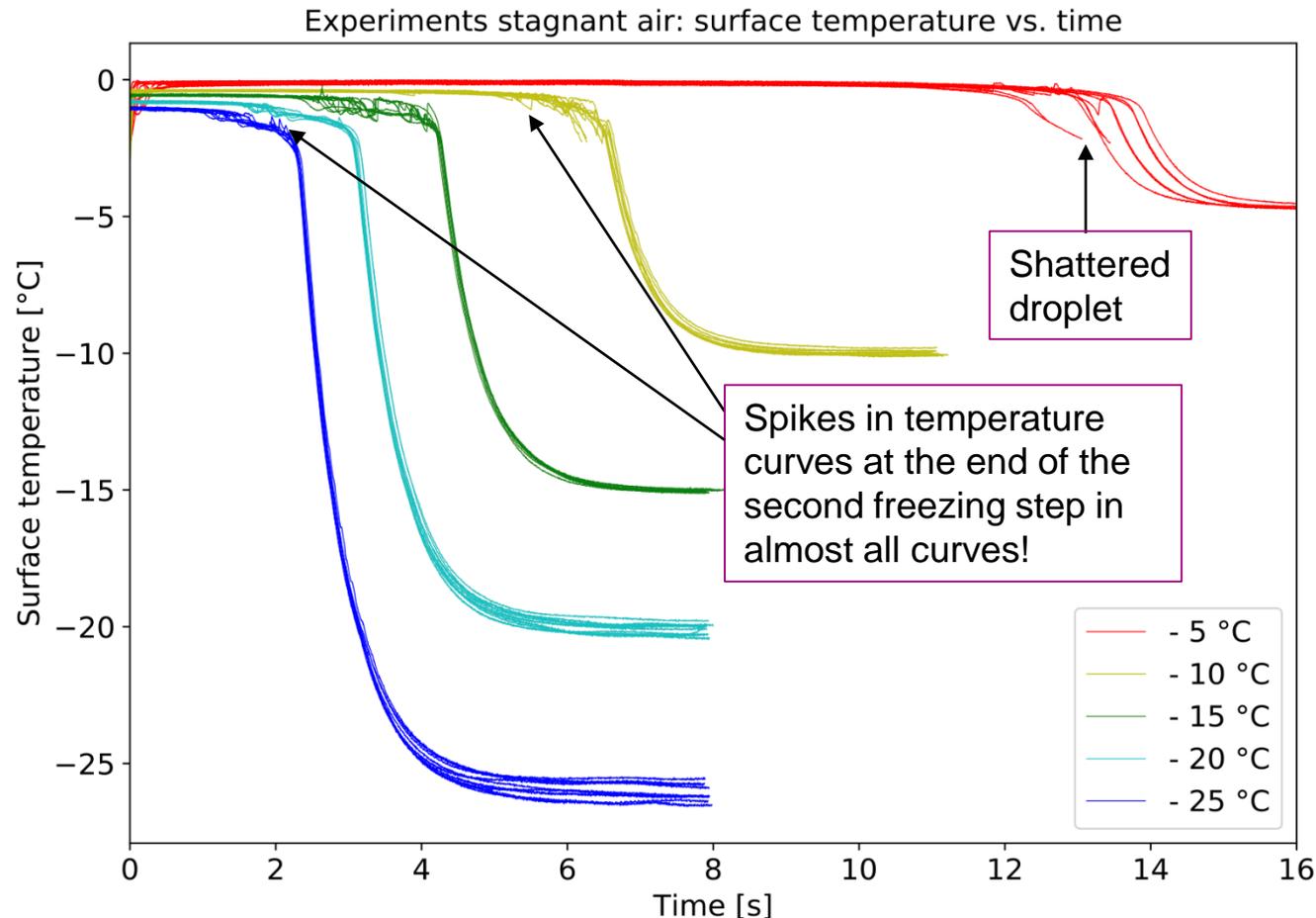
d = 345 μ m



Surface temperature: stagnant case

Surface temperature was measured for 10 droplets for every temperature setting. The temperature curves show very good reproducibility.

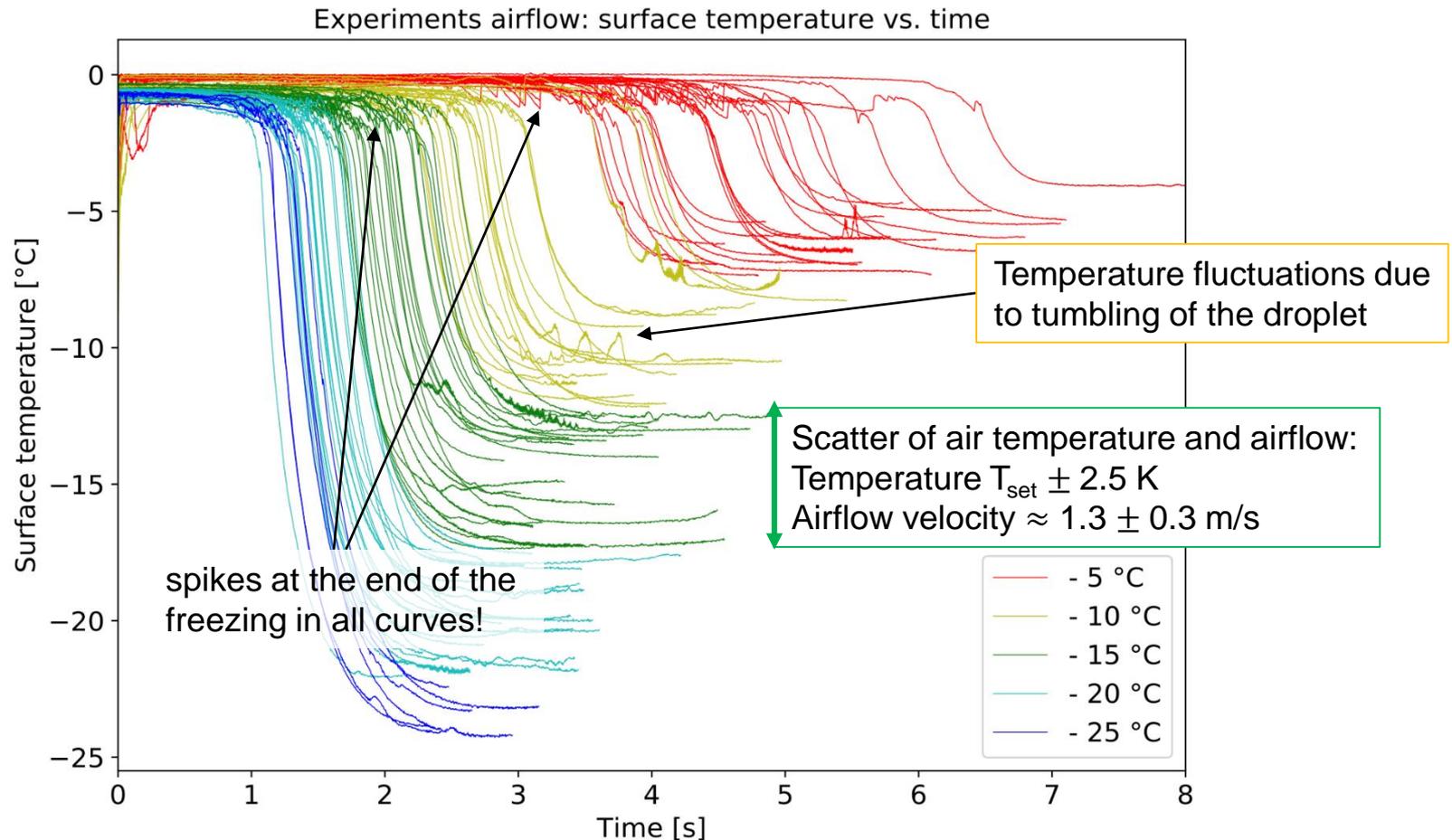
Note the spikes and fluctuations at the end of freezing visible in every curve!



Surface temperature: airflow case

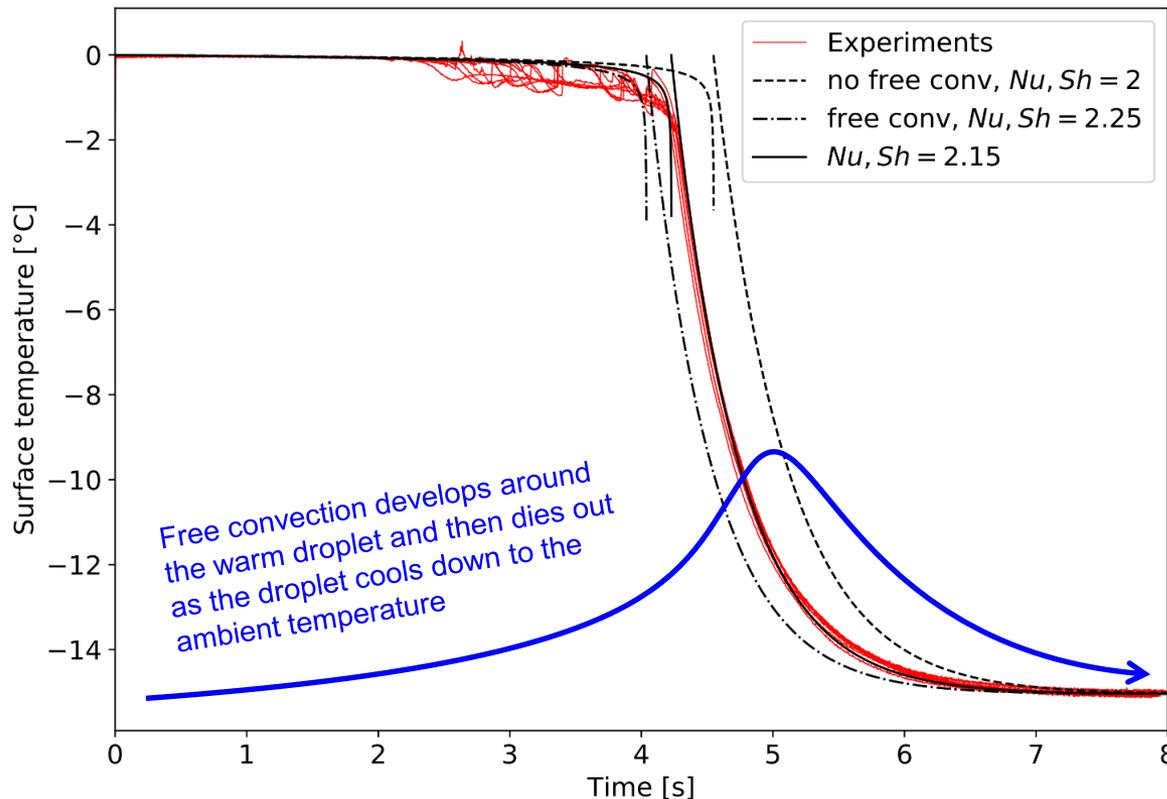
Surface temperature was measured for 25 droplets for every temperature setting. The actual temperature curves scatter due to variability in airflow and temperature.

Note even more spikes and fluctuations at the end of freezing visible in every curve!



Surface temperature: model vs. experiment

Good agreement of modelled surface temperature with experimental curves, except for the transition gap between second freezing and cooling step, as explained in the slide 6.



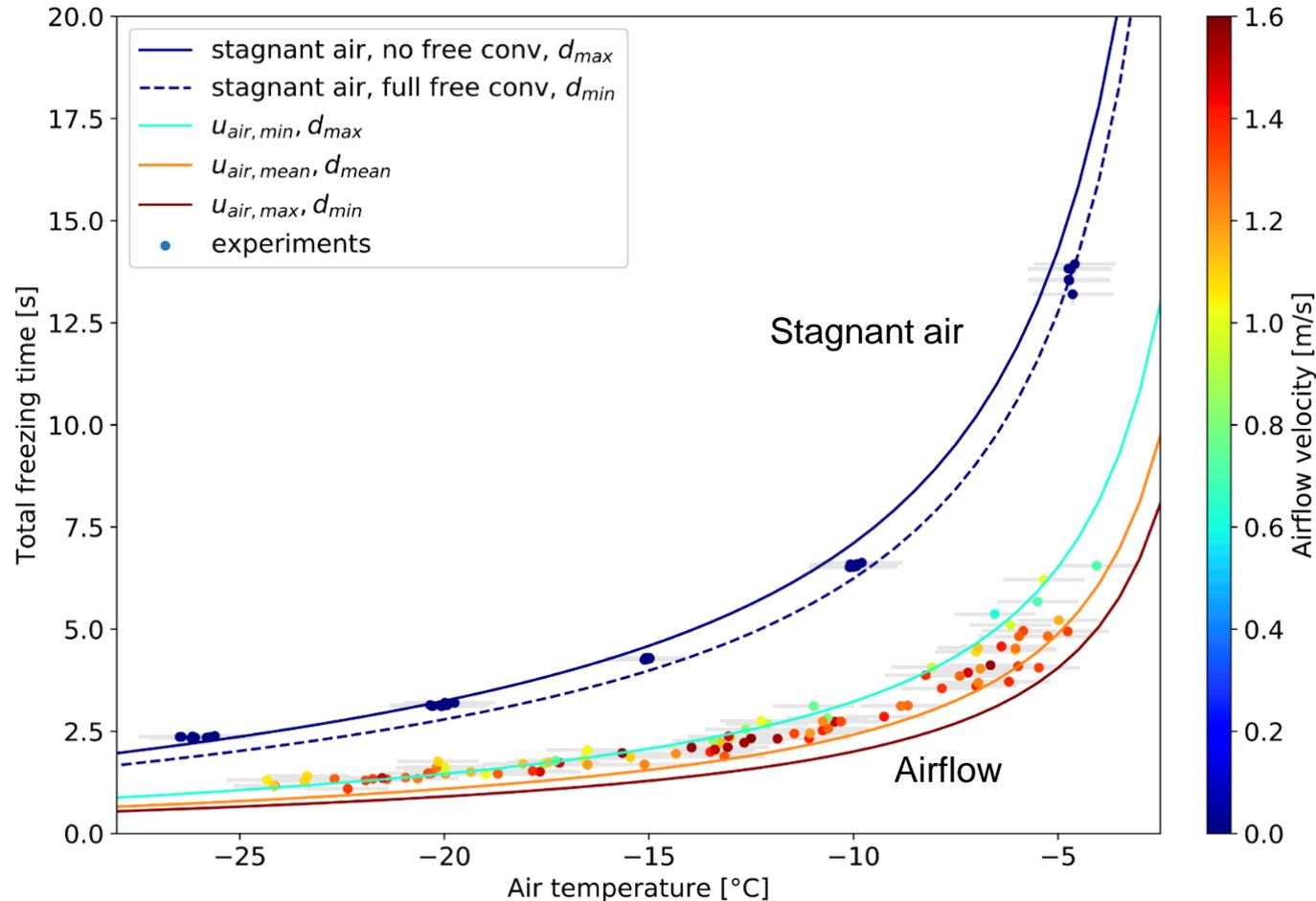
Comparison between the experimental surface temperature and the model for droplet freezing in stagnant air

Free convection:

In *stagnant air*, free convection around the droplet forms as the droplet is warmer than the ambient air. It is uncertain, to what extent the free convection is developing during the short freezing process. As the droplet cools down to ambient temperature, the free convection dies out.

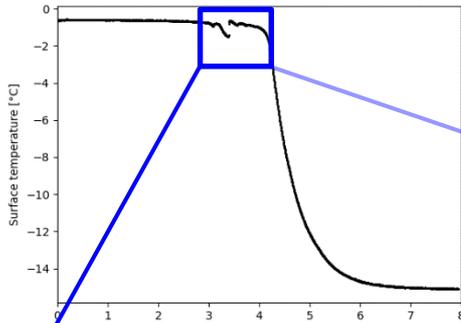
To account for the transient free convection, two extreme cases (no free convection and fully developed stationary free convection) have been modeled. *By choosing an intermediate Nusselt and Sherwood number, the model can be fitted to the experiment.*

Total freezing time: model vs. experiment



Taking into account the variability in temperature and airflow velocity, the **modelled total freezing time shows good agreement** with the experiments.

Temperature spikes give evidence for pressure build-up and release

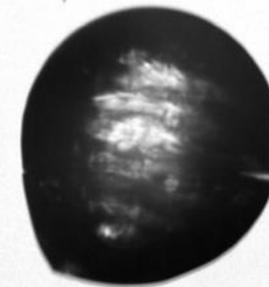
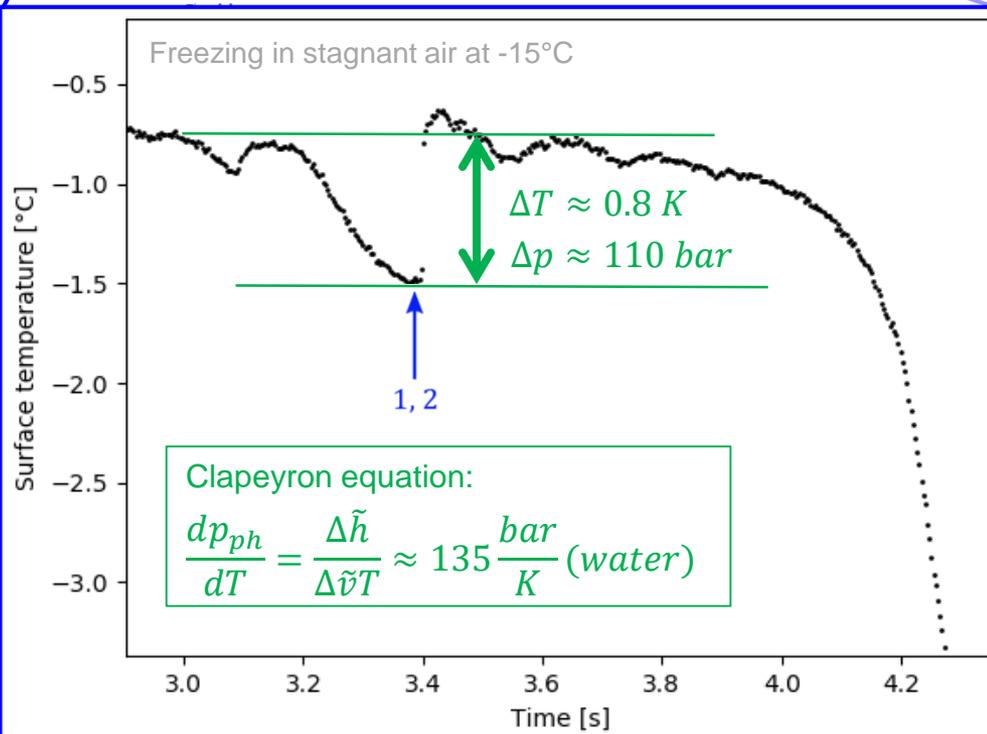


Major events of pressure release, like the incomplete break-up of this droplet, were always associated with a gradual decrease and sudden jump in temperature (spikes).

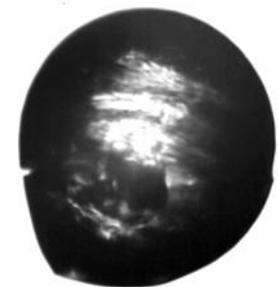
→ Pressure increase in the liquid core leads to melting point depression (via Clapeyron equation), followed by slowing of ice growth rate and therefore by an overall decrease of the droplet temperature.

→ In a pressure release event (fracturing), the pressure returns back to normal and melting point back to 273 K; the rate of latent heat release is again balanced by heat removal rate from the surface, the central temperature returns to 0°C.

→ Pressure measurement is possible by IR thermal imaging!



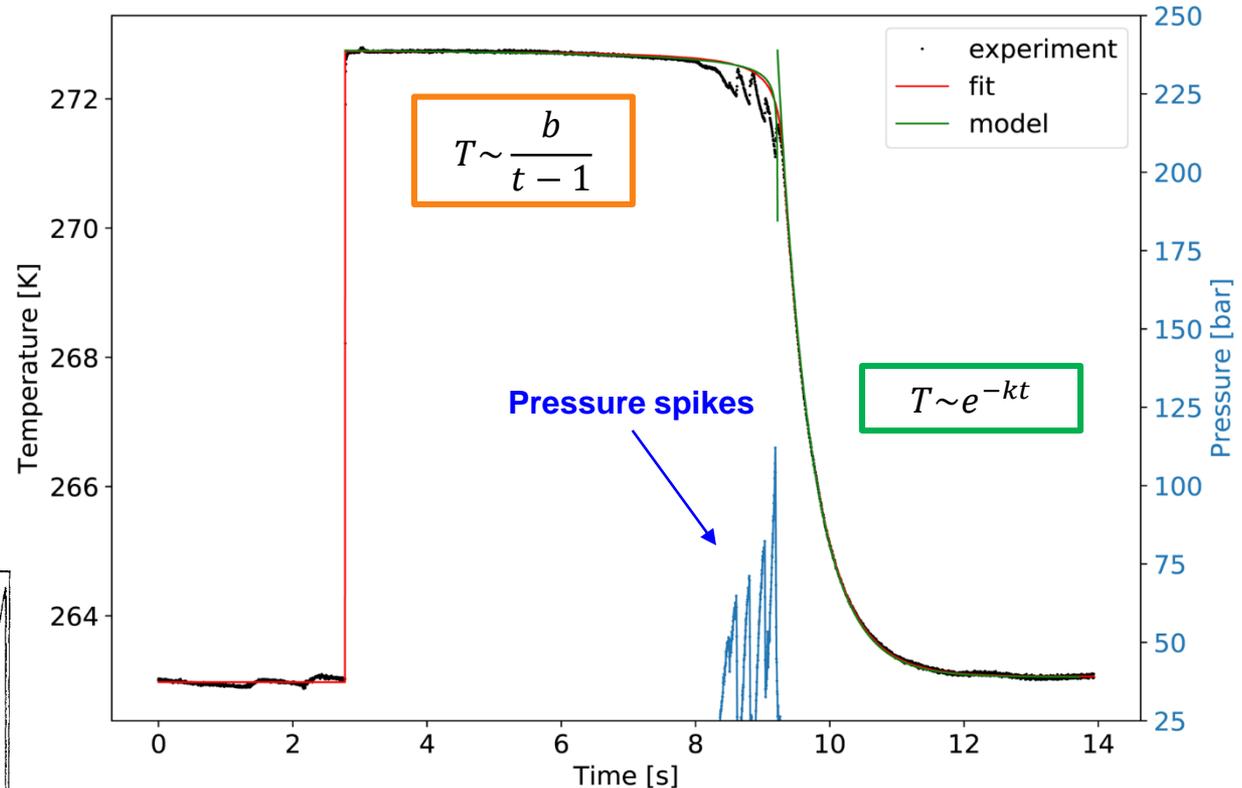
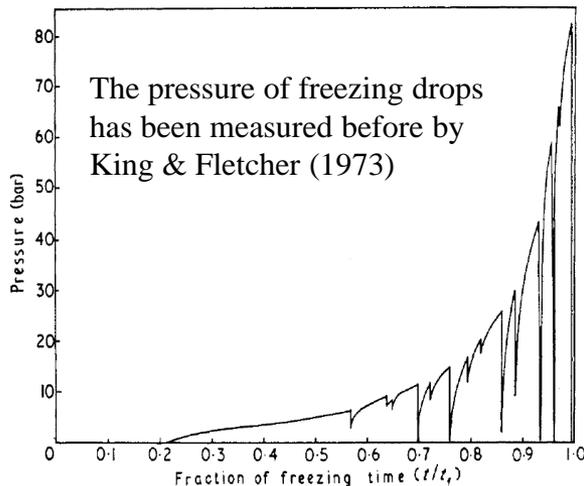
Incomplete break-up: 1



2
 $\Delta t_{1,2} = 0.2 \text{ ms}$

Calculation of internal pressure from spikes in surface temperature

The melting point depression was derived by subtraction of the measured temperature from the **envelope fit curve**, representing a baseline of „freezing without pressure change“. The **shape of the fit** was inspired by the **thermal model prediction** of the surface temperature for the second freezing and cooling stage.

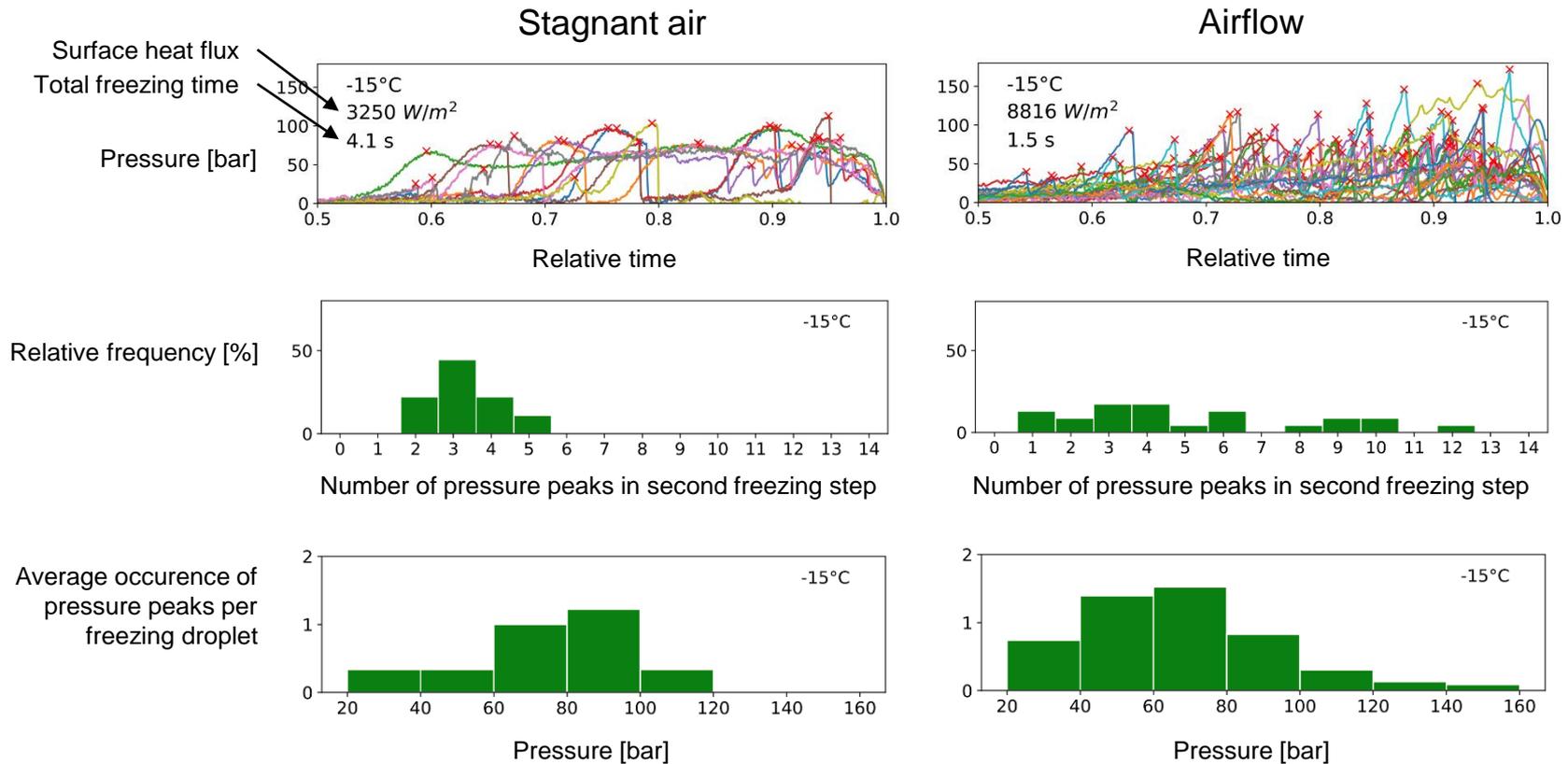


The perfect match between the envelope fit and model prediction allows using the fit curve as a baseline for pressure calculation:

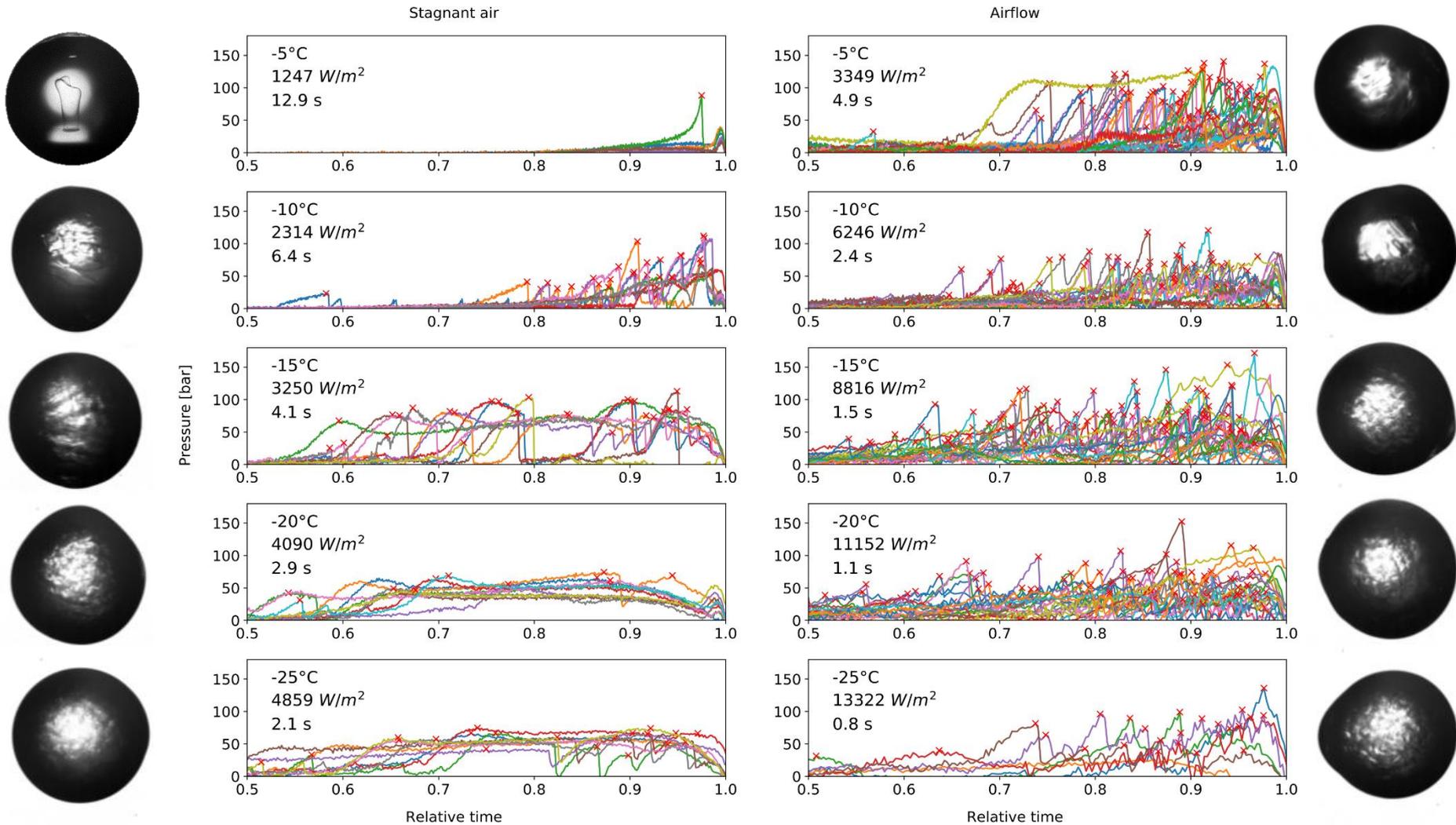
$$\text{Pressure} = (T_{\text{experiment}} - T_{\text{fit}}) * 135 \text{ bar/K}$$

Evolution of internal pressure: stagnant air vs. airflow

- Many pressure release events (spikes) could be detected per droplet, even if nothing is visible in the movies recorded by HSV camera.
- More pressure spikes and higher pressures achieved in airflow, apparently due to faster freezing

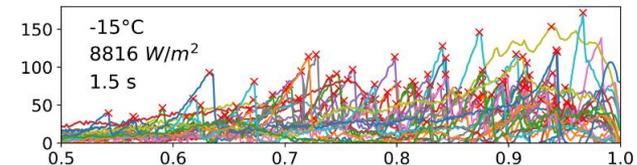
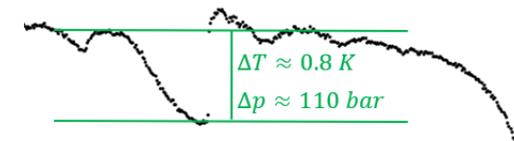
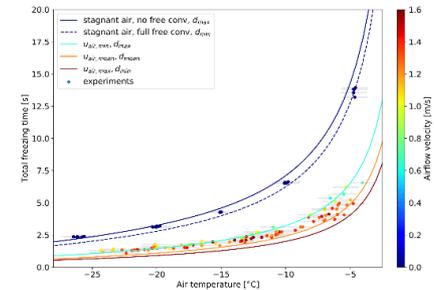
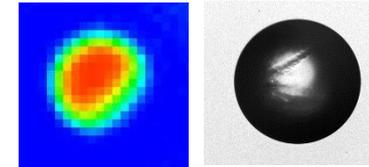


Overview of the results



Summary

- IR measurement of surface temperature with synchronized HSV recordings gave new insight into the freezing of free-falling water droplets.
- Our thermal model of droplet freezing was in good agreement with experiments with respect to the total freezing time and surface temperature.
- The internal pressure could be derived from the surface temperature measurements via melting point depression.
- Droplets freezing in free fall develop higher pressure and show higher frequency of pressure release events. Apparent role of faster freezing is suggested.



More pressure release events detectable with IR thermal imaging compared to HSV: Potentially higher SIP rate than previously thought

Acknowledgements

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