

# Floating on air



Physics of Fluids  
University of Twente

**Detlef Lohse**  
**Physics of Fluids**  
**University of Twente**

# Coworkers

Bram Borkent

Wilco Bouwhuis

Nicolas Bremond

Henri Lhuissier

Hanneke Gelderblom

Stefan Gekle

Alvaro Marin

Devaraj van der Meer

Ivo Peters

Andrea Prosperetti

David Rivas

Jacco Snoeijer

Erikjan Staat

Chao Sun

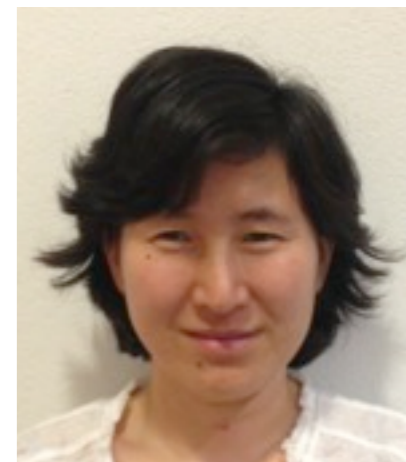
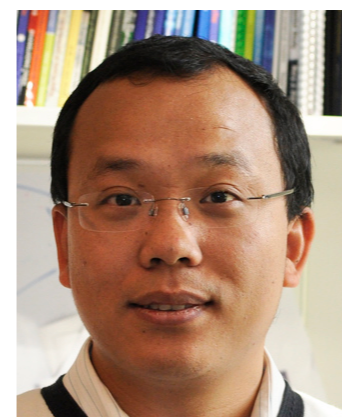
Peichun Amy Tsai

Tuan Tran

Roeland van der Veen

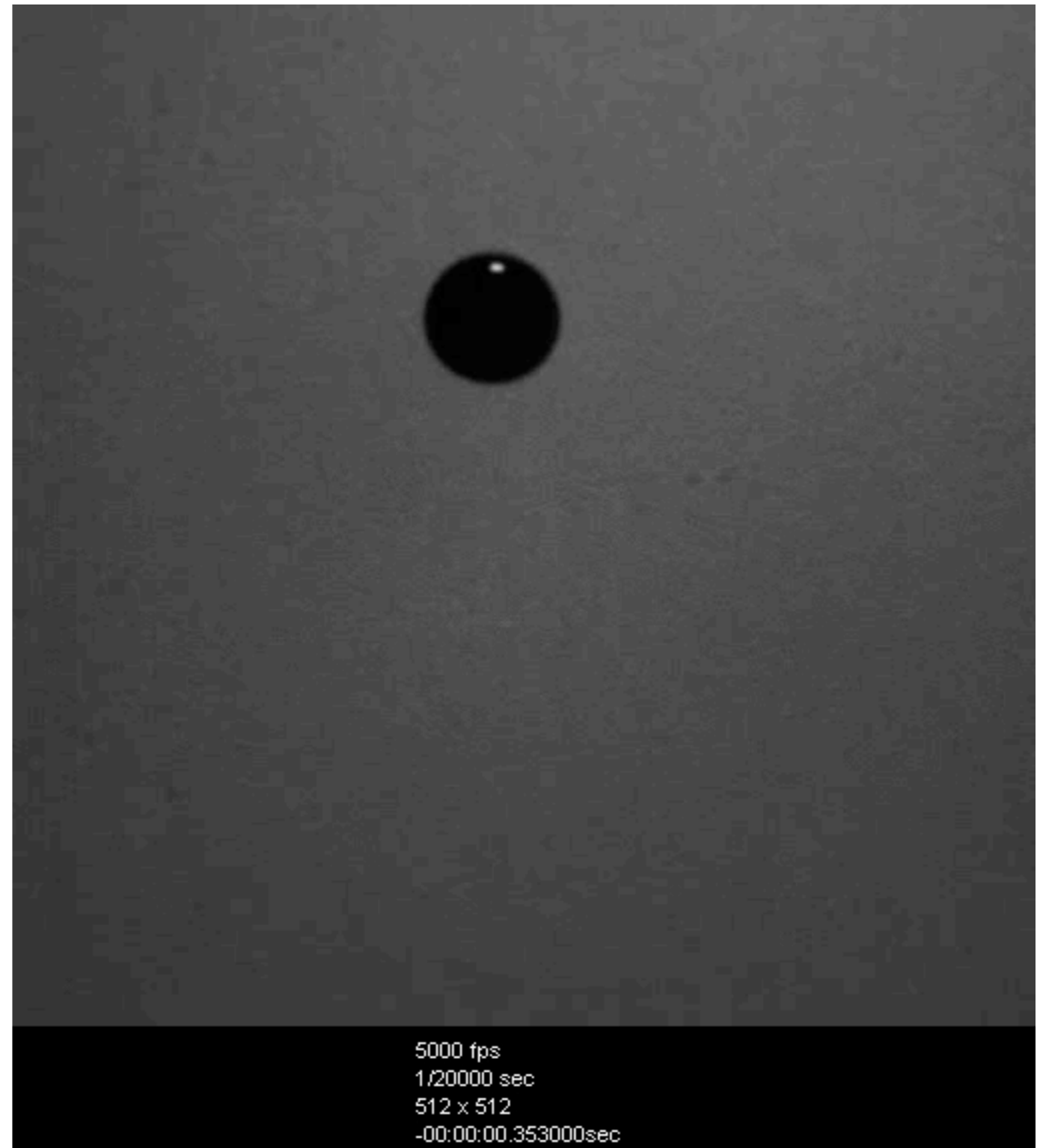
Koen Winkels

Xuehua Zhang



# Droplet impact

... on paper

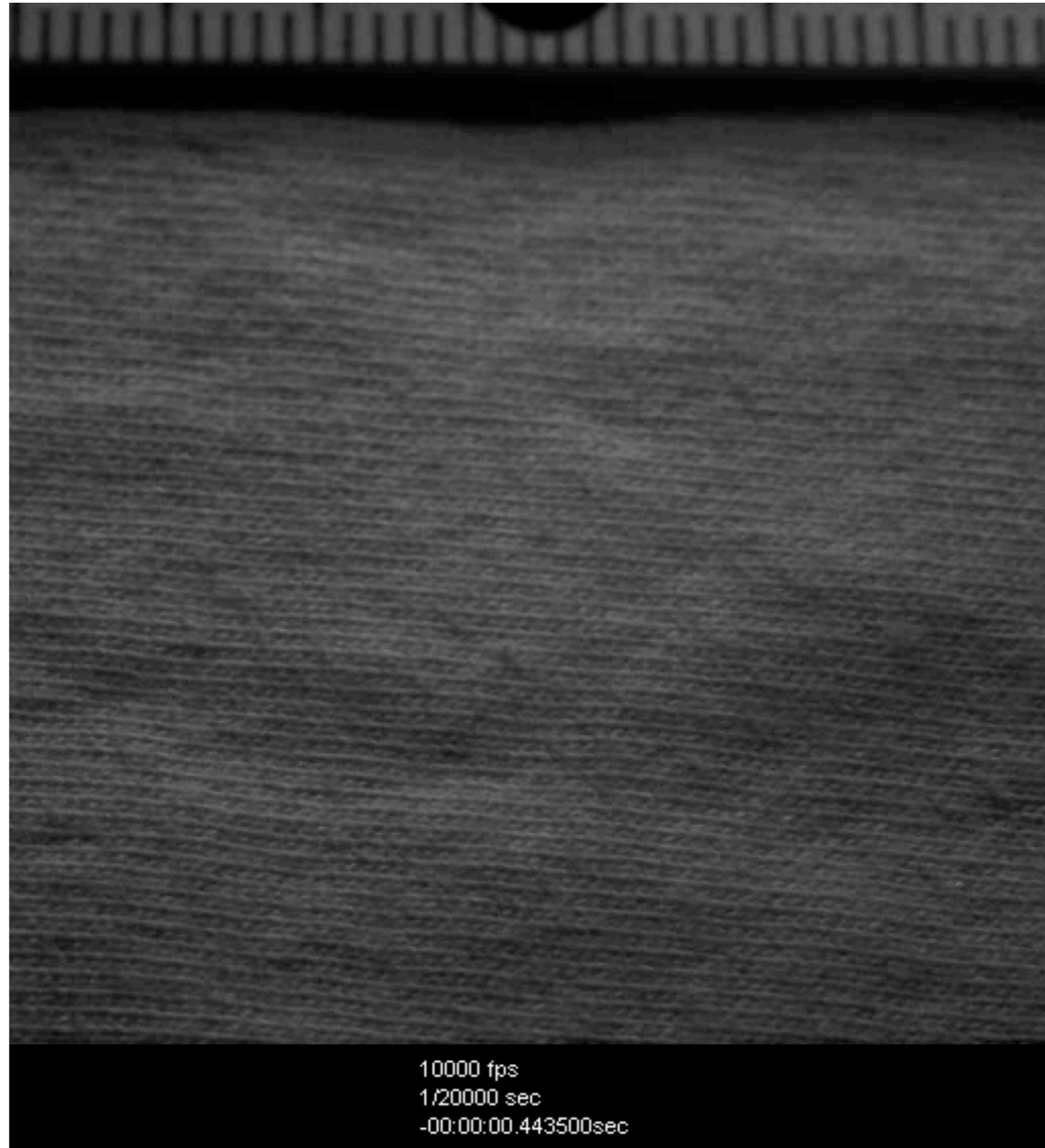


# Strong dependence on type of surface

... on cotton

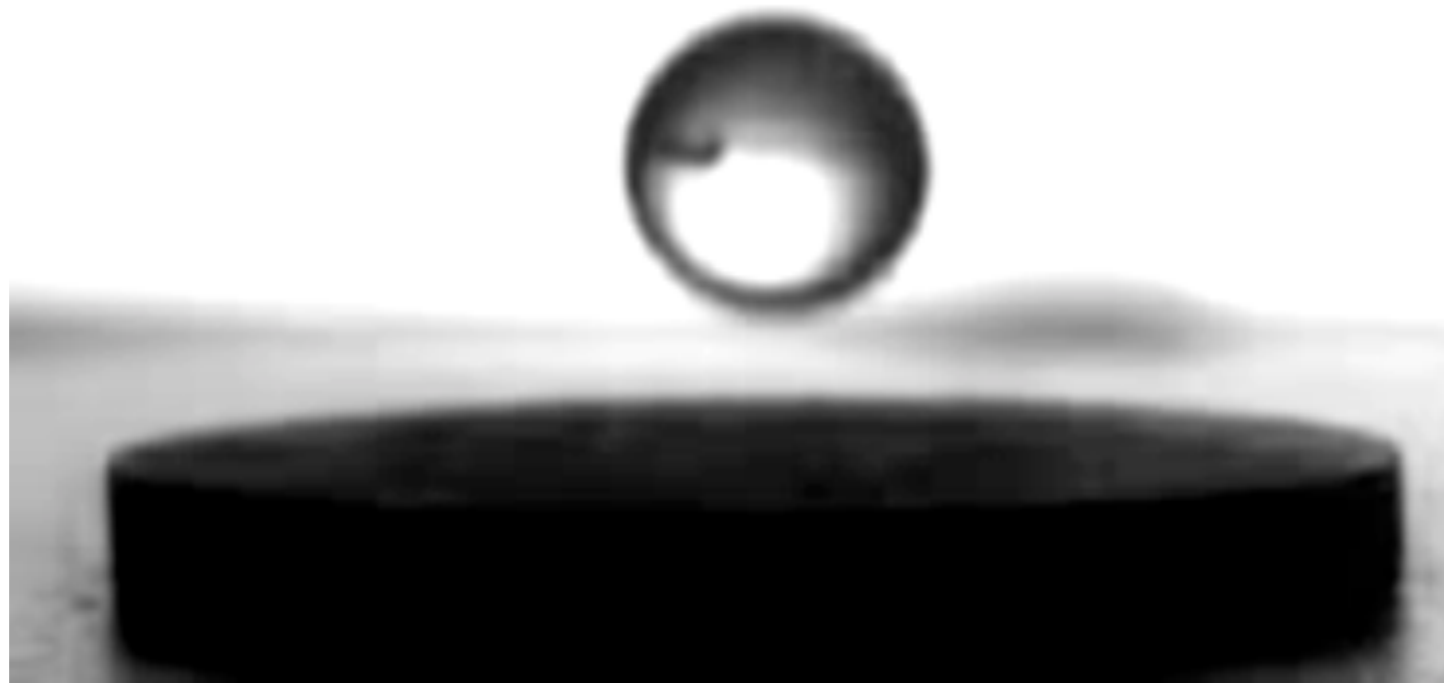
Liquid: blood

Nederlands Forensisch Institute



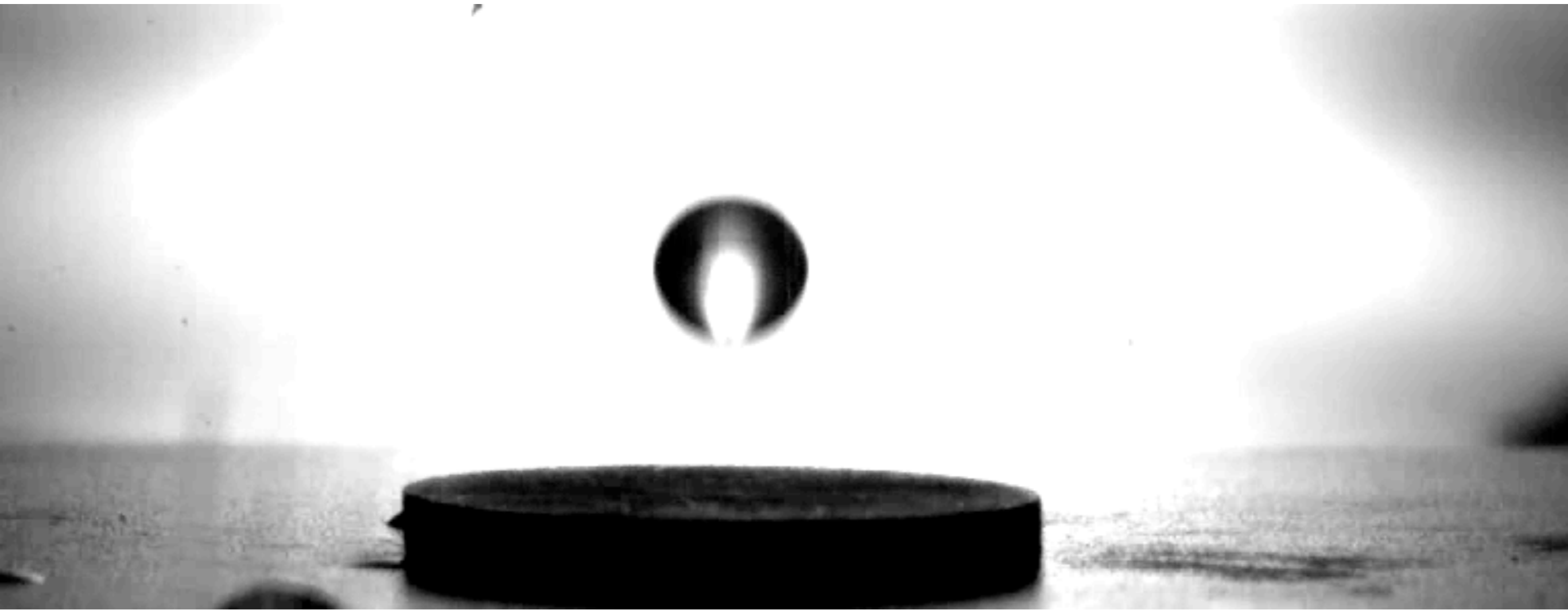


# Impact on hydrophobic surface



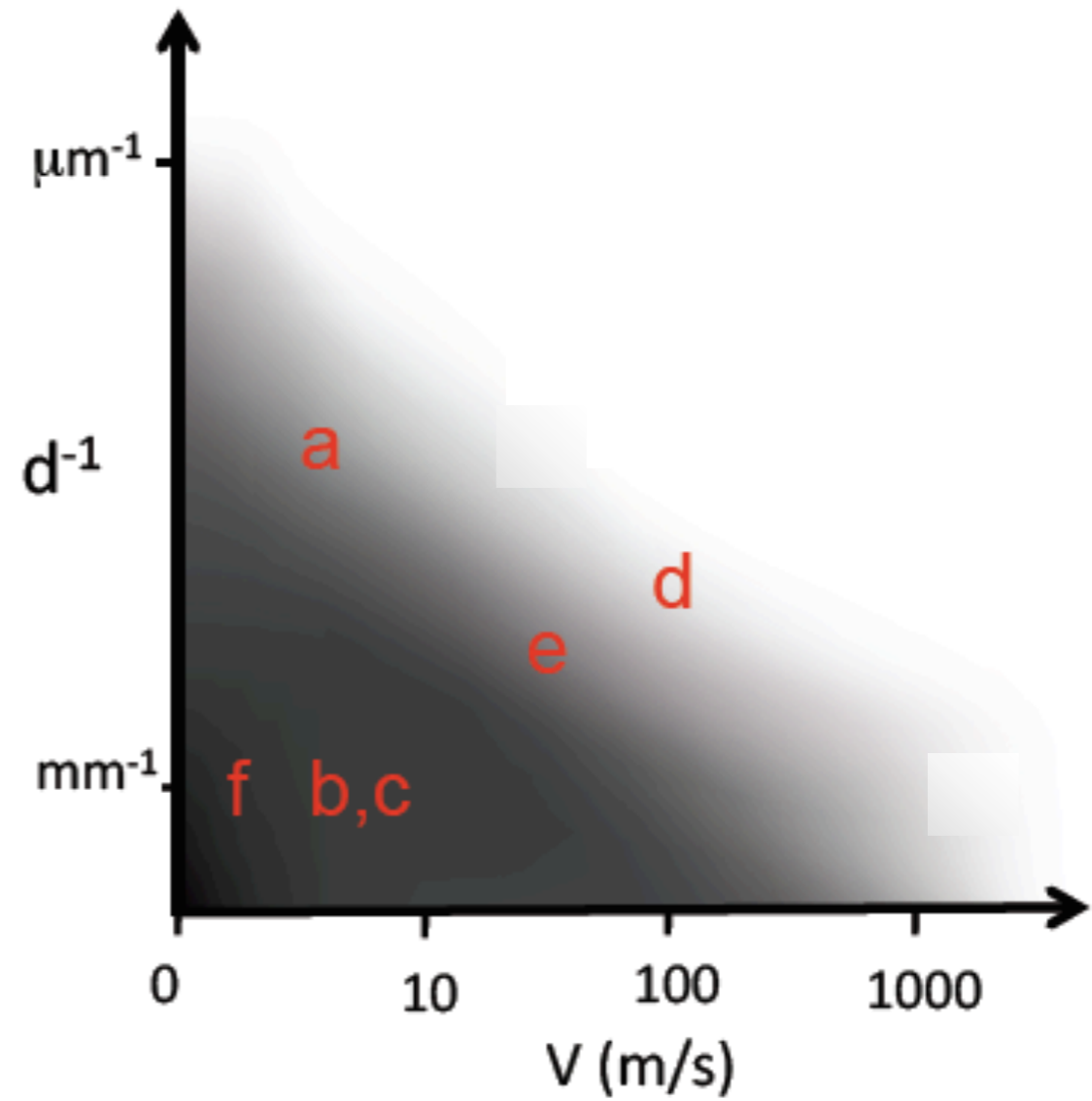
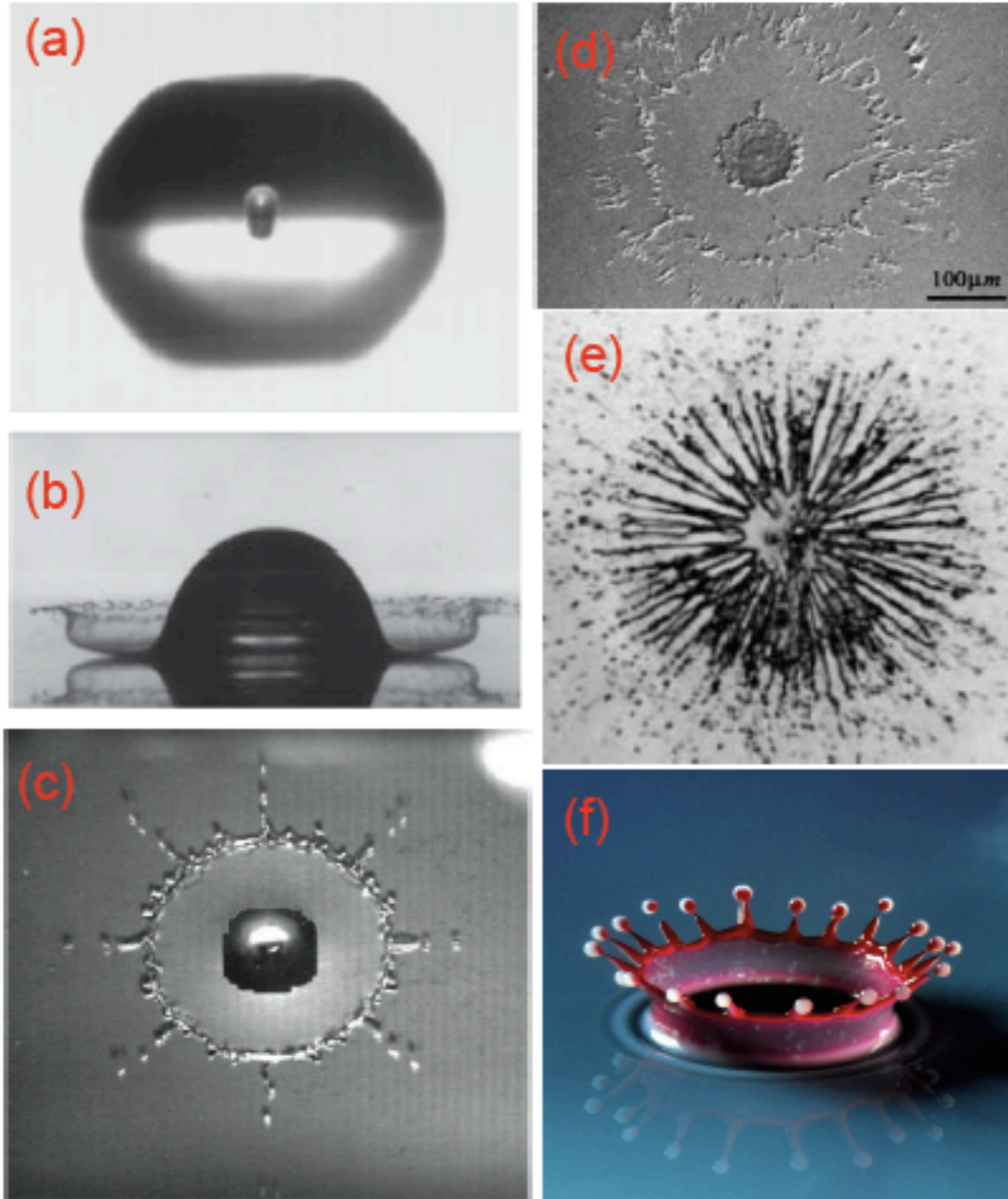
low  
velocity

# Impact on hydrophobic surface



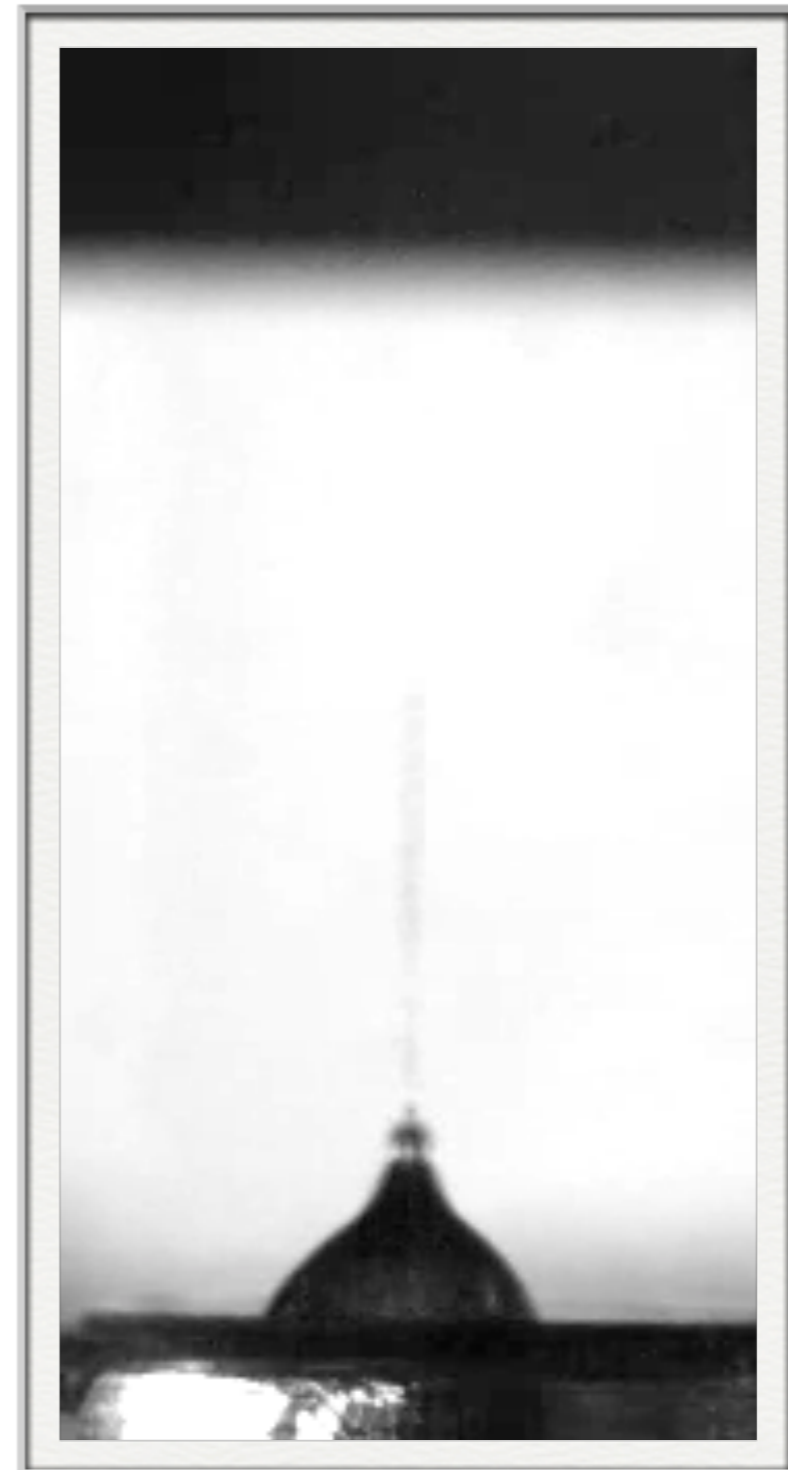
high velocity

# Phase space of impact



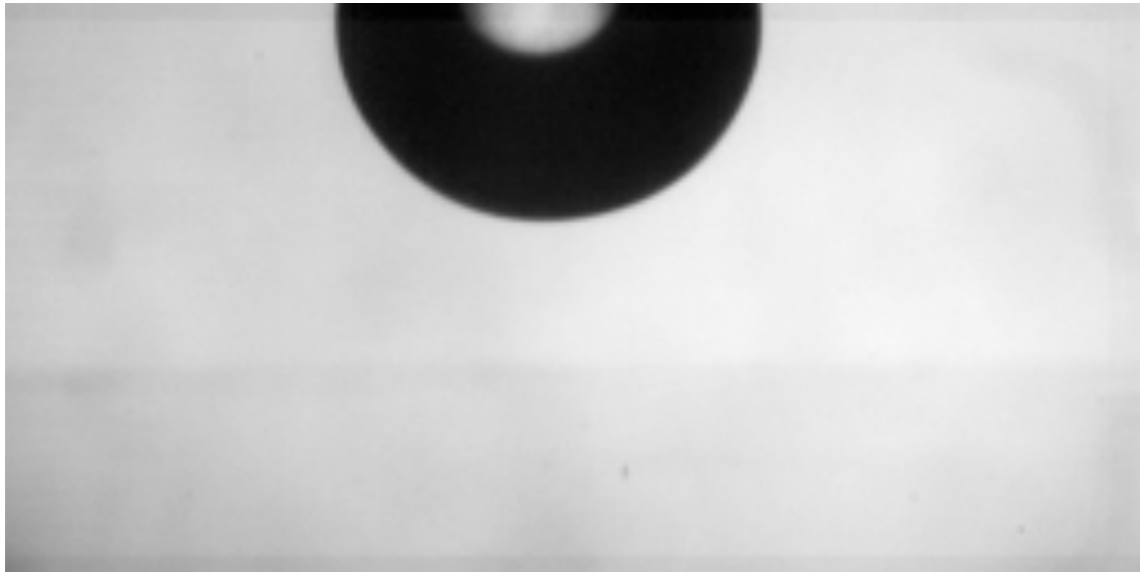
# Phase space of impact

- Velocity
- Diameter
- Viscosity
- Surface tension
- Roughness of surface
- Temperatures
- Air pressure
- ...

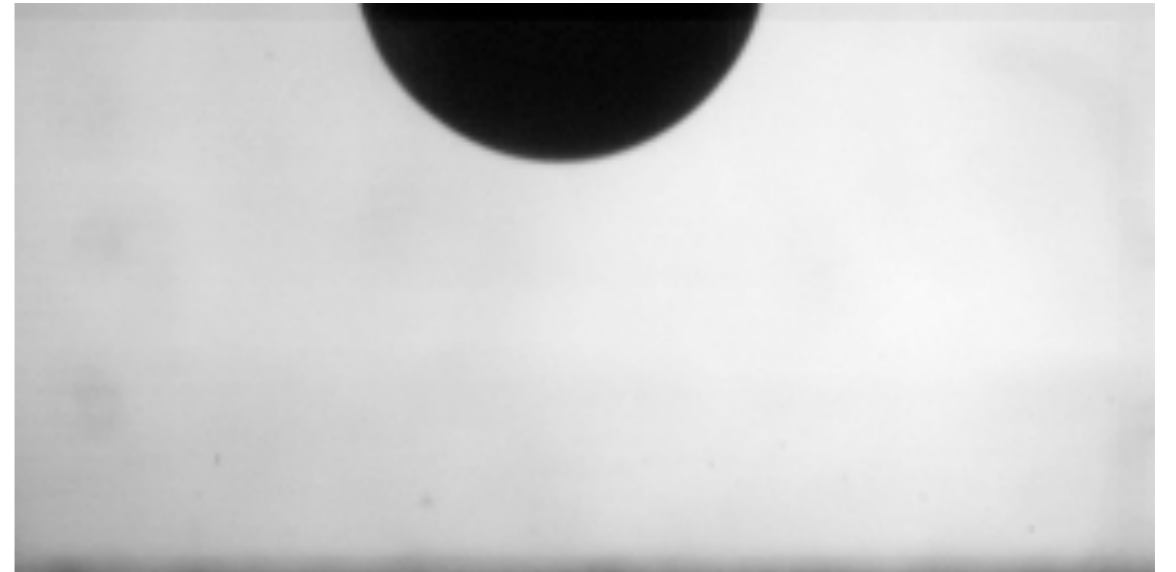


# Influence of air pressure on impact event

1 atm



0.2 atm

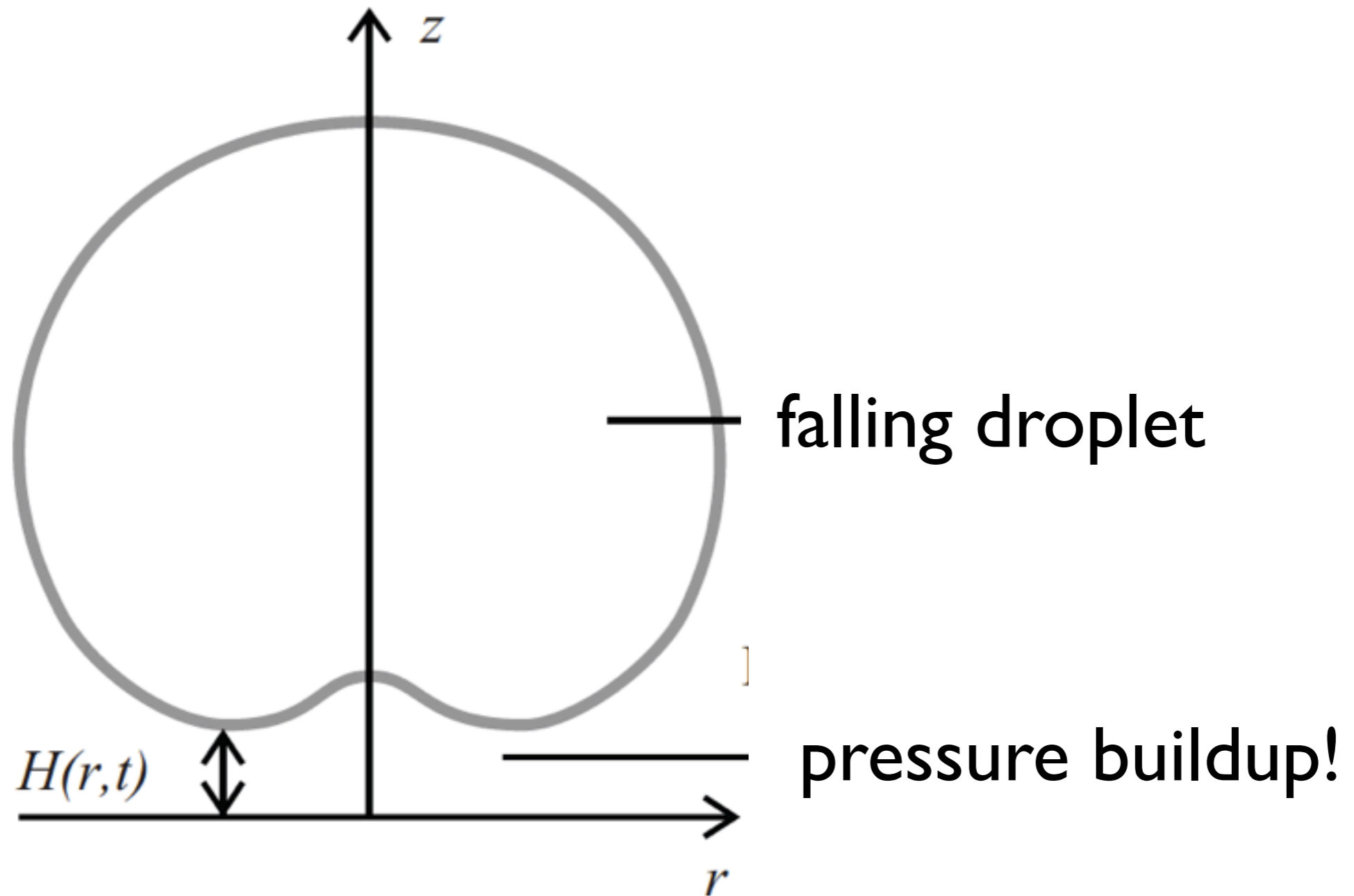


**no splash!**



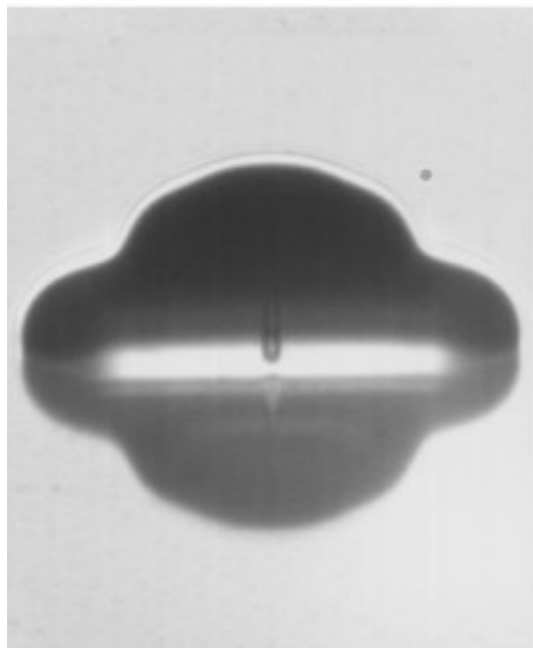
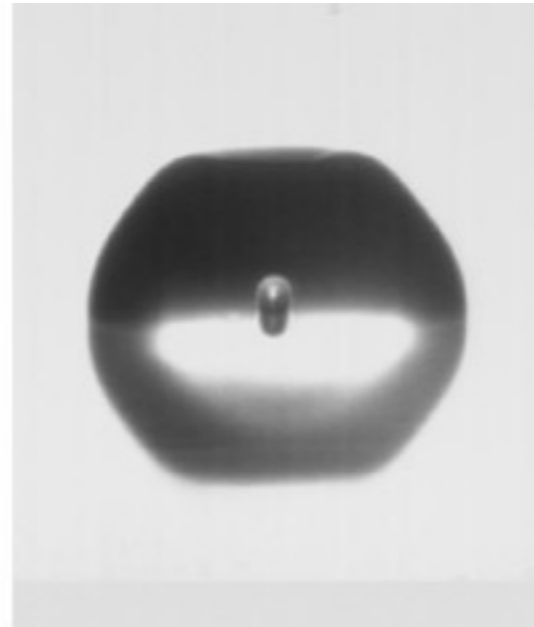
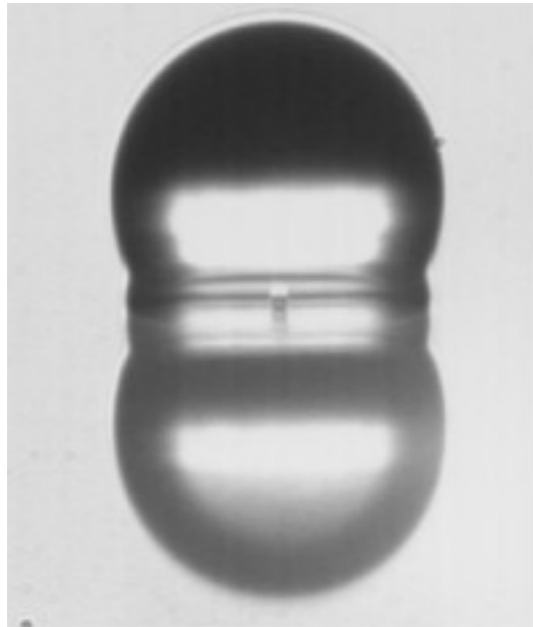
**How come?**

# Sketch of impacting drop

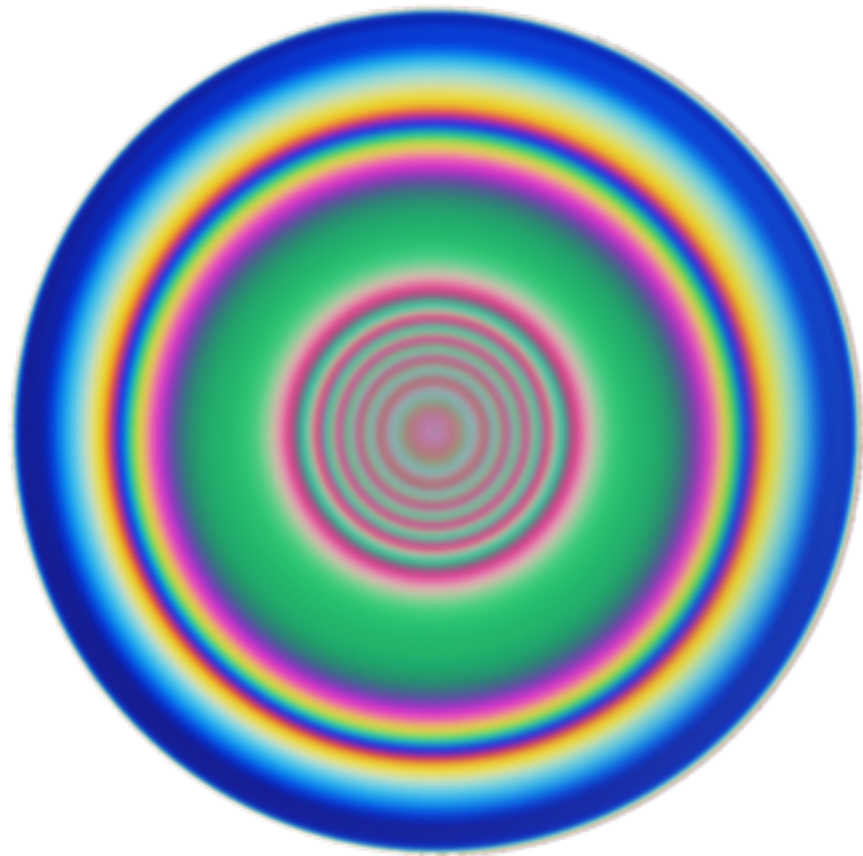


**Dimple-formation!**

# Consequence: entrained bubble under droplet!



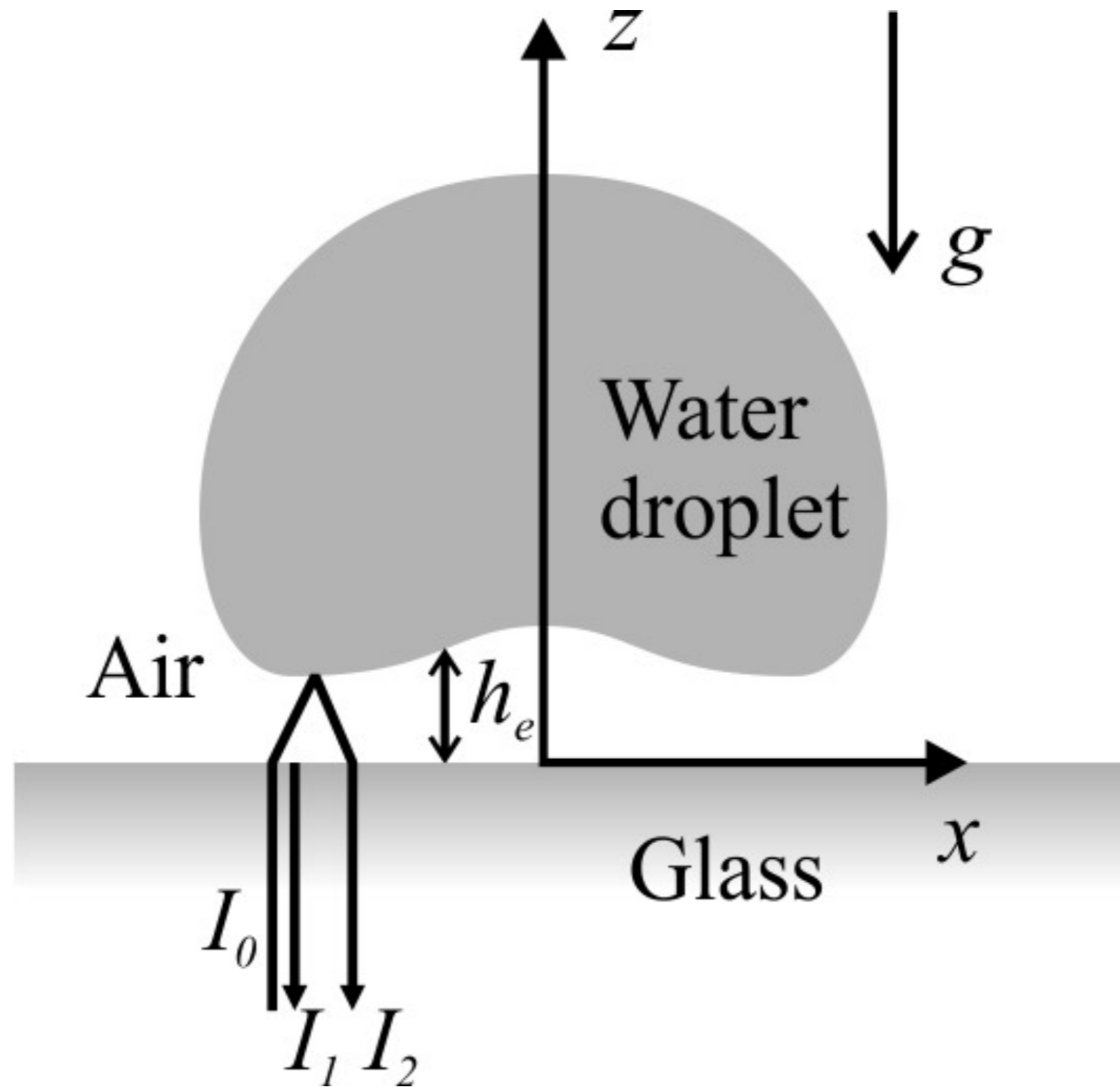
# Ultra-high-speed color interferometry



**Direct measurements  
of air layer profiles  
under impacting  
droplets**

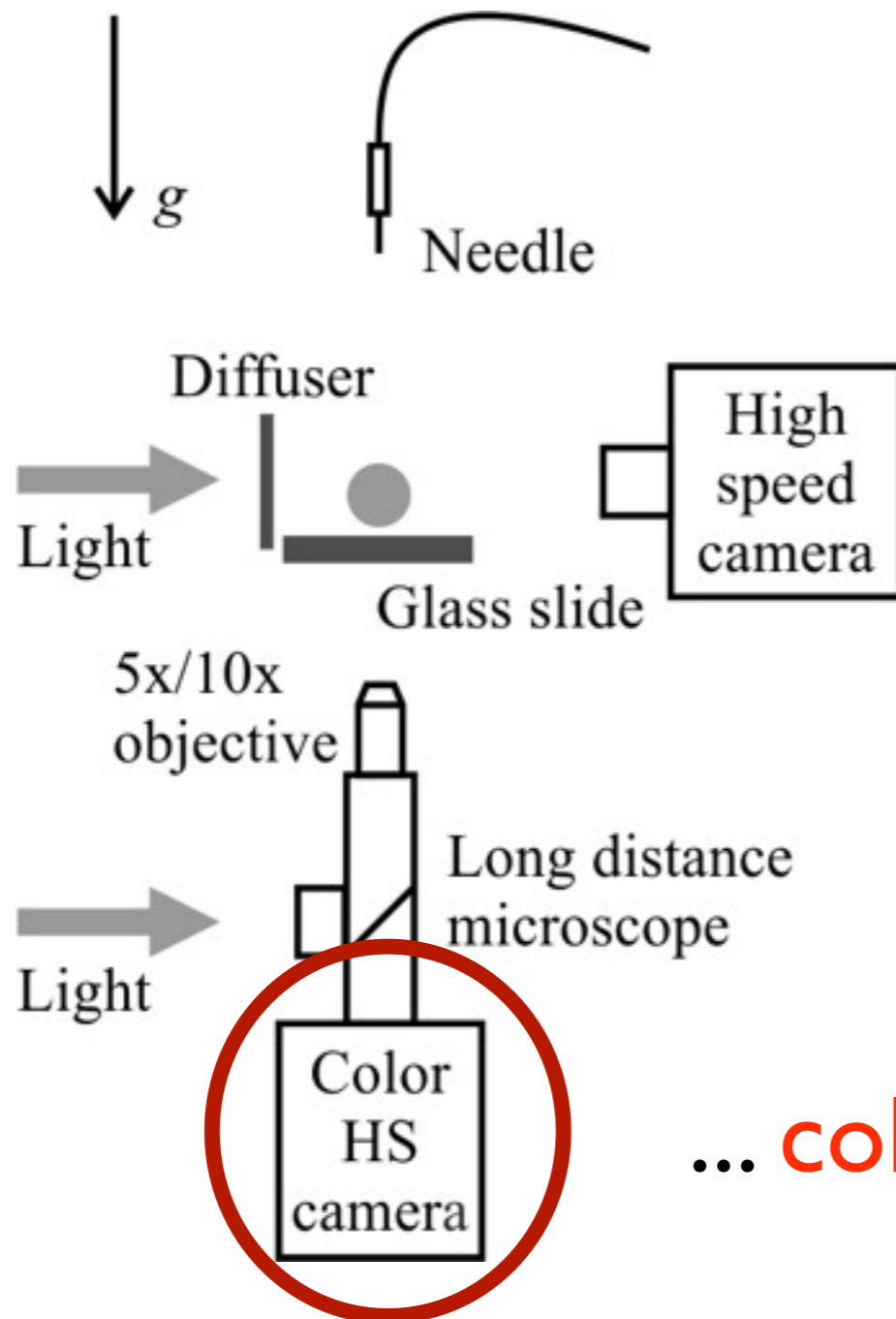
van der Veen, Tran, Lohse, Sun, Phys. Rev. E 85, 026315 (2012);  
Bouwhuis et al., Phys. Rev. Lett. 109, 264501 (2012)

# Interferometry for falling droplets



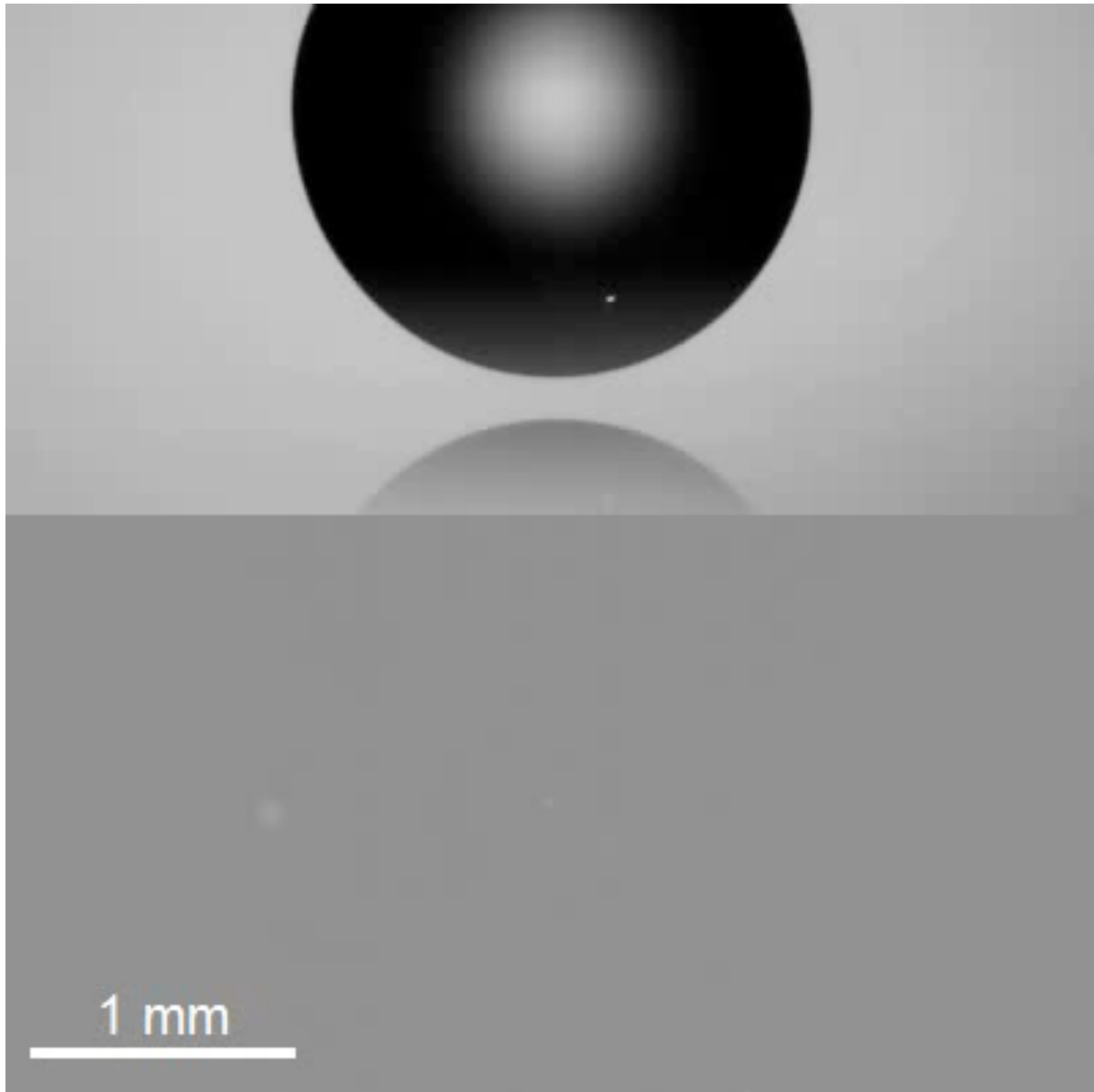


# Experimental setup

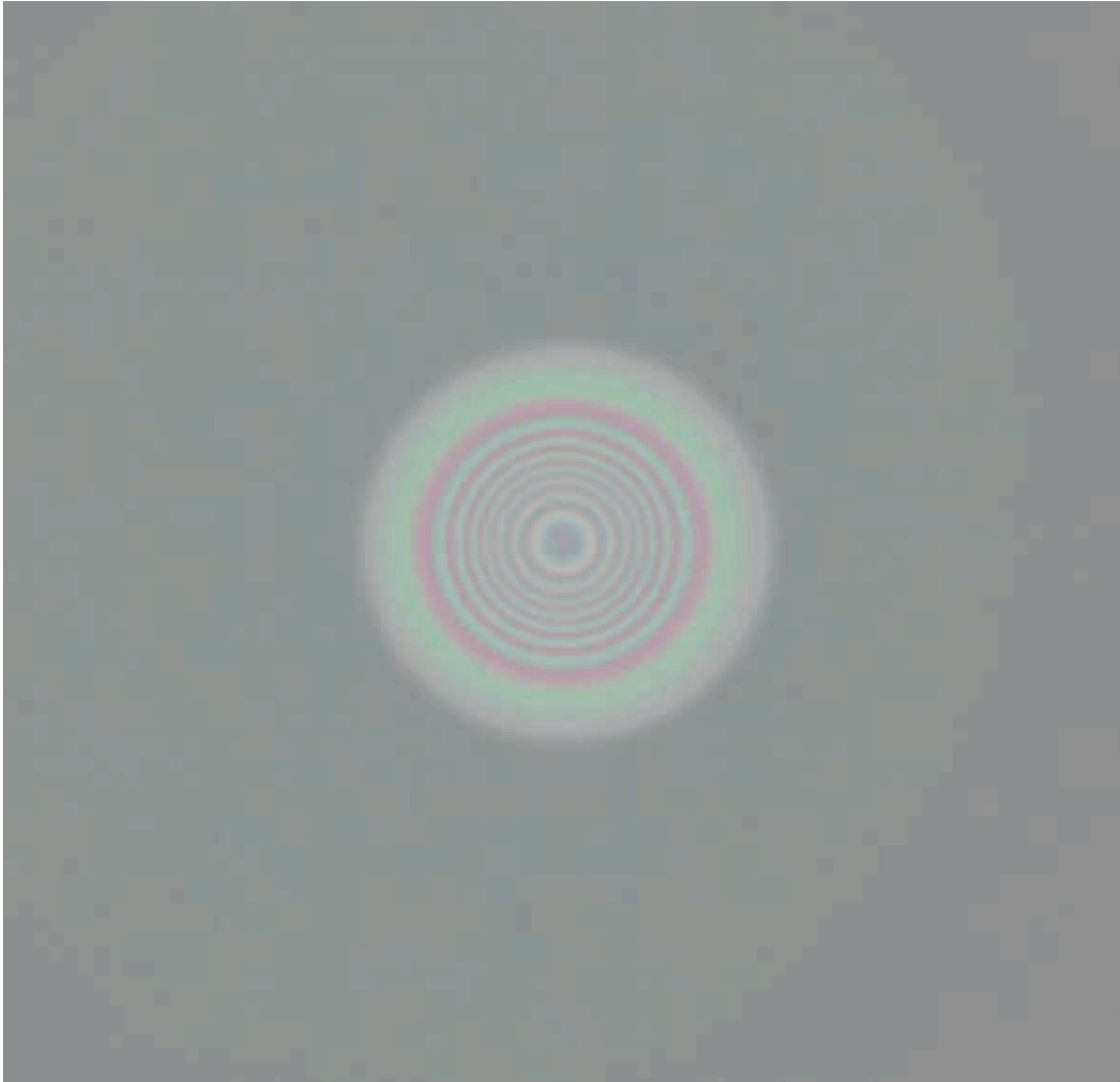


side view and...

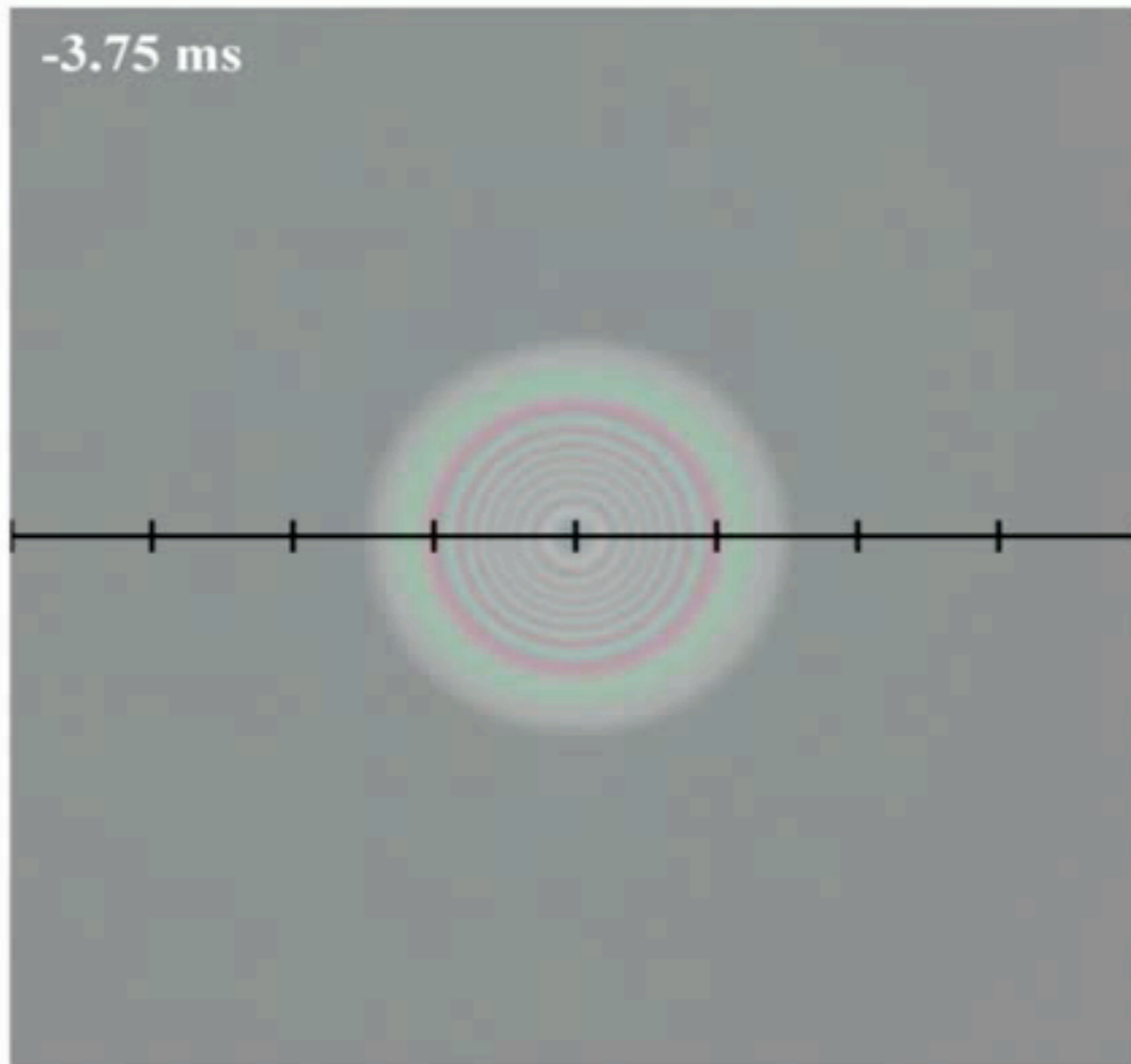
... colored bottom view



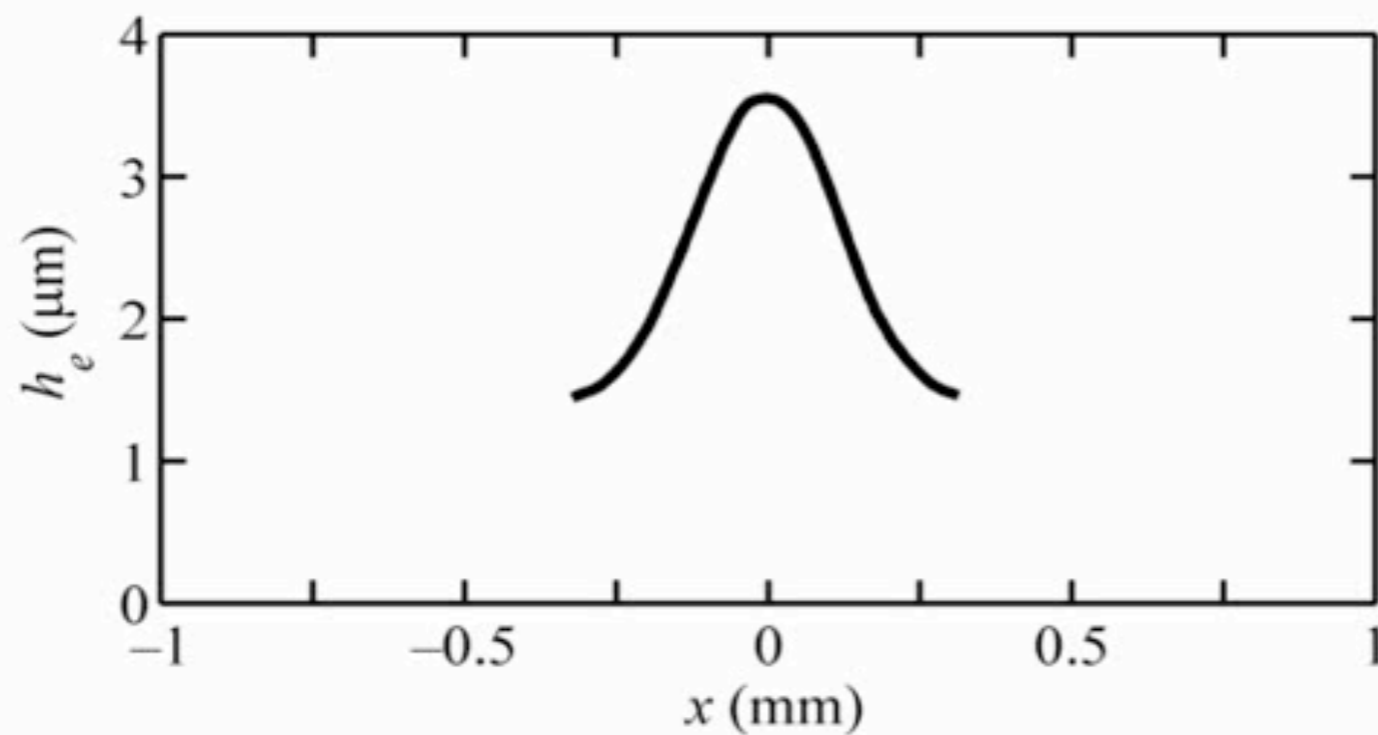
Water  
 $D = 2.0 \text{ mm}$   
 $U = 0.19 \text{ m/s}$   
 $We = 1.0$   
  
20000 fps



Water  
 $D = 2.0 \text{ mm}$   
 $V = 0.22 \text{ m/s}$   
 $We = 1.3$   
  
4800 fps

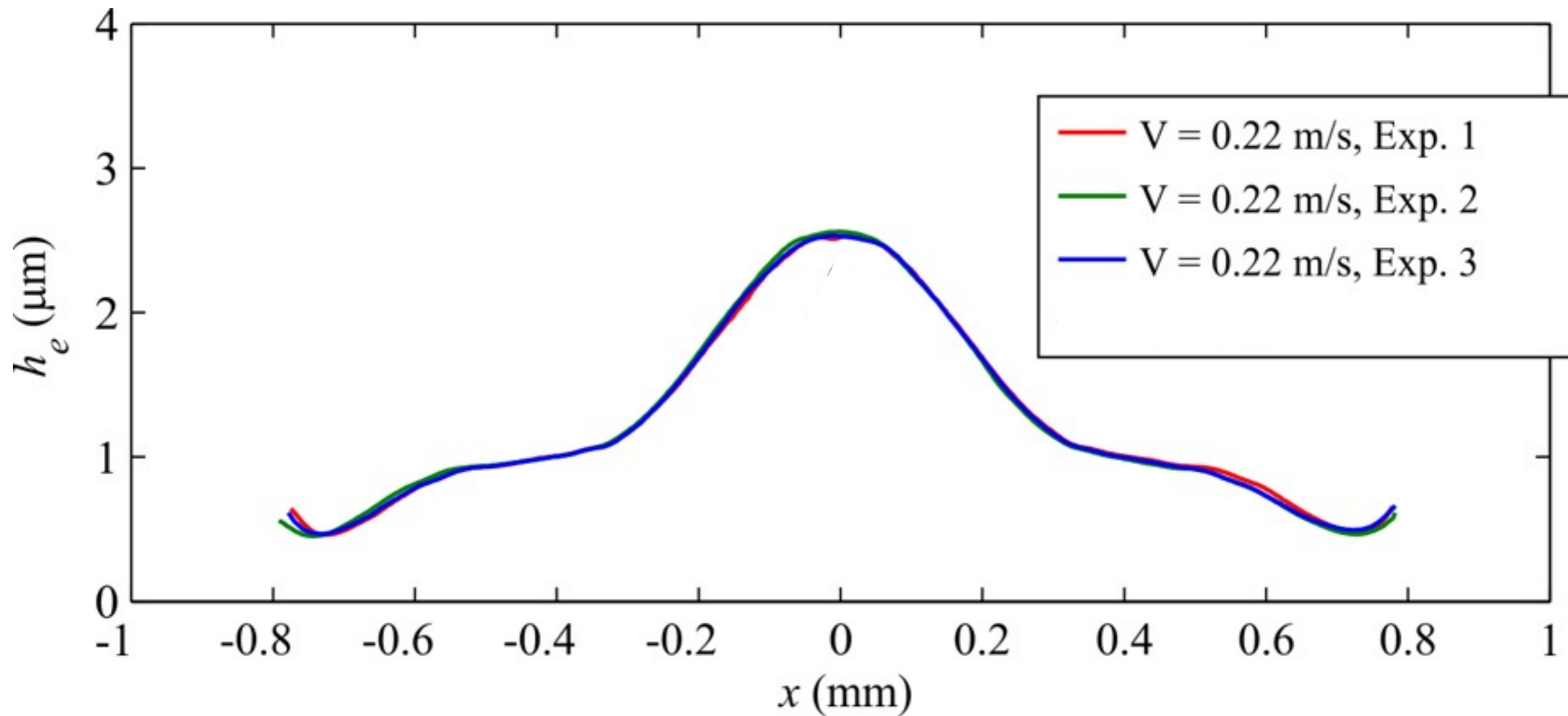


Water  
 $D = 2.0$  mm  
 $V = 0.22$  m/s  
 $We = 1.3$



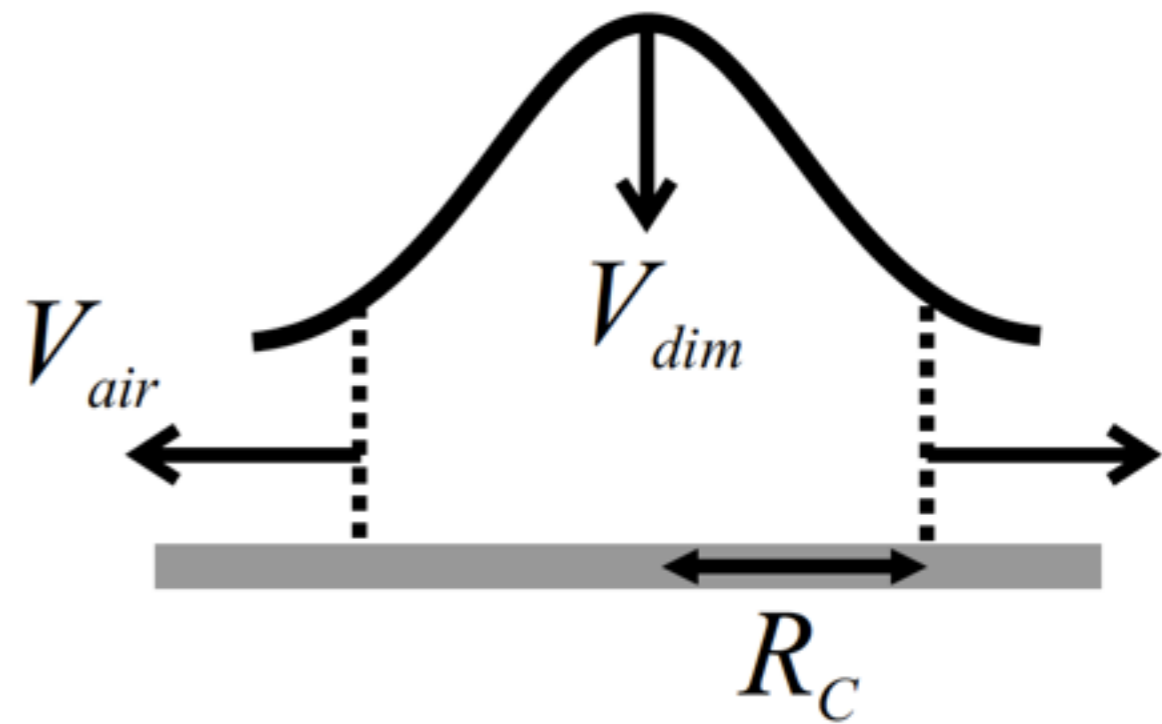
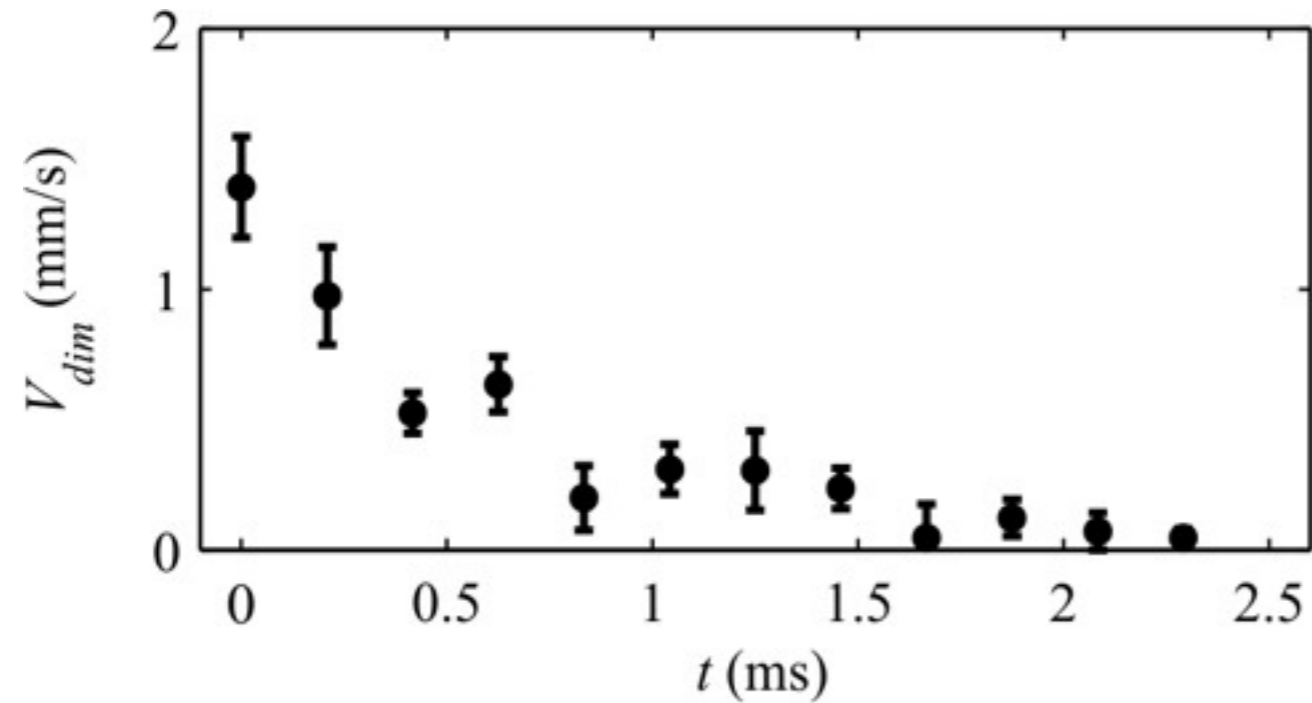
4800 fps

# Reproducibility perfect!





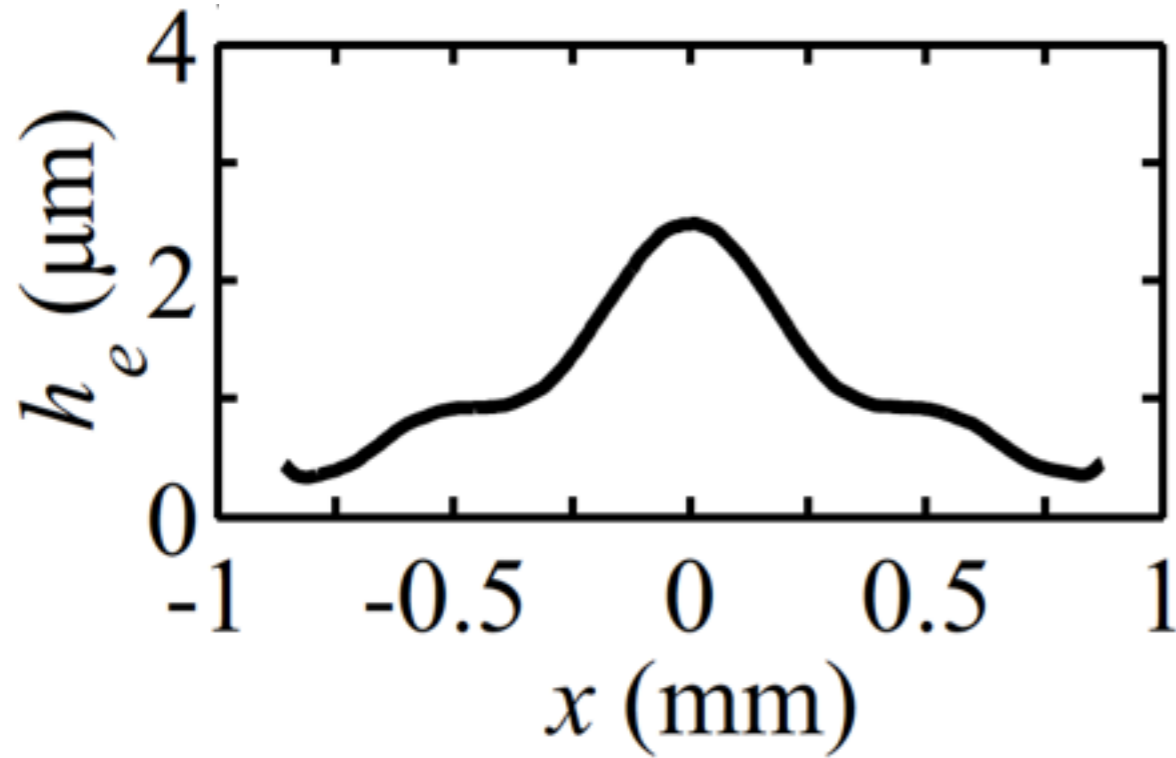
# Profile dynamics & dimple velocity allows to calculate velocity of outwards air flow



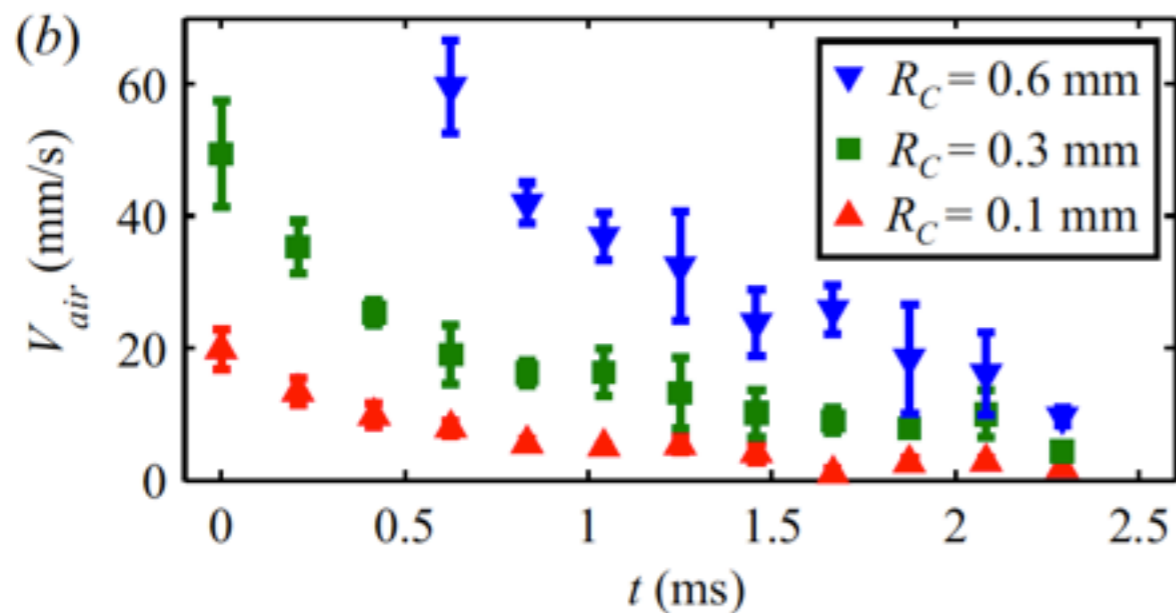
assume  
incompressibility

60 x higher air  
flow velocities!

# Note the numbers...



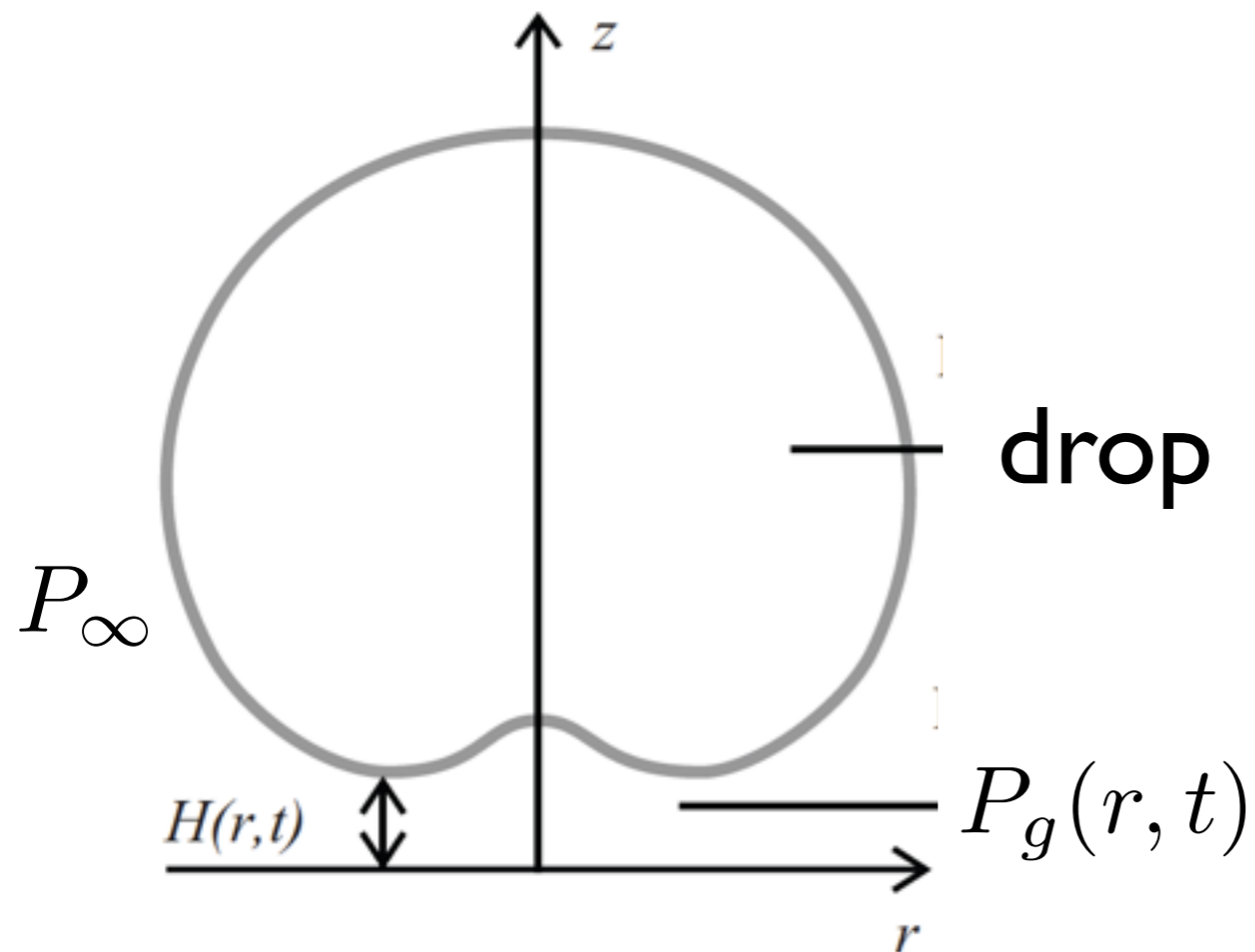
$$Re \sim \frac{V_{air} h_e}{\nu_{air}}$$
$$\sim 0.01$$



**Air-flow under  
the drop is  
viscous flow!**

# **Numerics & theory**

# Compare to BI calculations



Laplace equation:

$$\nabla^2 \phi = 0$$

Solve for

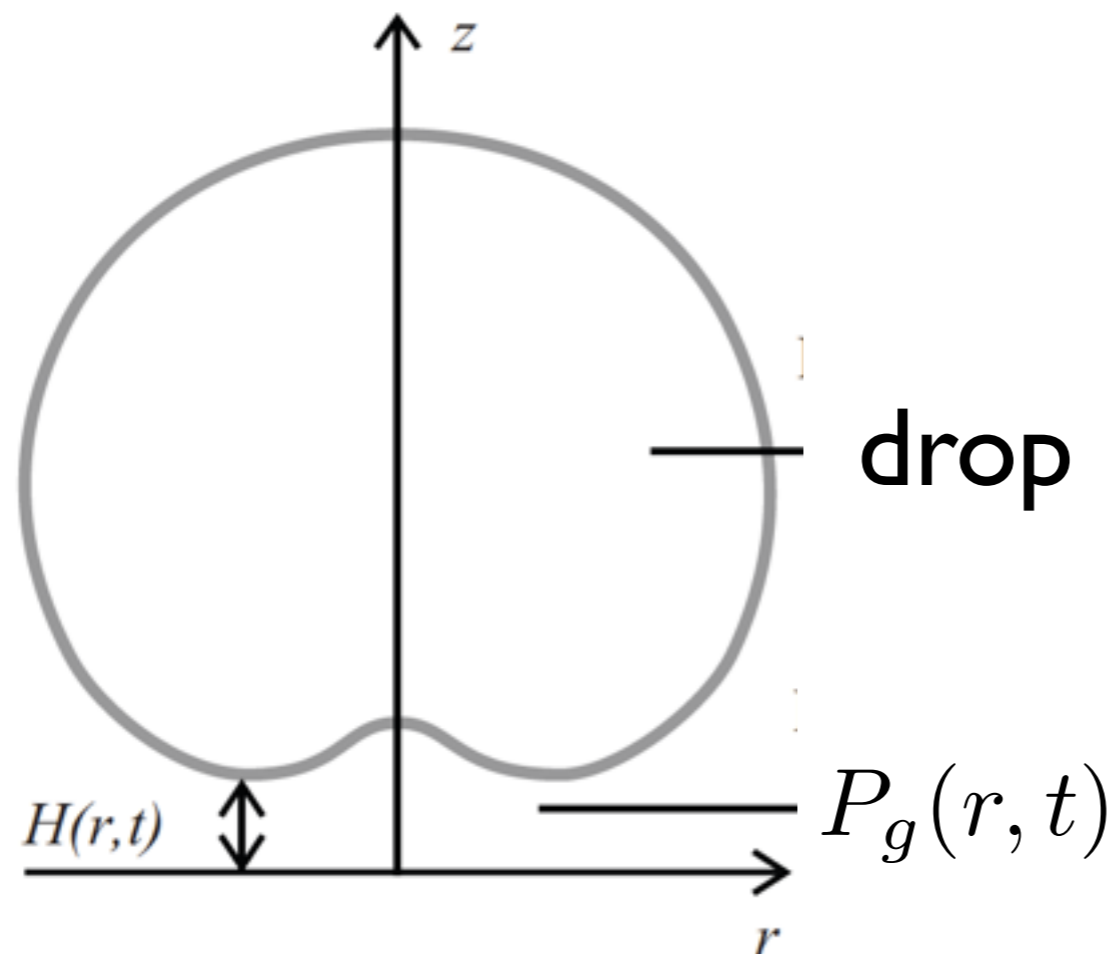
$$H(r, t) \text{ \& } P_g(r, t)$$

Unsteady Bernoulli equation for liquid:

$$\partial_t \phi + 1/2 |\nabla \phi|^2 = -gz - \frac{\gamma}{\rho l} \kappa(H(r, t)) - \frac{1}{\rho l} (P_g(r, t) - P_\infty)$$

# Obtain pressure from Stokes equation for gas (viscous!) in lubrication approximation

$$\partial_t H(r, t) - \frac{1}{r} \frac{\partial}{\partial r} \left( \frac{r(H(r, t))^3}{12\eta_g} \partial_r P_g(r, t) \right) = 0$$



Solve the two coupled PDEs for  $H(r, t)$  &  $P_g(r, t)$



# Dimensionless numbers

Weber number:

$$We = \frac{\rho_l R U^2}{\gamma}$$

Bond number:

$$Bo = \frac{\rho_l g R^2}{\gamma}$$

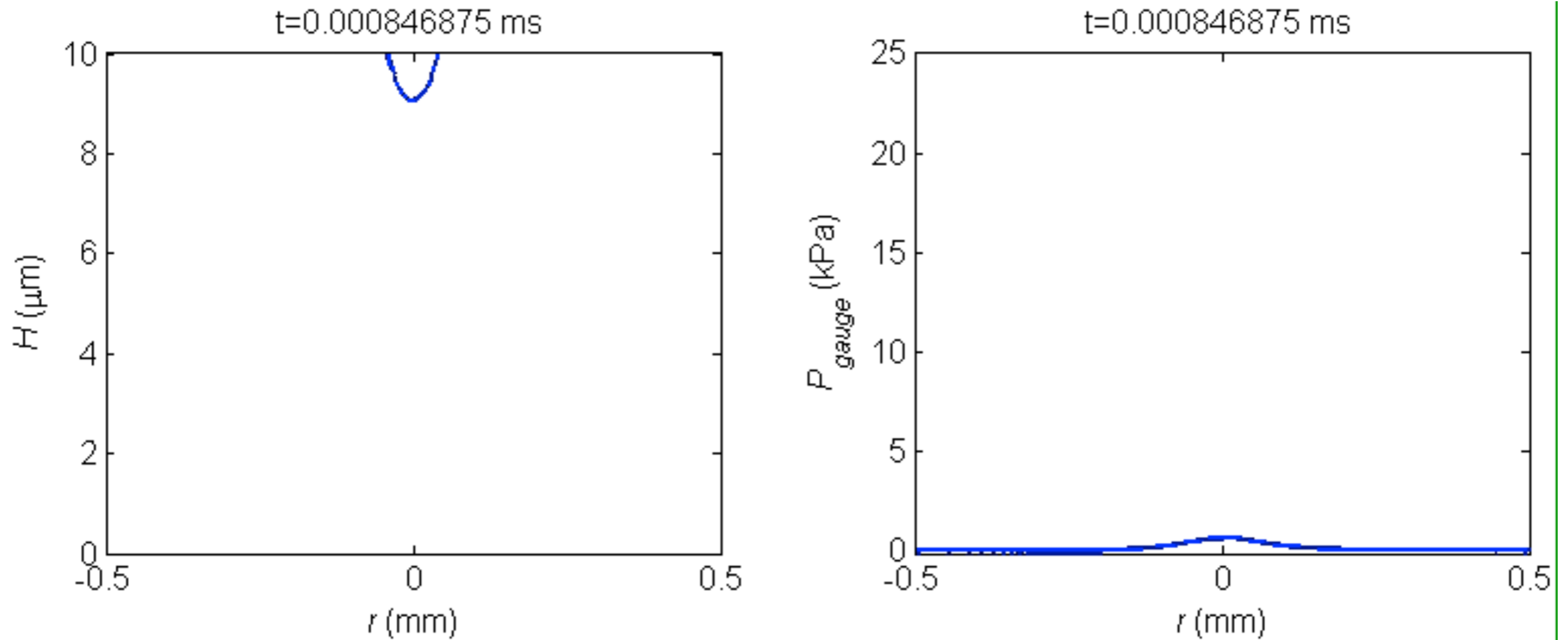
Gas flow relevant:  
(inverse) Stokes number:

$$St = \frac{\eta_g}{\rho_l U R}$$

Capillary number:

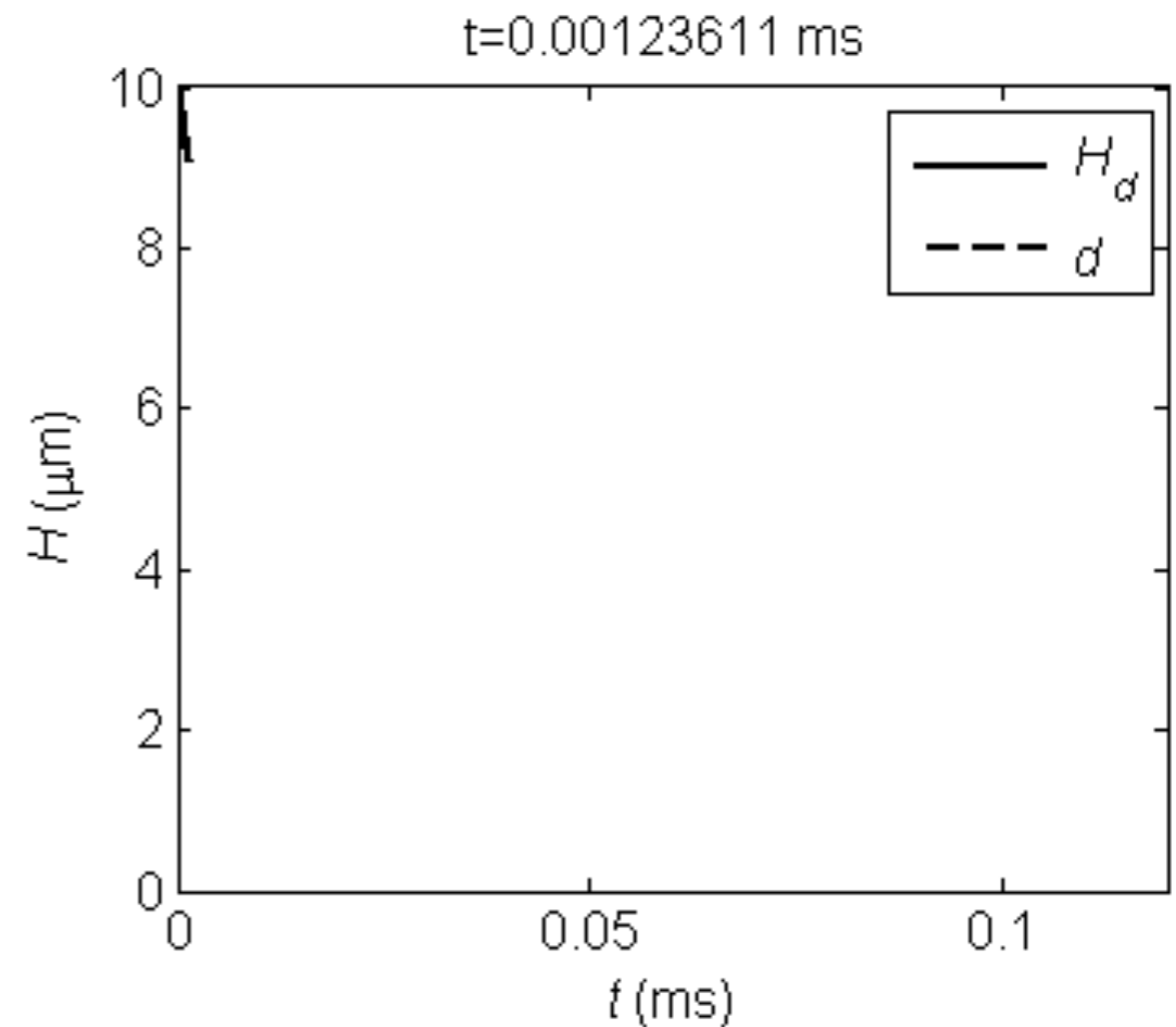
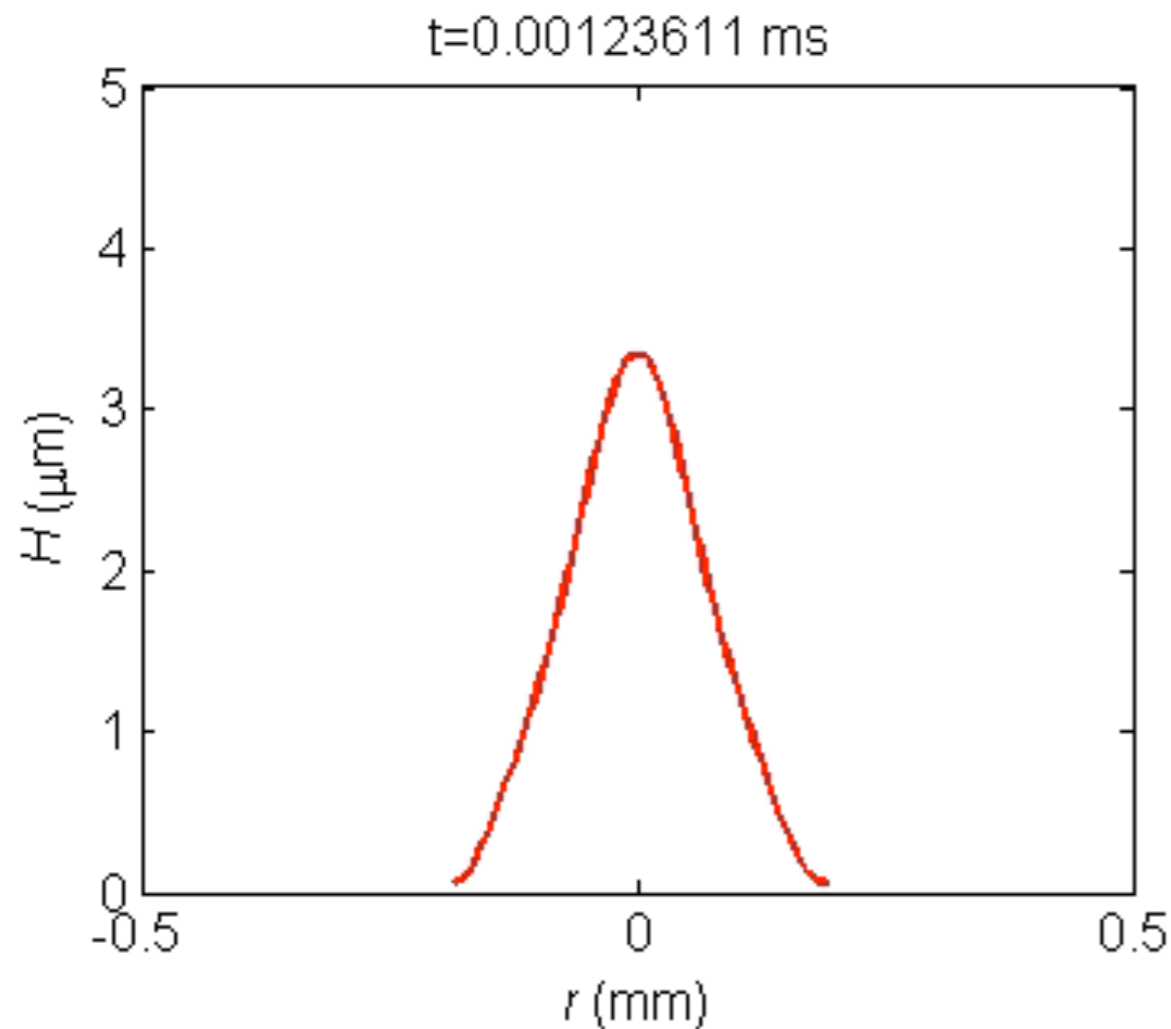
$$Ca = \frac{\eta_g U}{\gamma} = St \cdot We$$

# Profile & pressure



ethanol,  $U=1.12\text{m/s}$ ,  $D=1.8\text{mm}$ ,  $We=40$

# Profile dynamics in inertial regime: comparison with **experiment**



$$U=1.12\text{m/s}, \quad We=40$$

no free parameter!

# **Under what conditions is the entrained bubble maximal?**

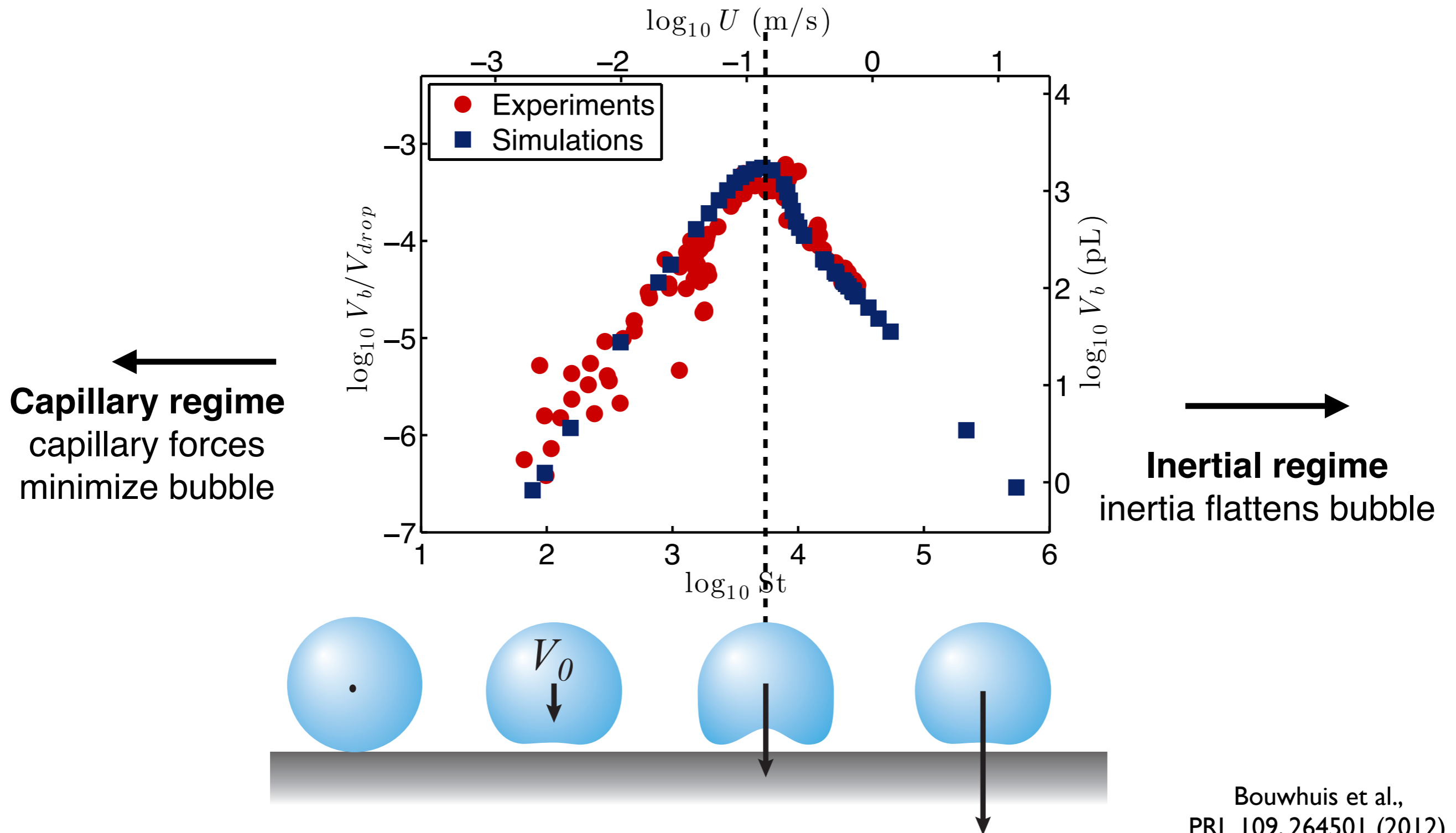
large  $U$  (and/or larger  $R$ ):

inertia of impact makes smaller dimples

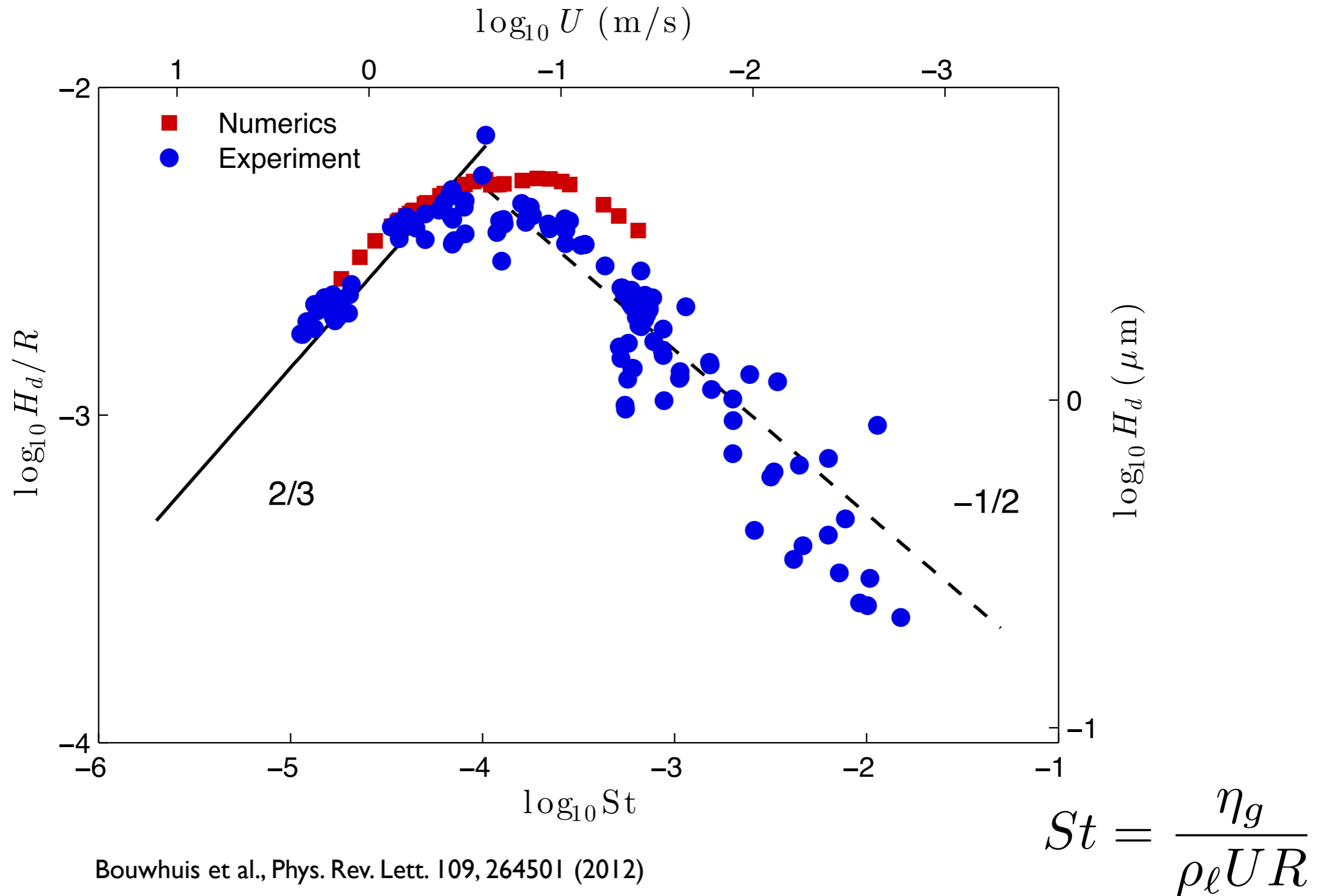
small  $U$  (and/or smaller  $R$ ):

capillary forces make smaller dimples

# Entrapped bubble size at impact



# Experimental & numerical data: dimple height

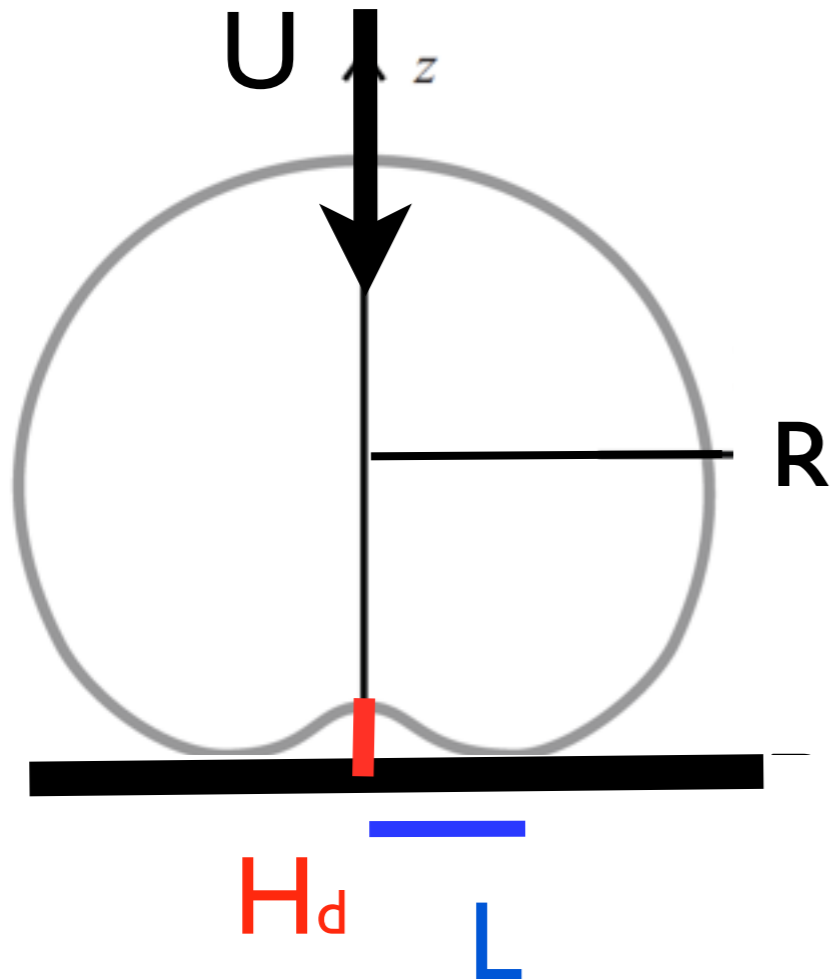


# **Indeed two regimes:**

- **inertial regime**
- **capillary regime**

**Analytical derivation of scaling laws in the two regimes**

# Inertial regime: Derivation of 4/3-scaling



horizontal lengthscale:

$$L \sim \sqrt{RH_d}$$

horizontal velocity:

$$u_x \sim LU/H_d$$

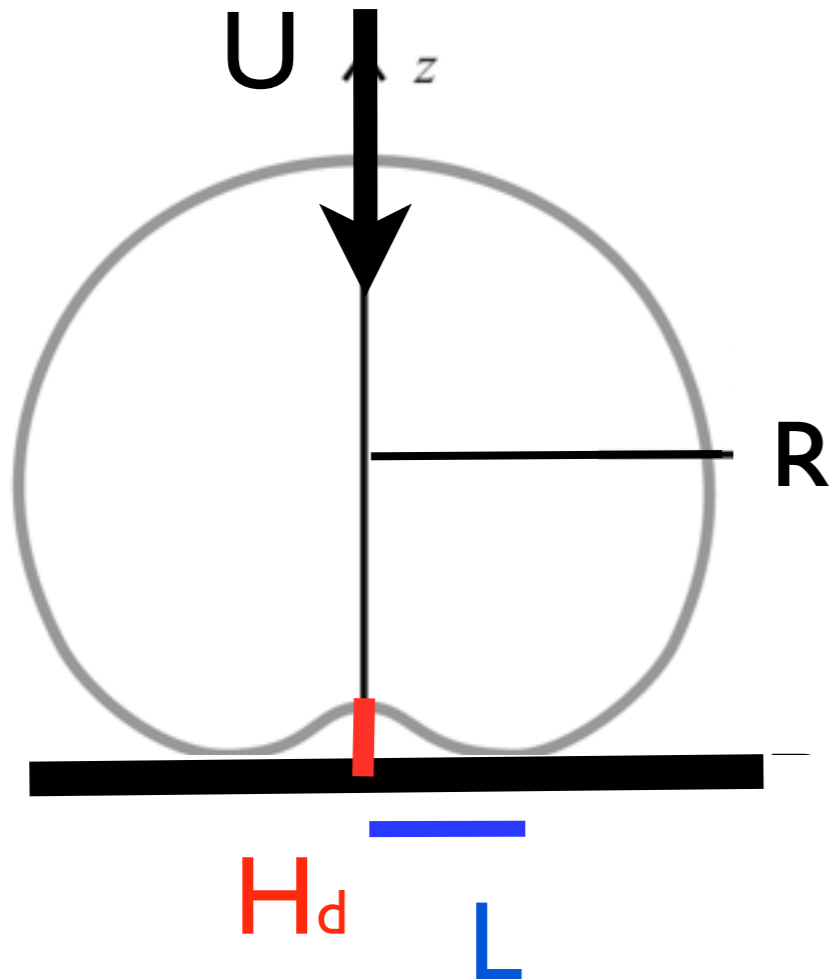
gas pressure: Stokes equation:

$$\partial_x p \sim \eta_g \partial_y^2 u_x$$

$$\frac{P_g}{L} \sim \eta_g \frac{u_x}{H_d^2}$$



# Inertial regime: Derivation of 4/3-scaling



liquid pressure from  
unsteady Bernoulli eq.:

$$\partial_t \phi \sim P_l / \rho_l$$

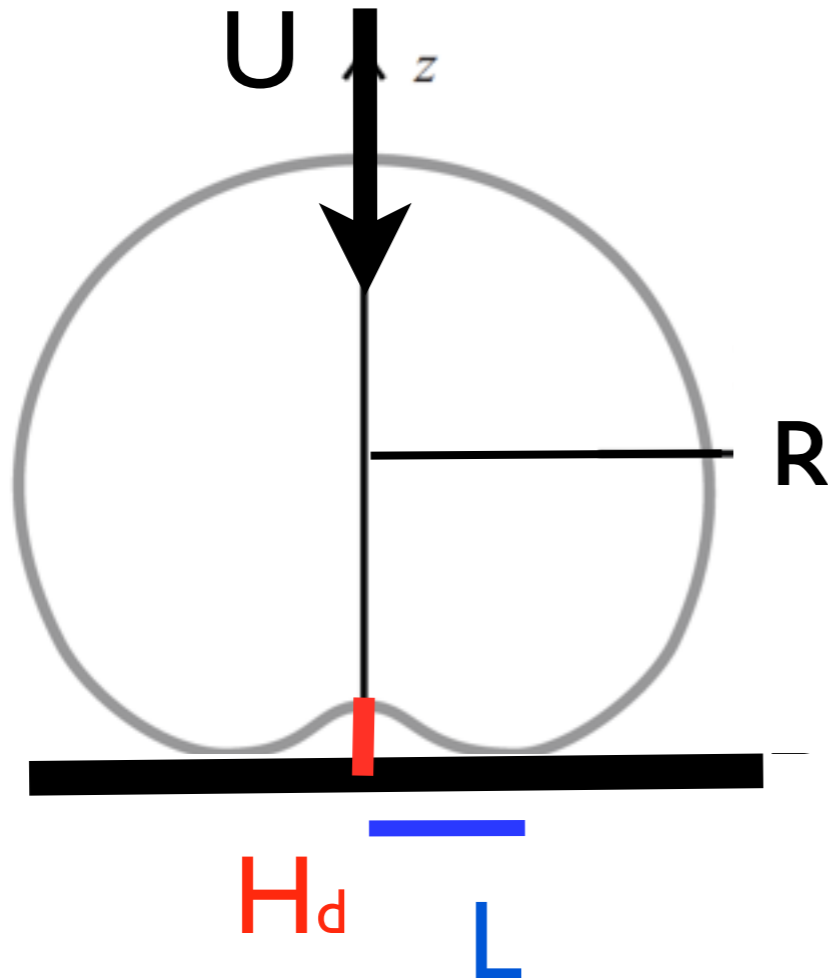
dimensional analysis:

$$\phi \sim UL \quad \partial_t \sim U/H_d$$

liquid pressure:

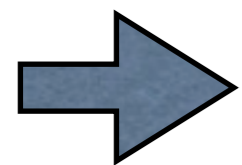
$$P_l \sim \rho_l U^2 L / H_d$$

# Inertial regime: Derivation of 4/3-scaling

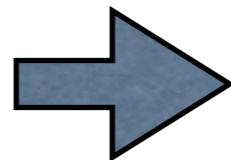


Droplet deforms once:

$$P_g \sim P_l$$

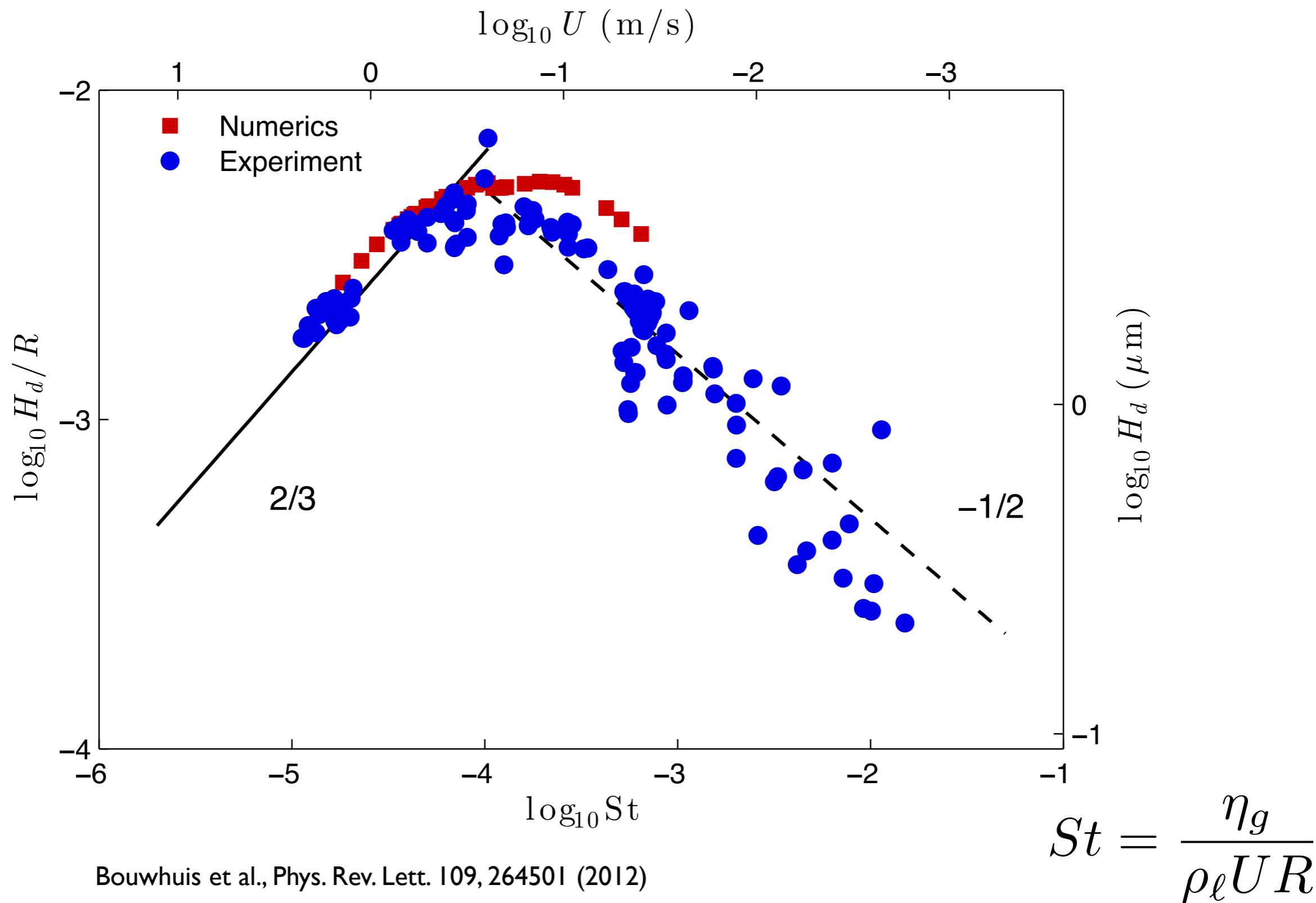


$$H_d \sim R St^{2/3}$$



$$V_b \sim L^2 H_d \sim R^3 St^{4/3}$$

# Experimental & numerical data: dimple height



# Scaling in capillary regime

balance capillary and viscous forces:

$$\partial_x(\gamma\kappa(H(x))) \sim \eta_g \partial_z^2 u_x$$

$$\frac{H_d}{R} \sim \sqrt{Ca} \sim \sqrt{St We} \sim \frac{\eta_g}{\sqrt{\gamma\rho_l R}} St^{-1/2}$$

Crossover, when:  $St \sim Ca^{3/4}$

# Maximum in dimple height

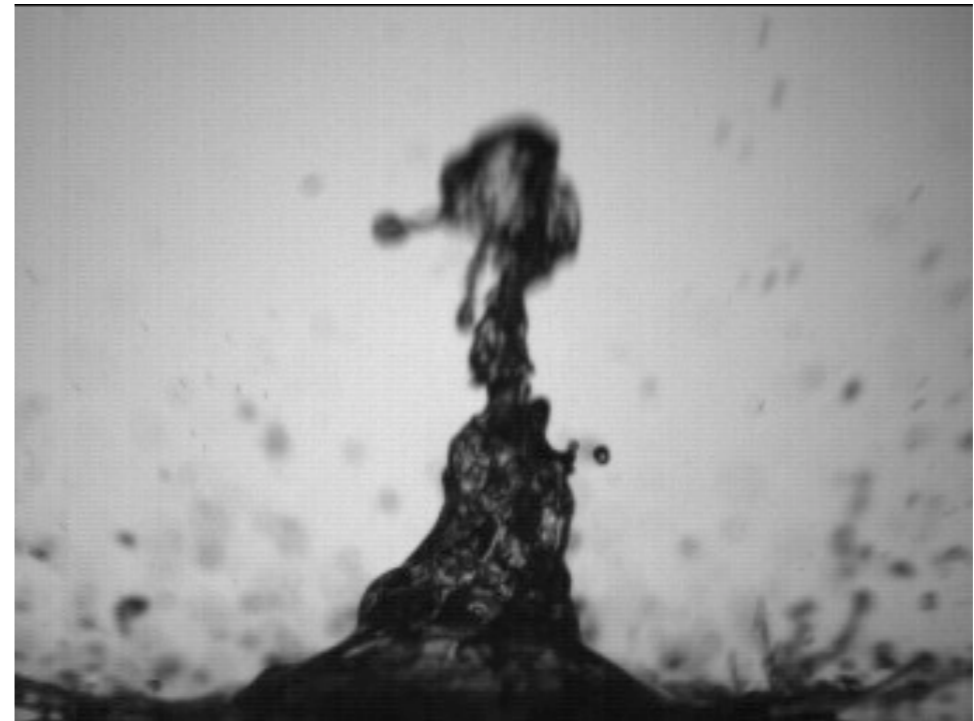
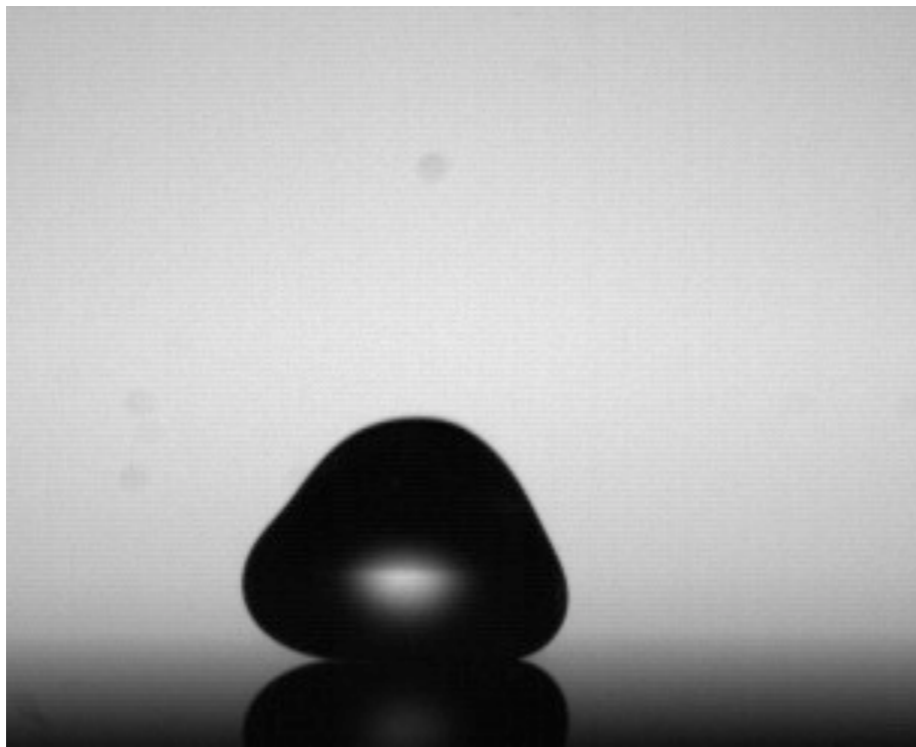
at:

$$St \sim Ca^{3/4} \sim \frac{\eta_g^{6/7}}{(\gamma \rho_l R)^{3/7}}$$

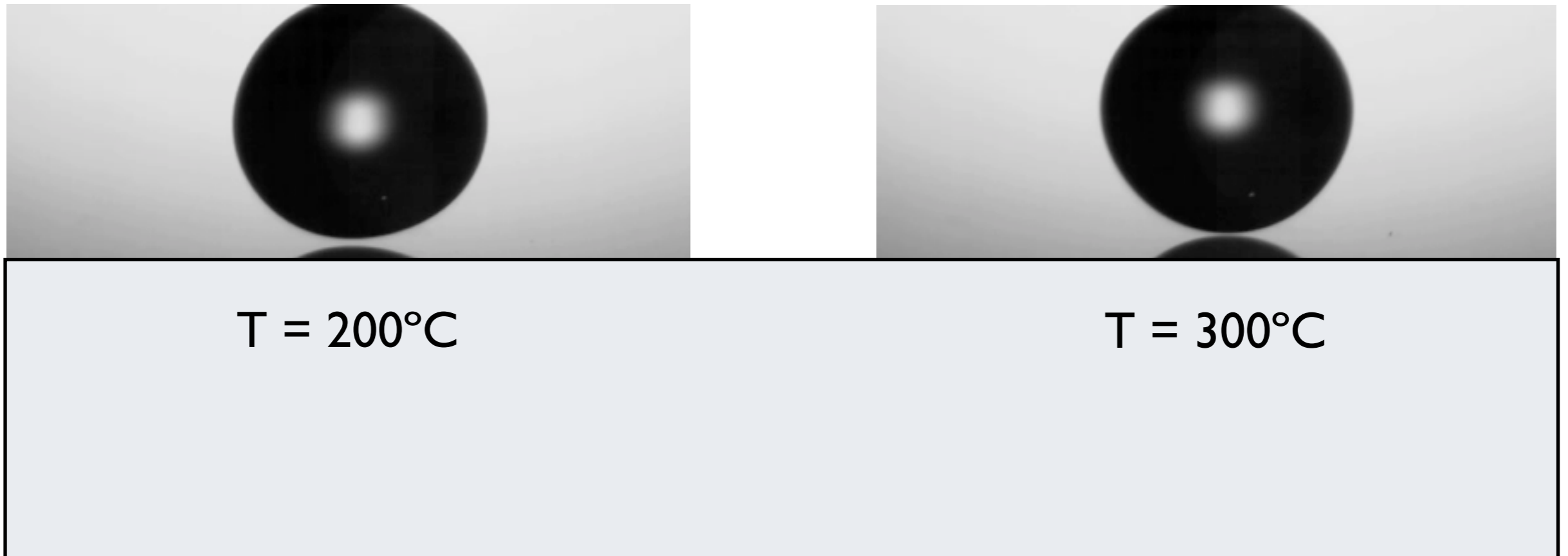
smaller  $St$  = larger  $U$  (and/or larger  $R$ ):  
inertia of impact makes smaller dimples

larger  $St$  = smaller  $U$  (and/or smaller  $R$ ):  
capillary forces make smaller dimples

# Impact on **superheated** surfaces

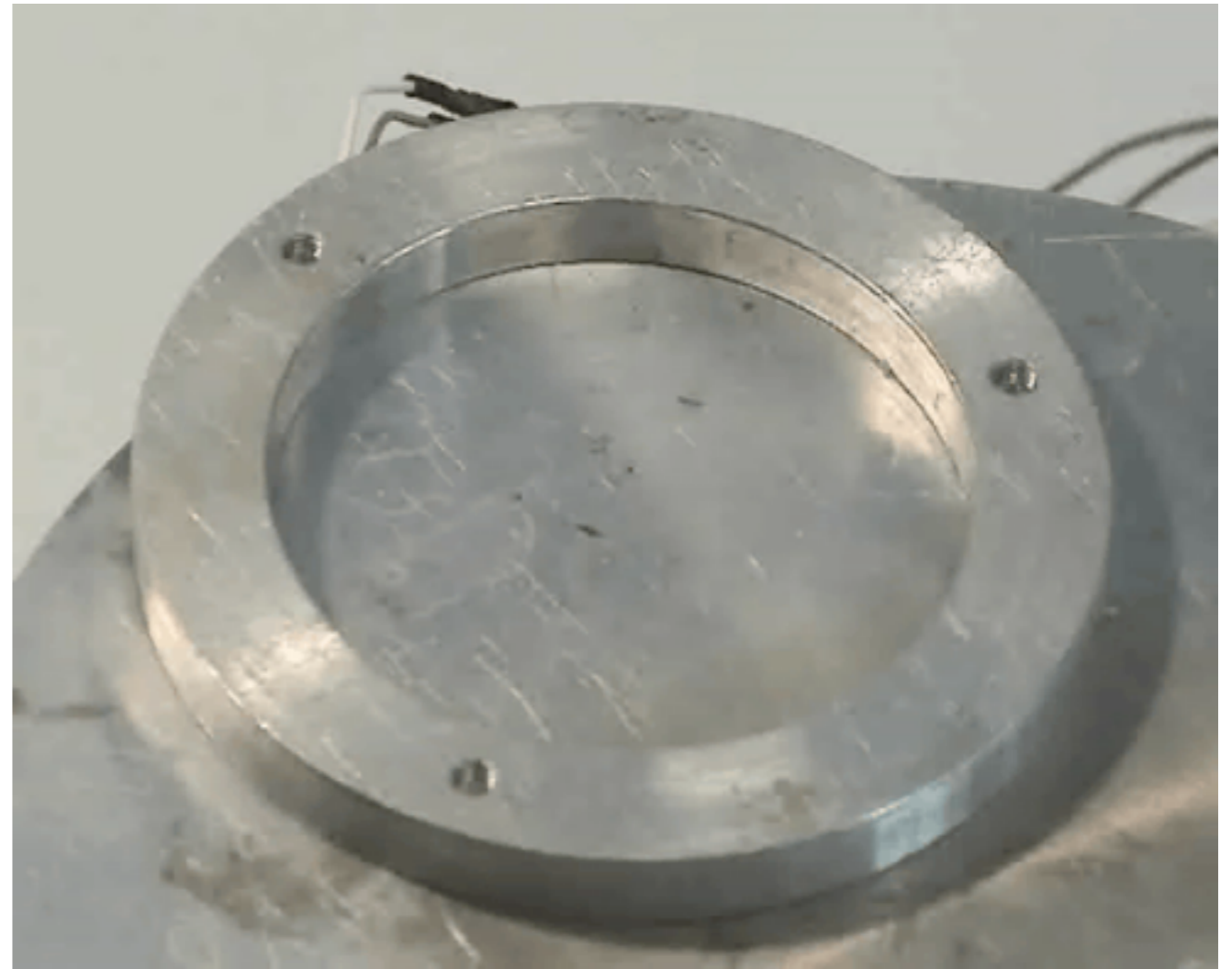


# Influence of surface temperature

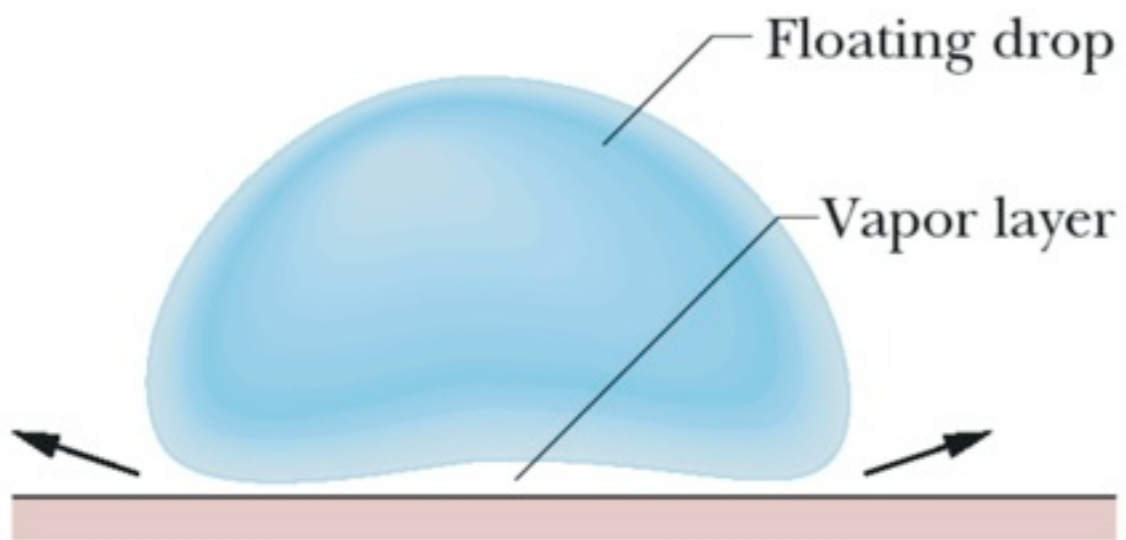


$$T_{Leidenfrost} = 180^{\circ}C \text{ (static)}$$

# Static Leidenfrost effect



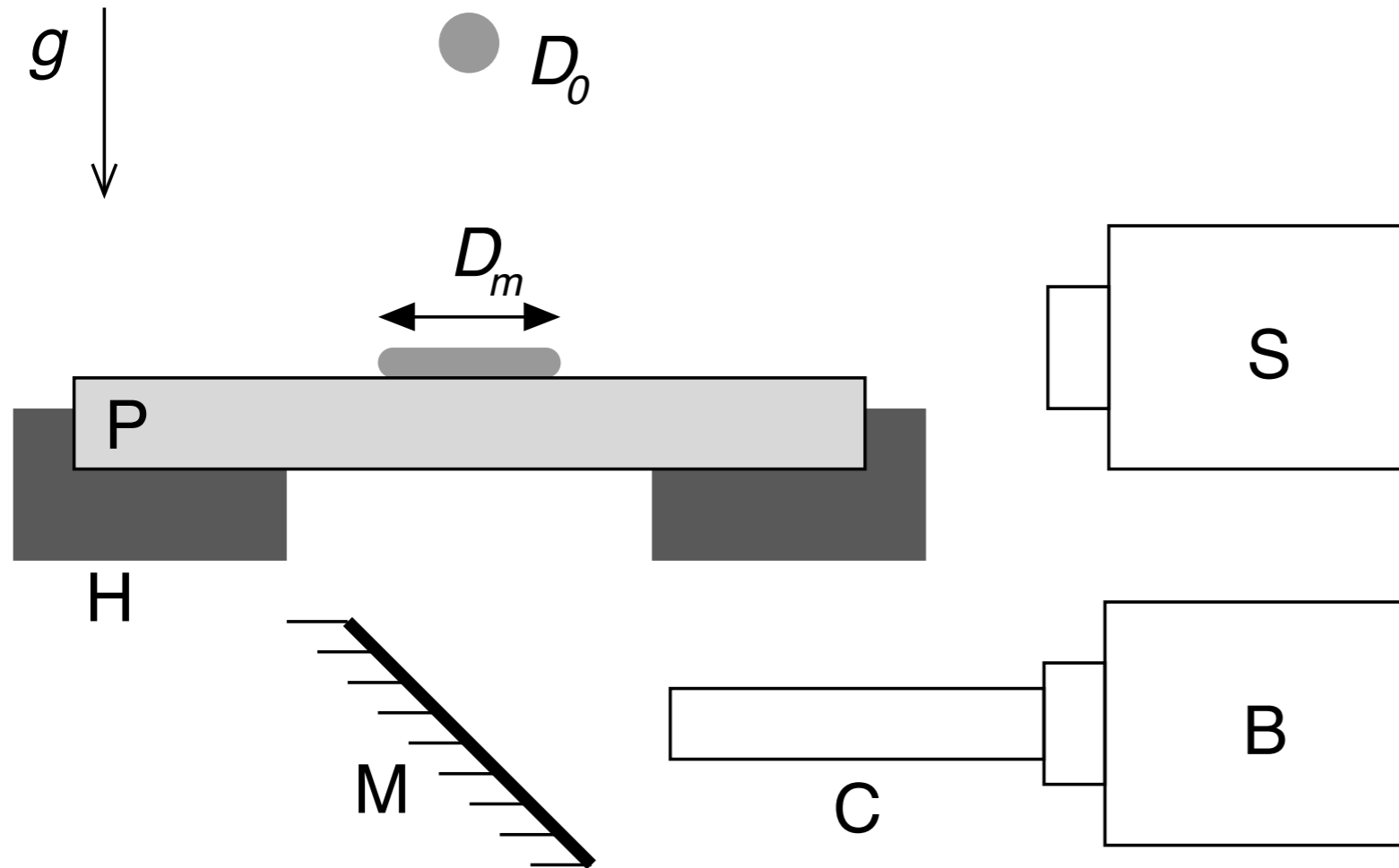
*Johann Gottlob Leidenfrost (1756)*





# Towards the **dynamic** Leidenfrost effect

# Experimental setup

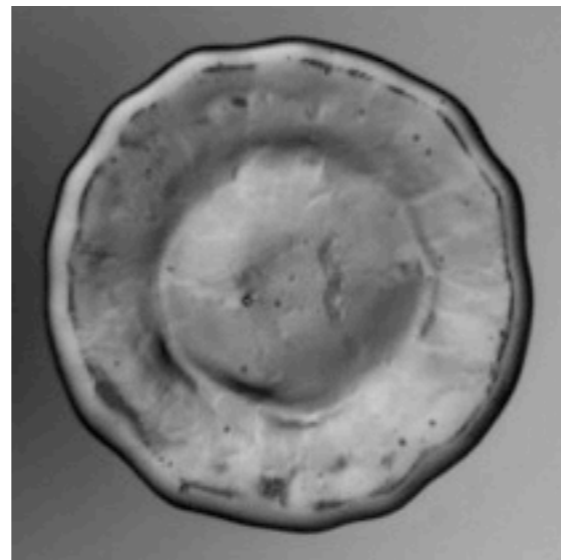
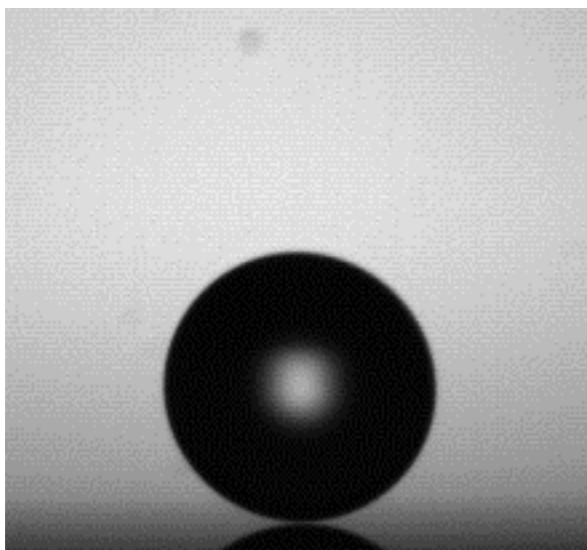


- Liquids: water & FC-72
- Surfaces: polished silicon, structured silicon, **sapphire** & carbon nano fibers

- Control parameters:

$$200^{\circ}\text{C} \leq T \leq 600^{\circ}\text{C}$$

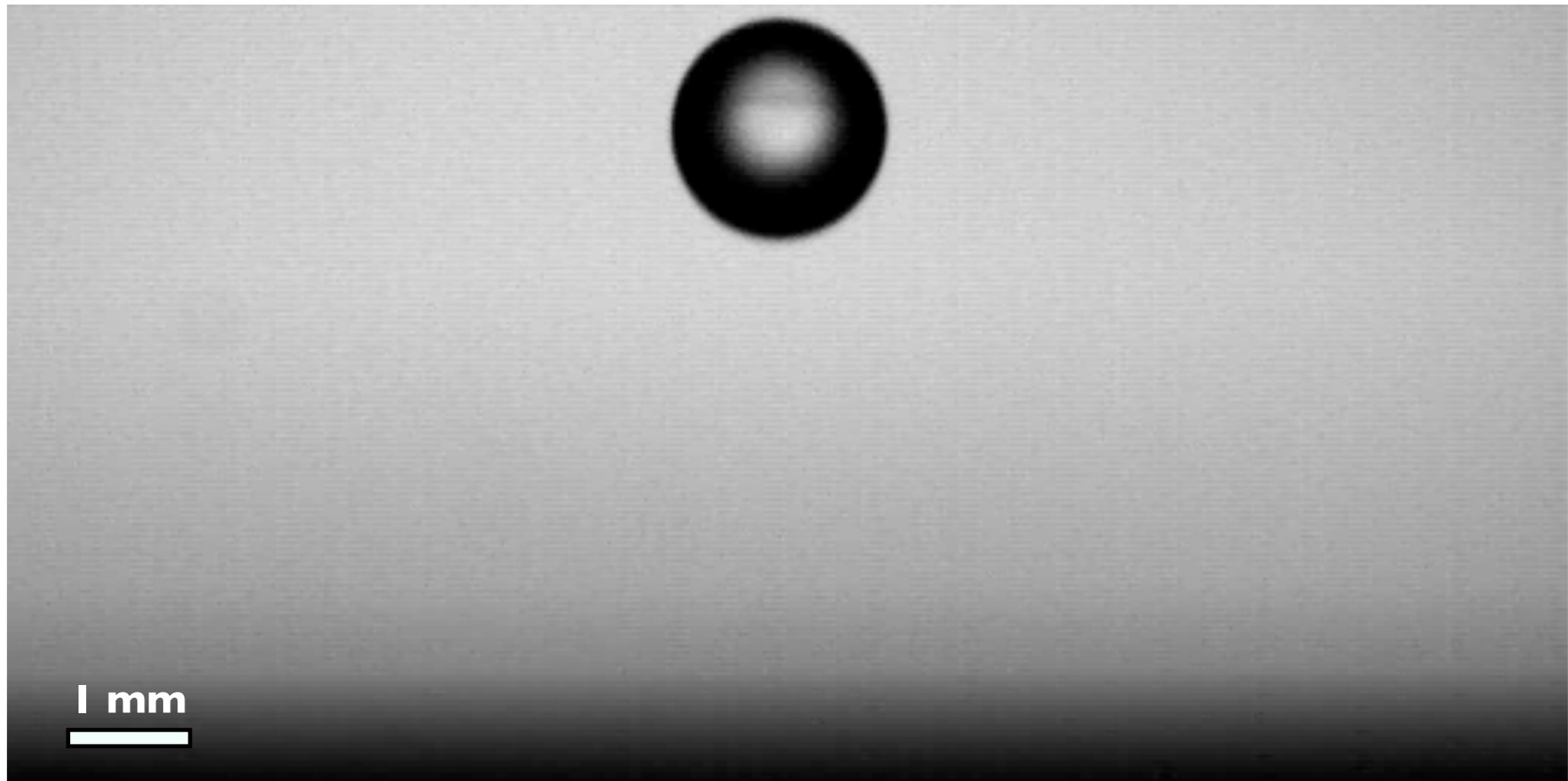
$$0.5 \leq \text{We} = \frac{\rho D_0 V^2}{\sigma} \leq 500$$



# **Different boiling regimes:**

# I. Contact boiling

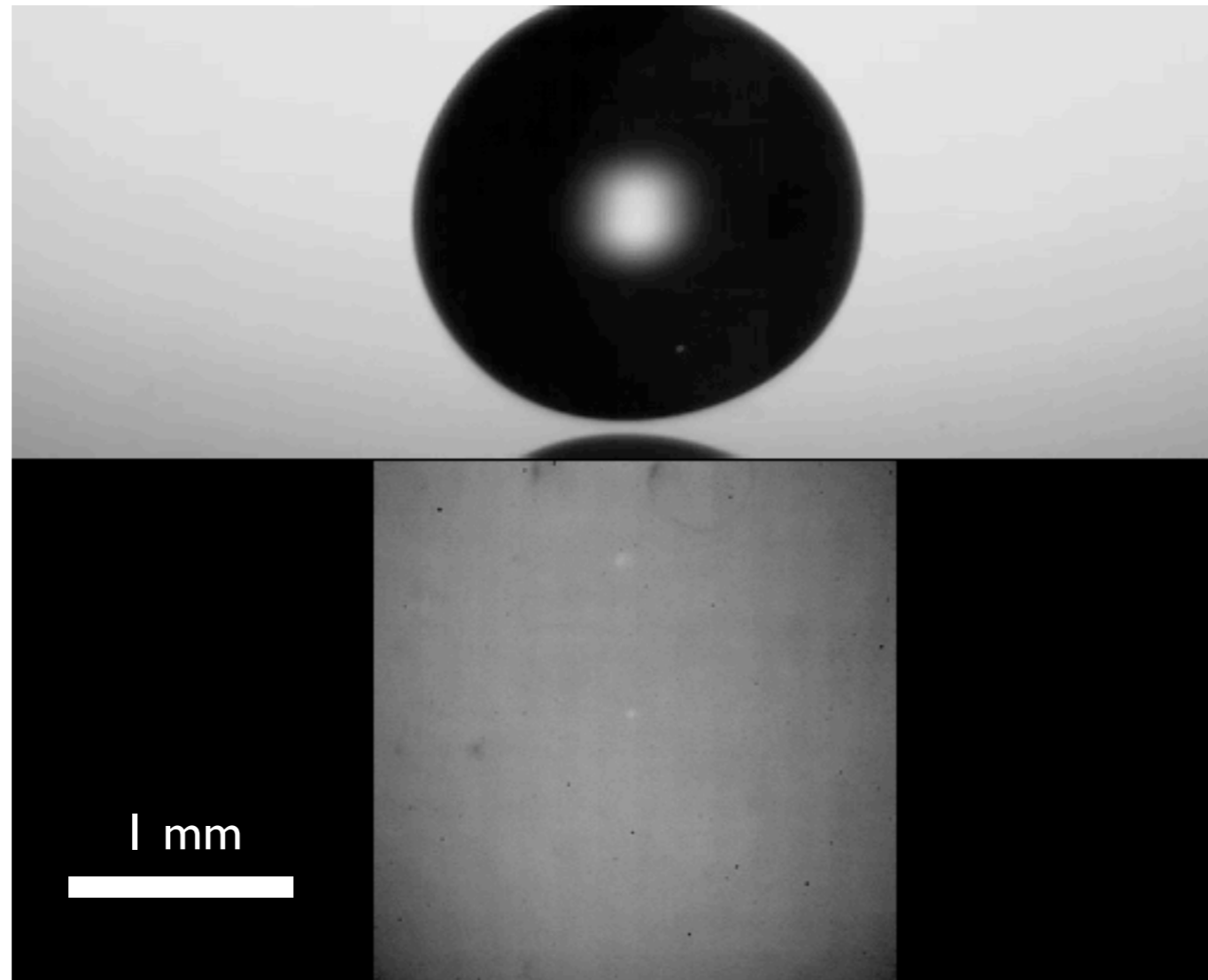
Side view recording



$T = 400 \text{ }^\circ\text{C}$   
 $We = 67.7$

# I. Contact boiling

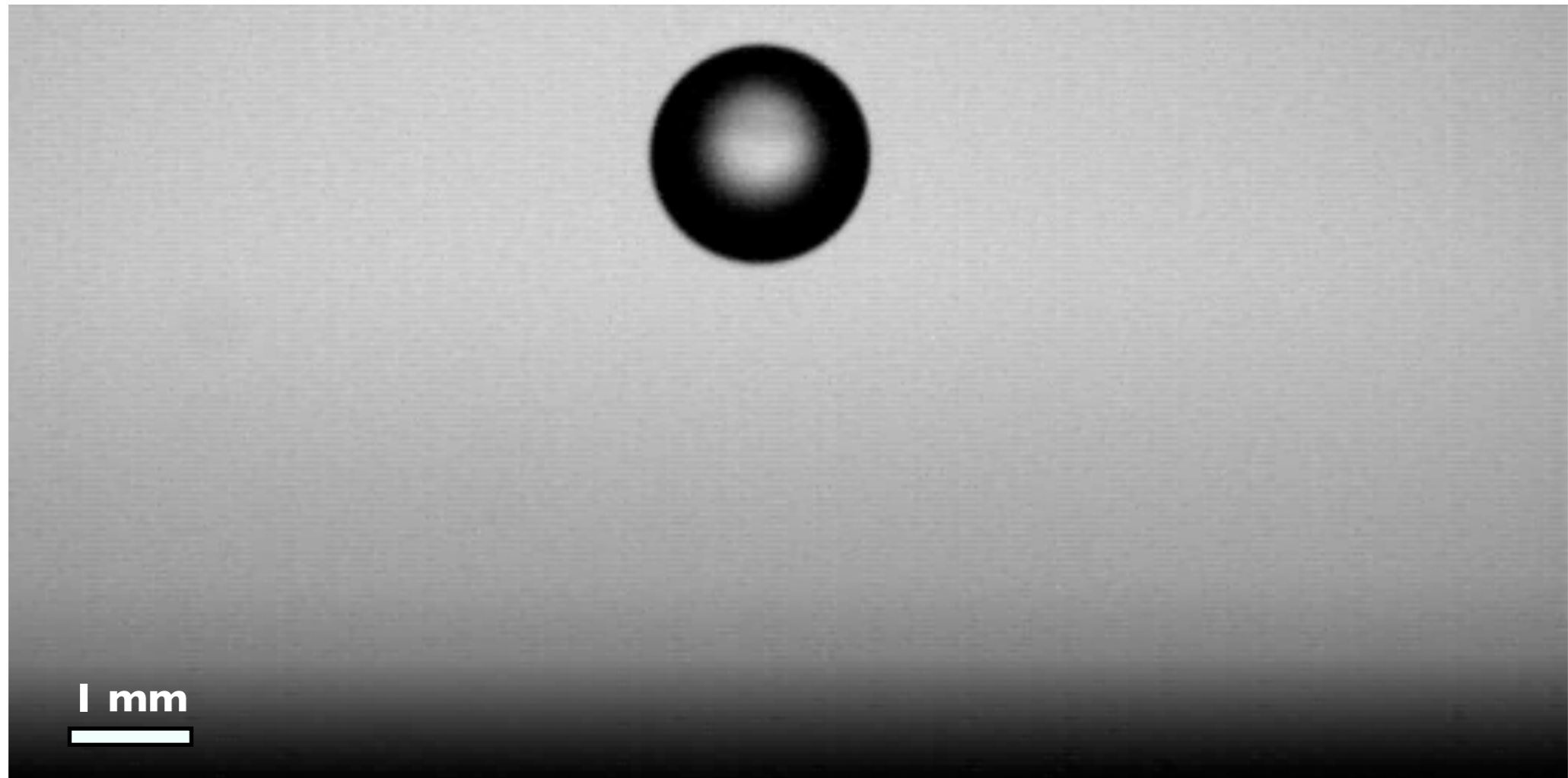
Bottom interferometric view



Liquid makes contact with the surface

## 2. Gentle film boiling

Side view recording

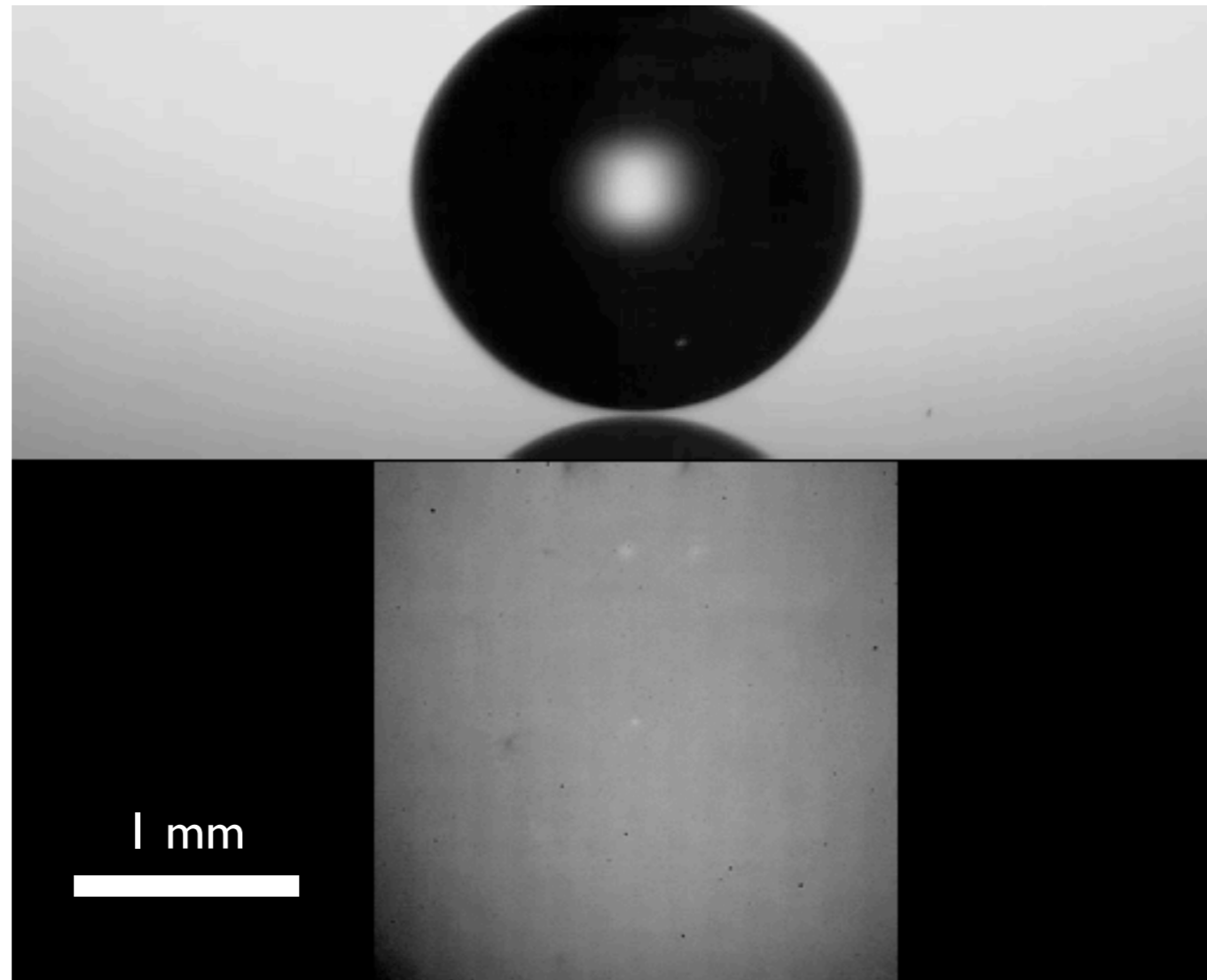


$$T = 460 \text{ }^{\circ}\text{C}$$

$$We = 66.7$$

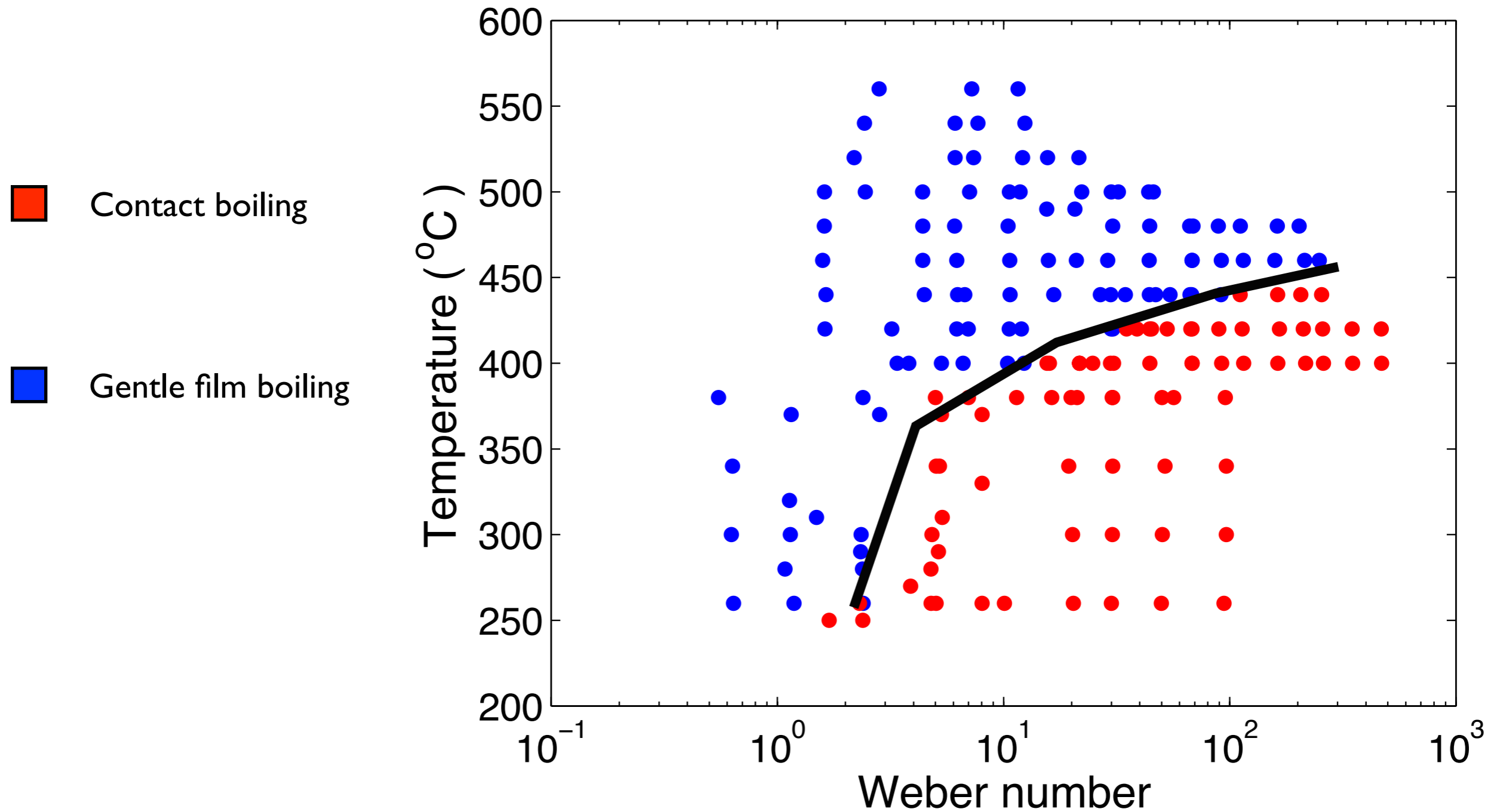
## 2. Gentle film boiling

Bottom interferometric view



Liquid makes **no contact** with the surface:  
(dynamic) Leidenfrost state

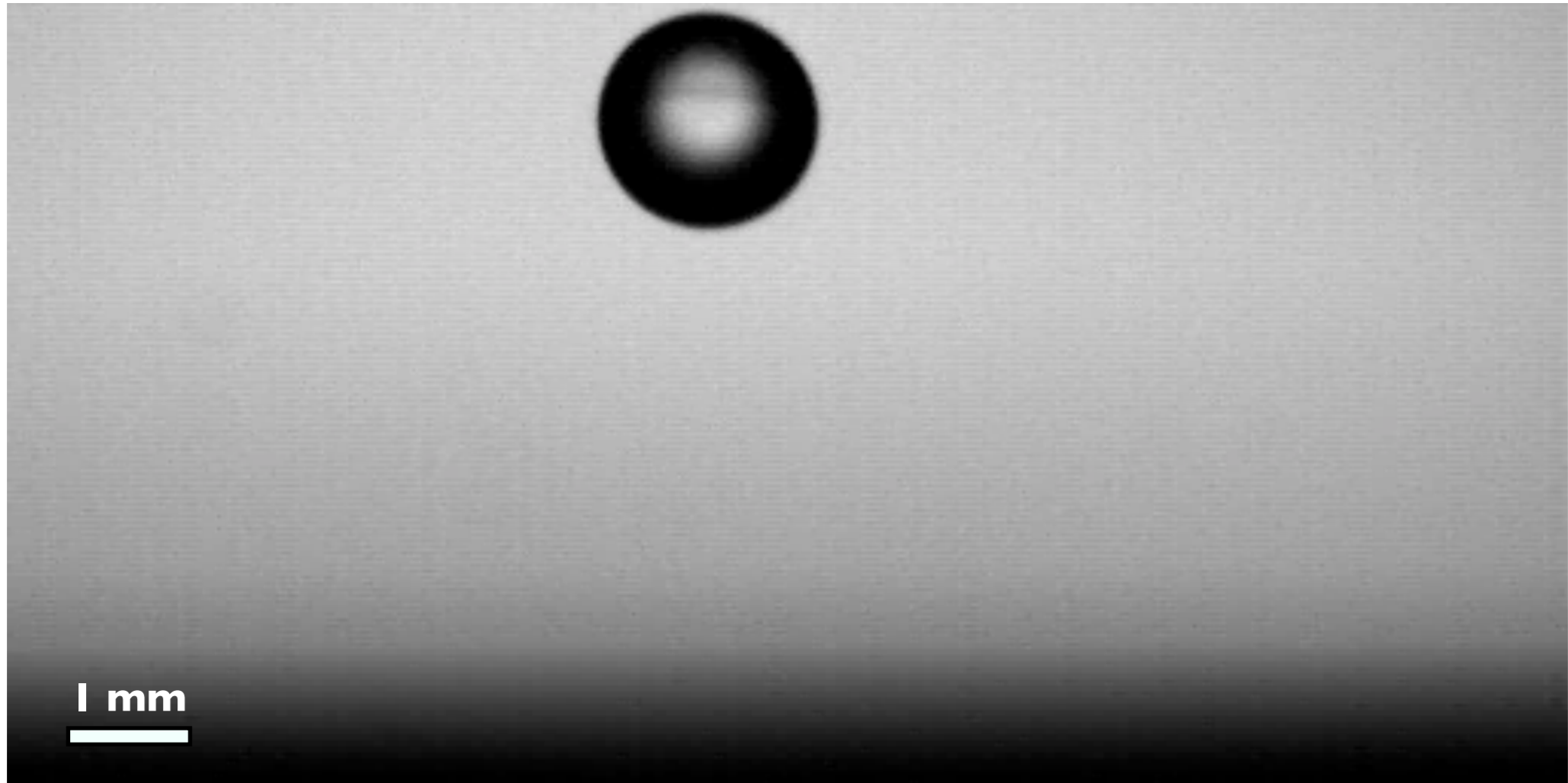
# Phase diagram water on smooth silicon





# 3. Spraying film boiling

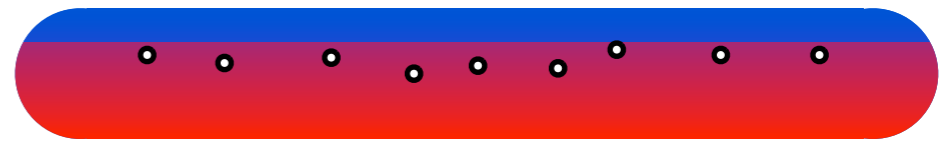
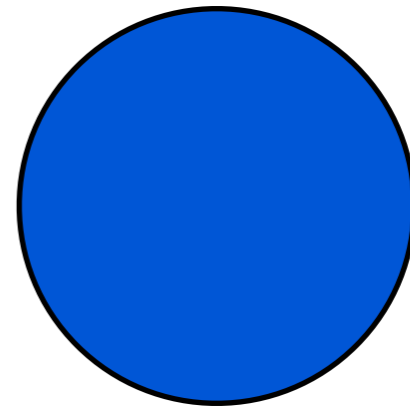
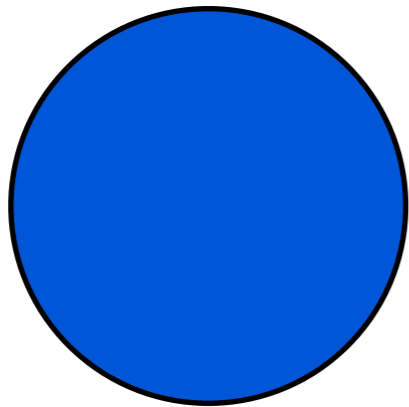
Side view recording



$T = 520 \text{ }^{\circ}\text{C}$

$We = 65.4$

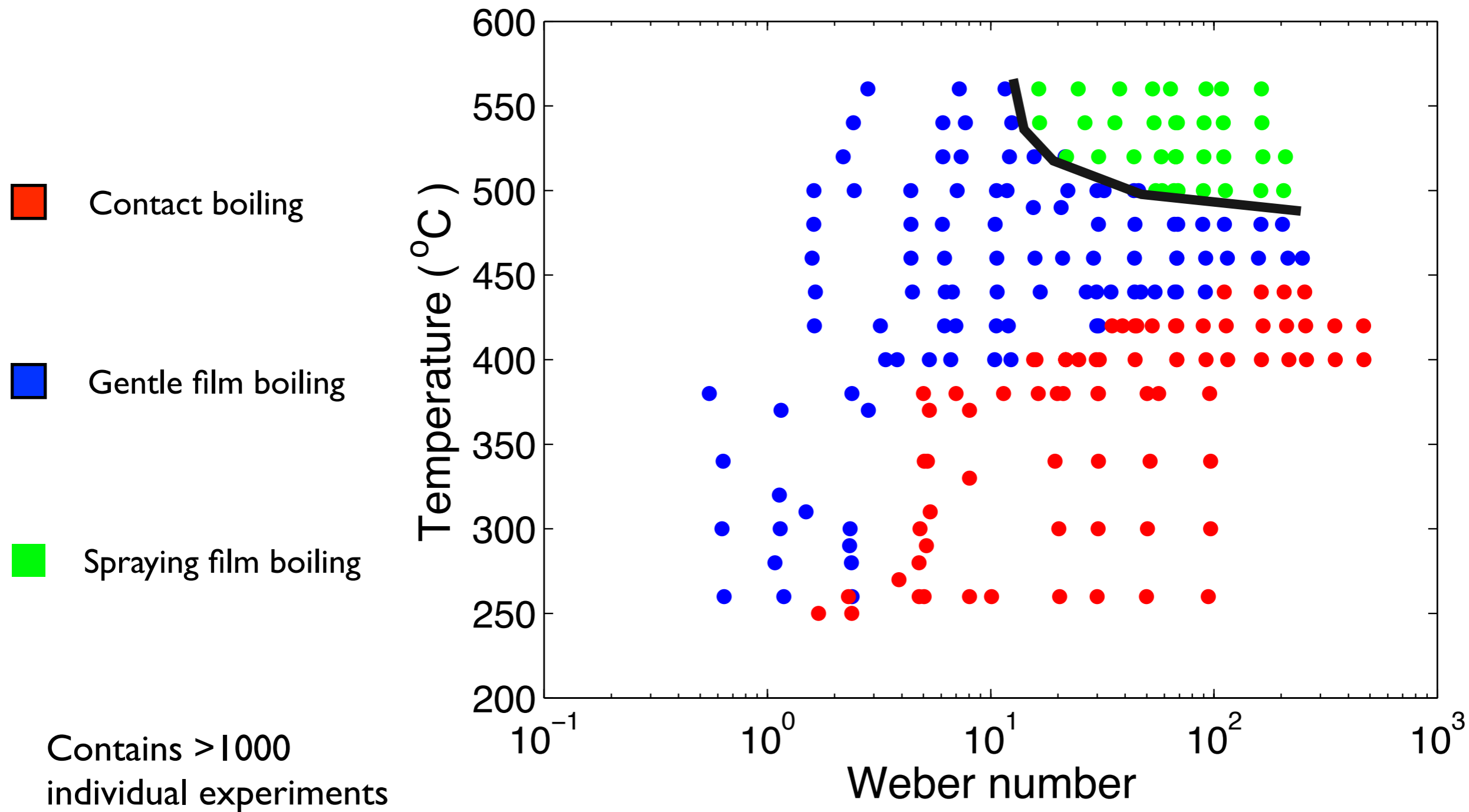
# Mechanism of spraying film boiling



Low Weber number

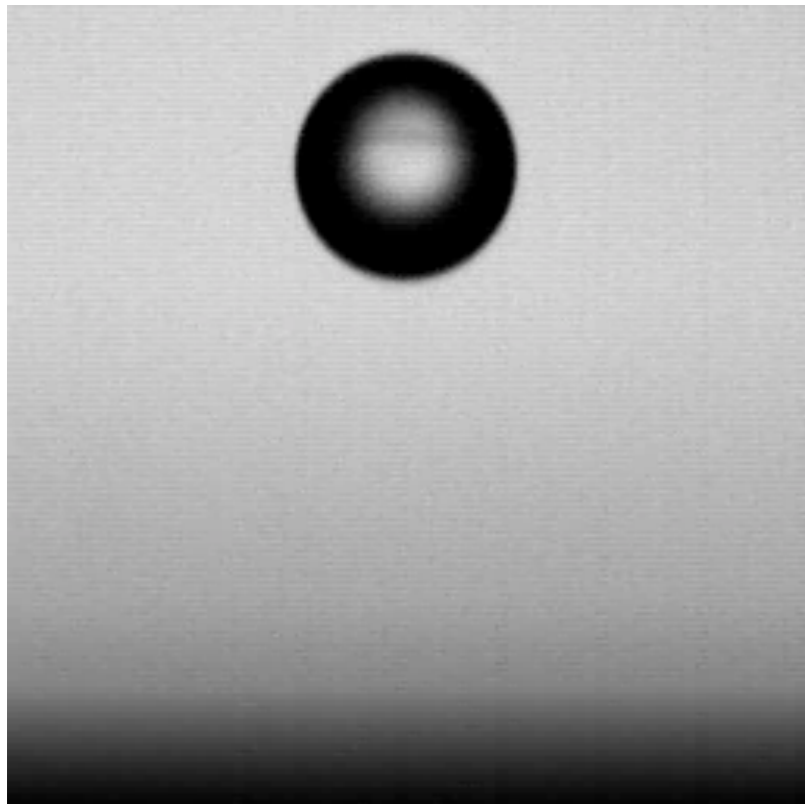
High Weber number

# Phase diagram water on smooth silicon



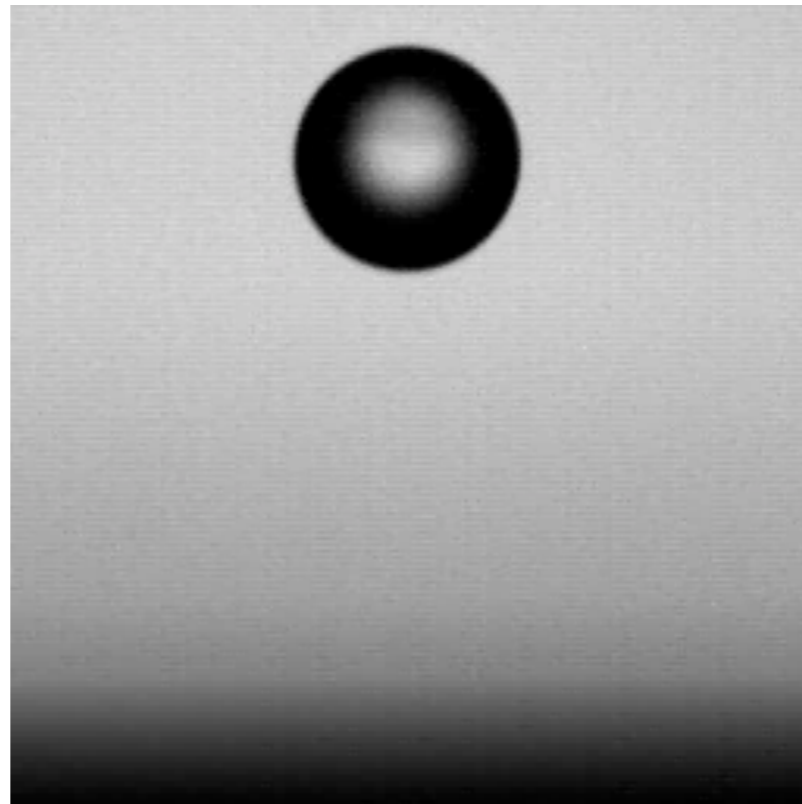
# The three scenarios in comparison

Contact boiling



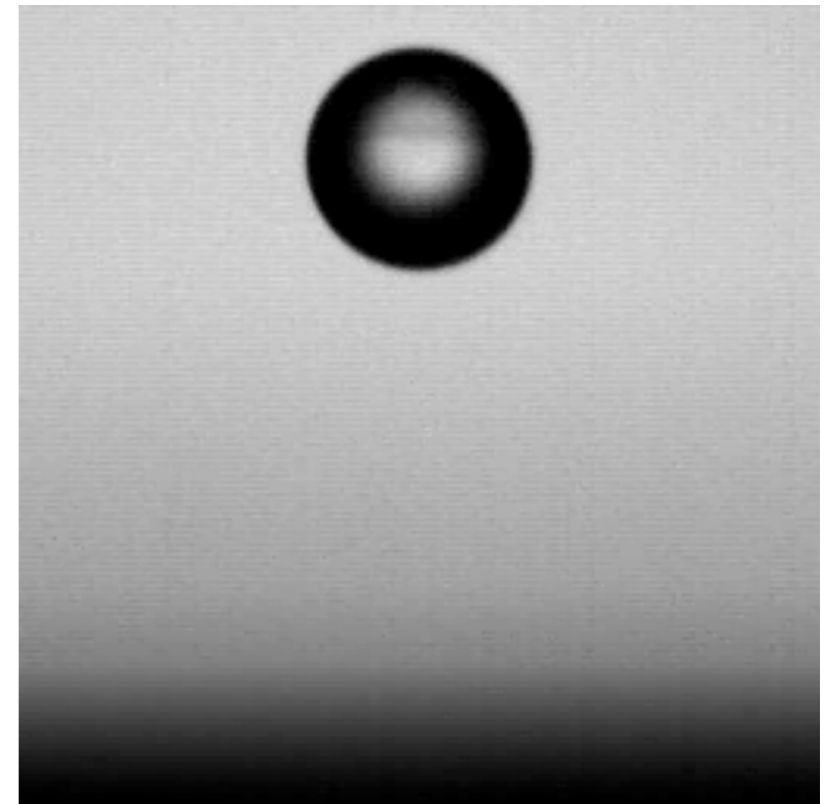
$T = 400 \text{ }^{\circ}\text{C}$

Gentle film boiling



$T = 460 \text{ }^{\circ}\text{C}$

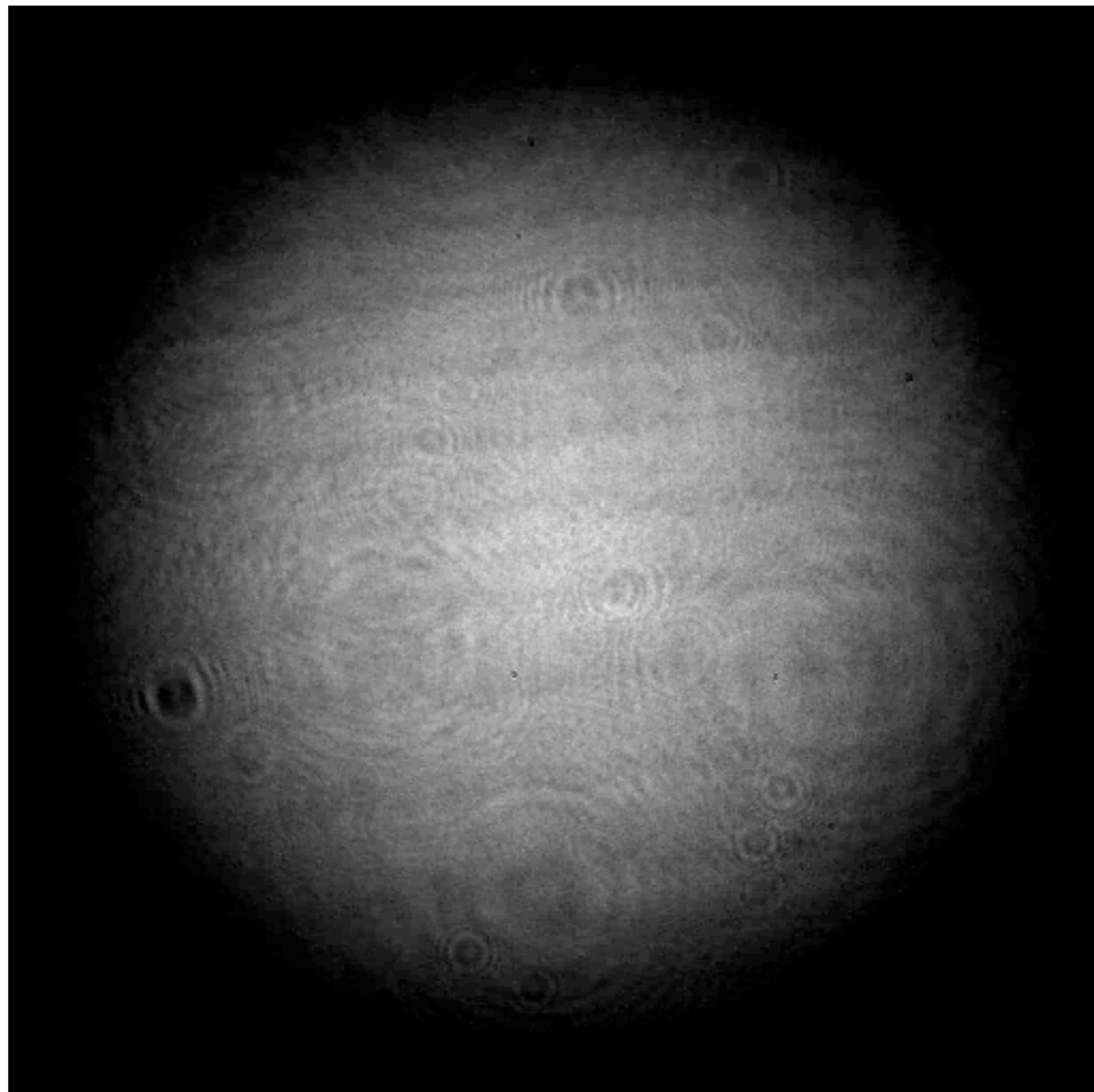
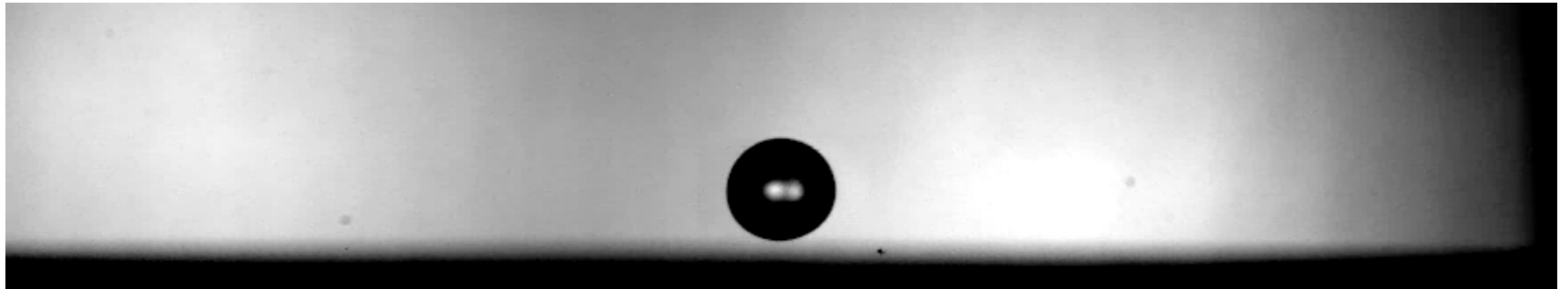
Spraying film boiling



$T = 520 \text{ }^{\circ}\text{C}$

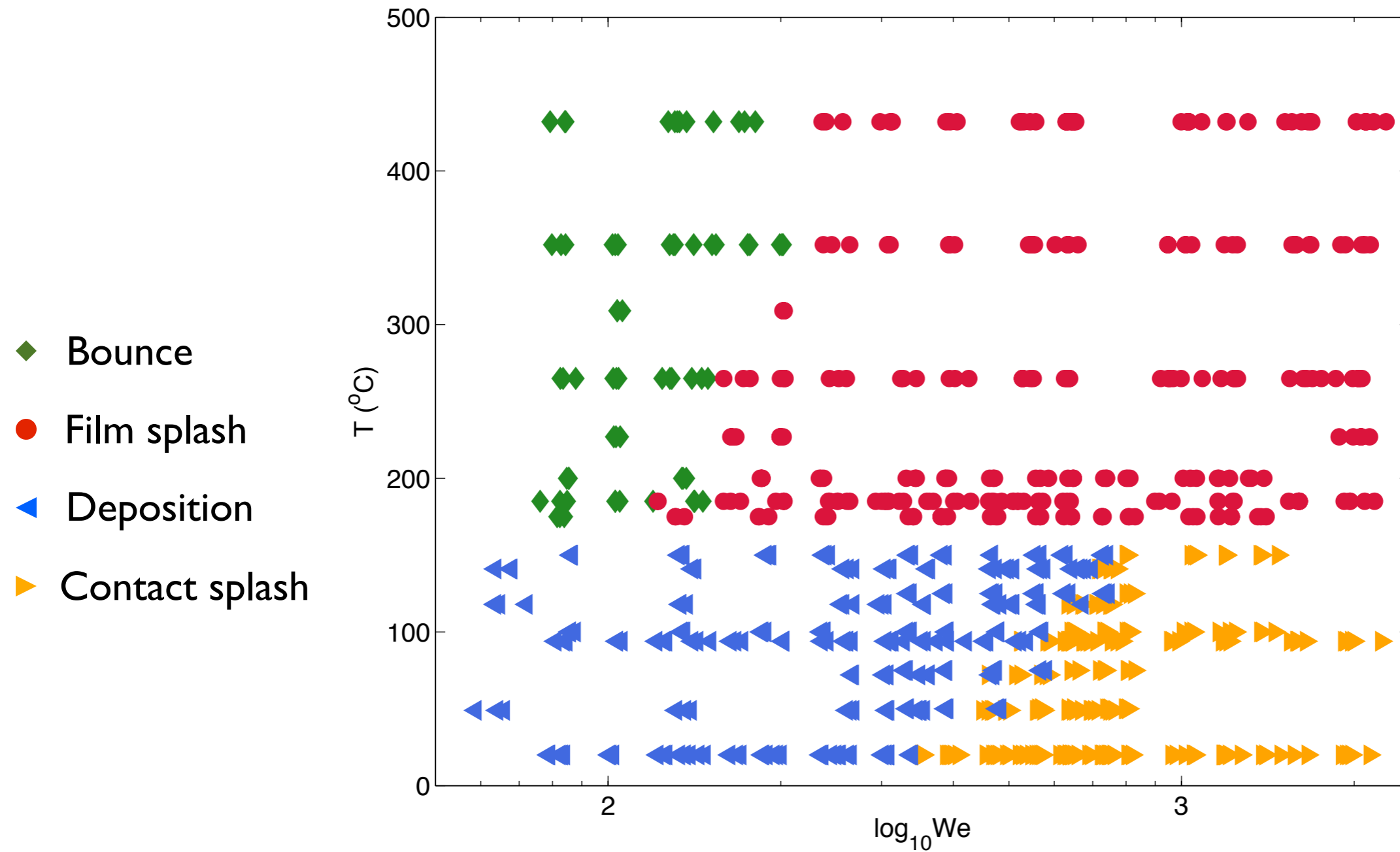
**Is splashing possible in the  
Leidenfrost regime?  
(no contact!)**

# Film boiling splash

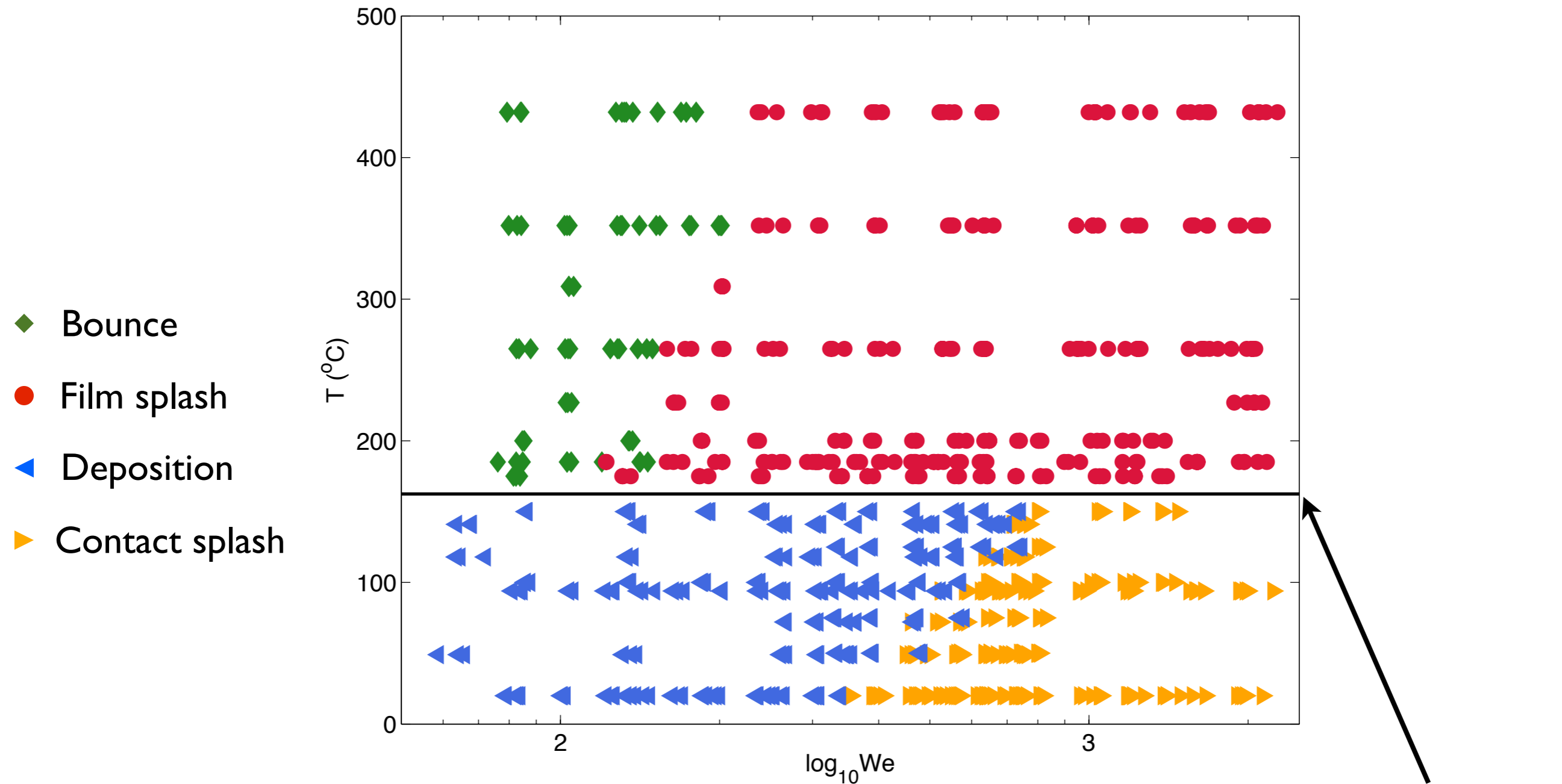


$T = 200 \text{ }^\circ\text{C}$   
 $D = 2.5\text{mm}$   
 $We = 1390$   
ethanol

# Full phase space of boiling & splashing behavior



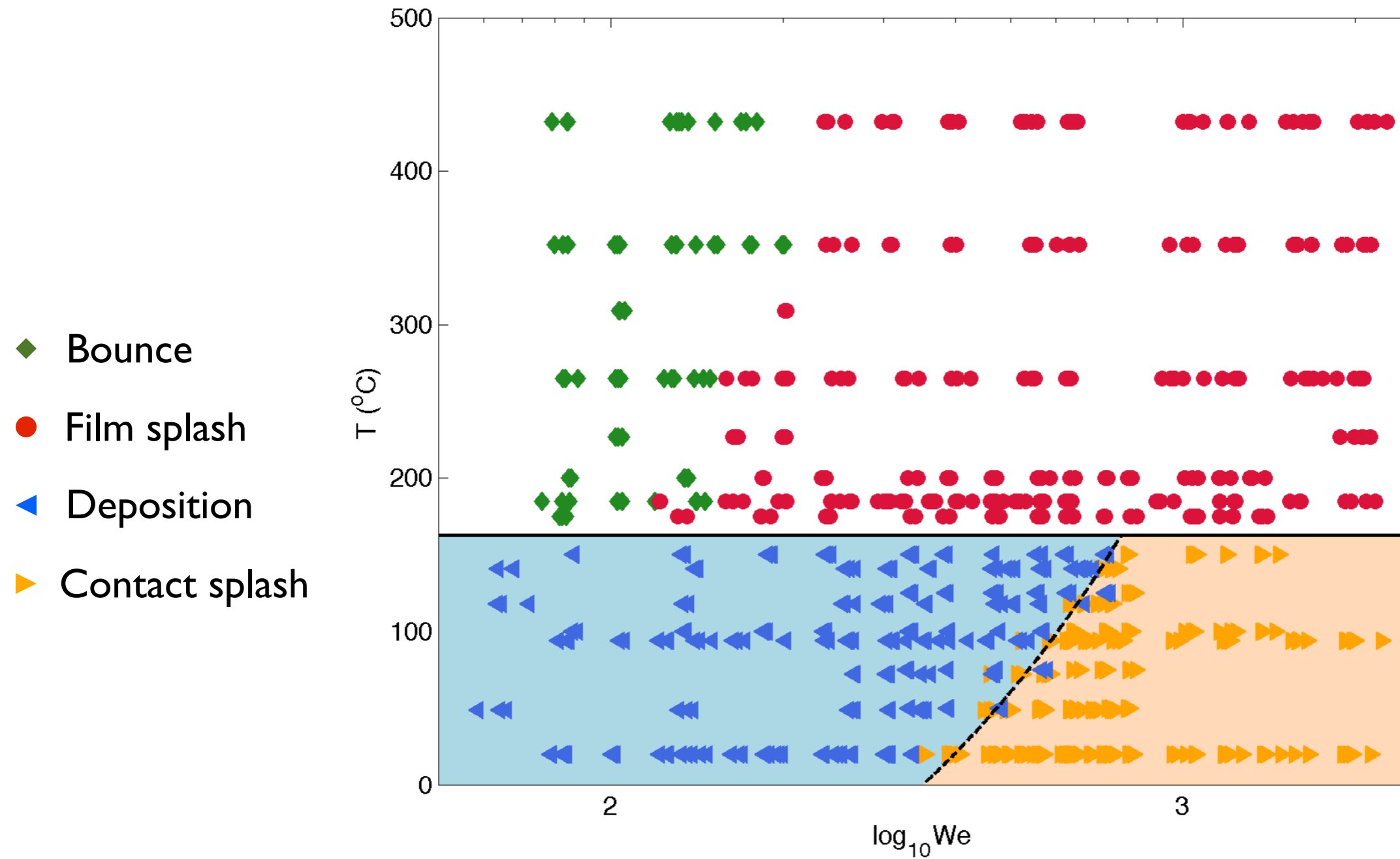
# Transition to film boiling



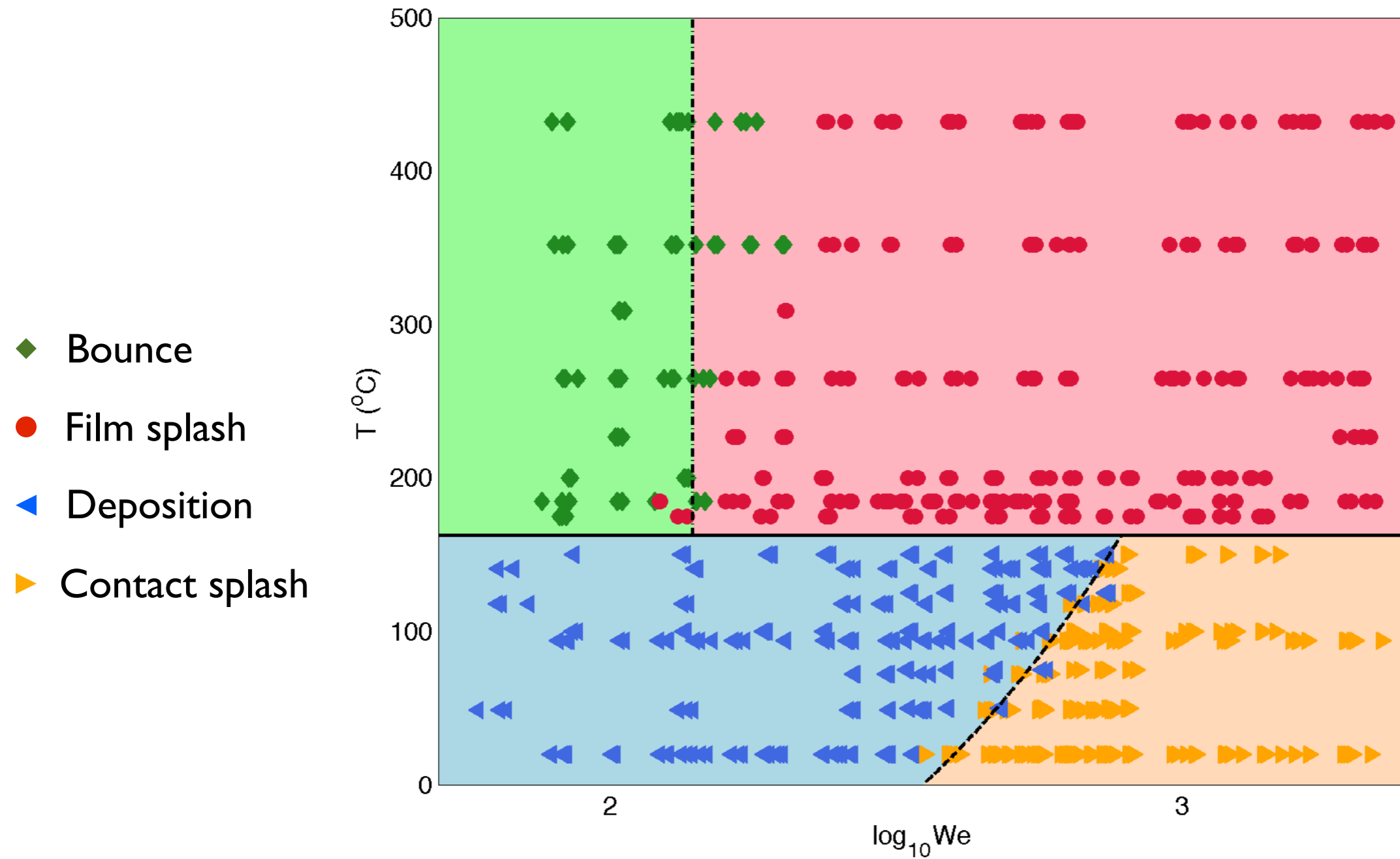
Transition temperature to film boiling regime:  $T_L = 162$  °C



# below $T_L$ : increasing temperature suppresses splashes!



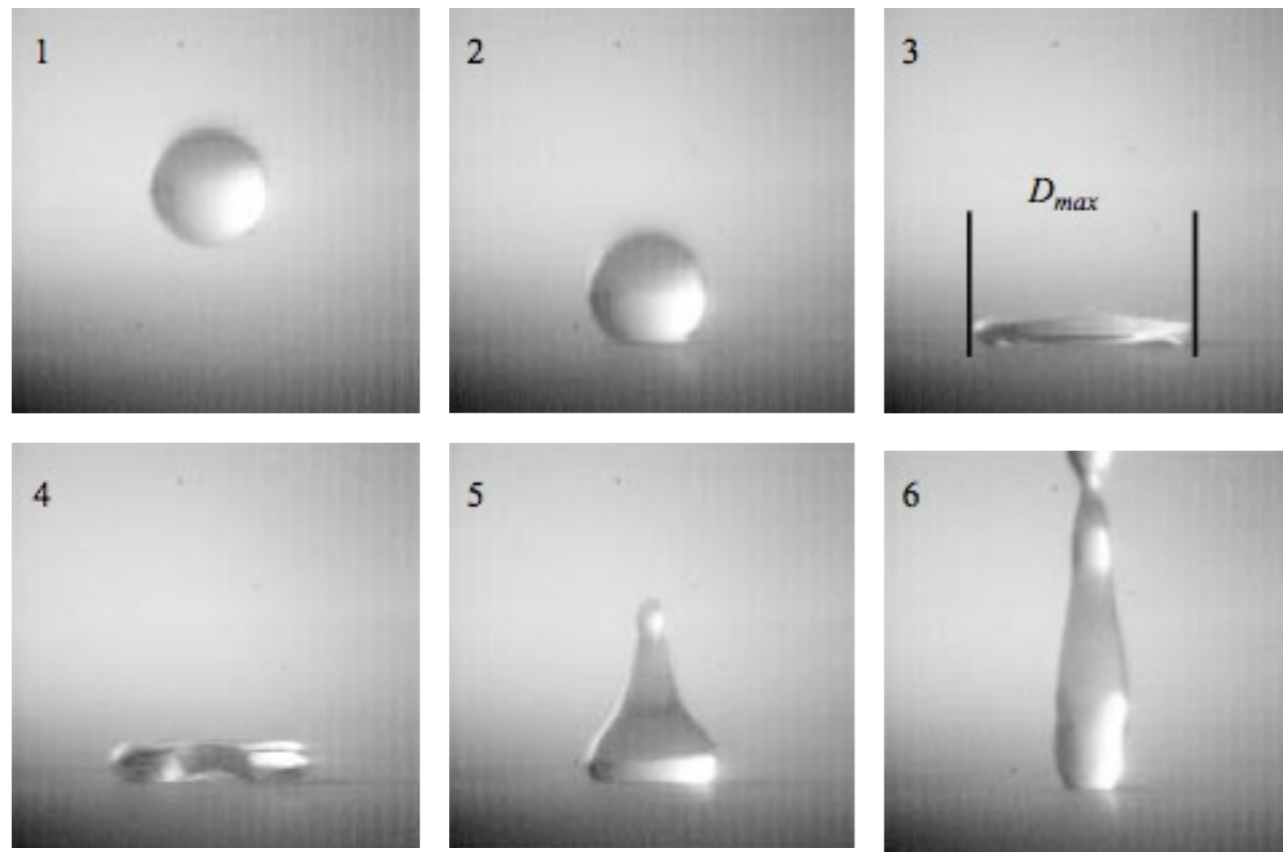
**above  $T_L$ : much lower We-  
threshold for splashing !!**



**How much do droplets spread?**

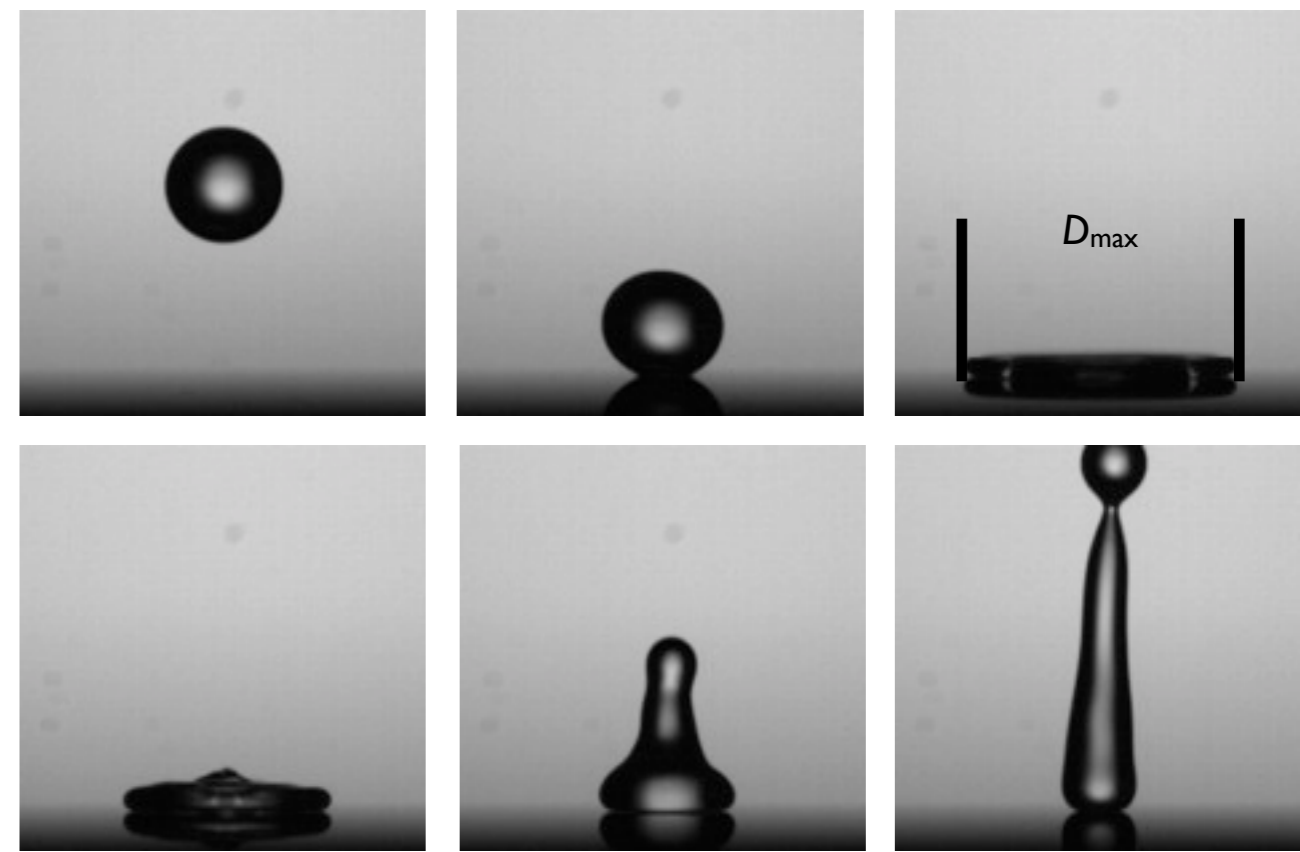
# Spreading of impacting droplets

Impact on (non-heated)  
**superhydrophobic** surface



$We = 24$

Impact on **superheated** surface  
in film boiling regimes



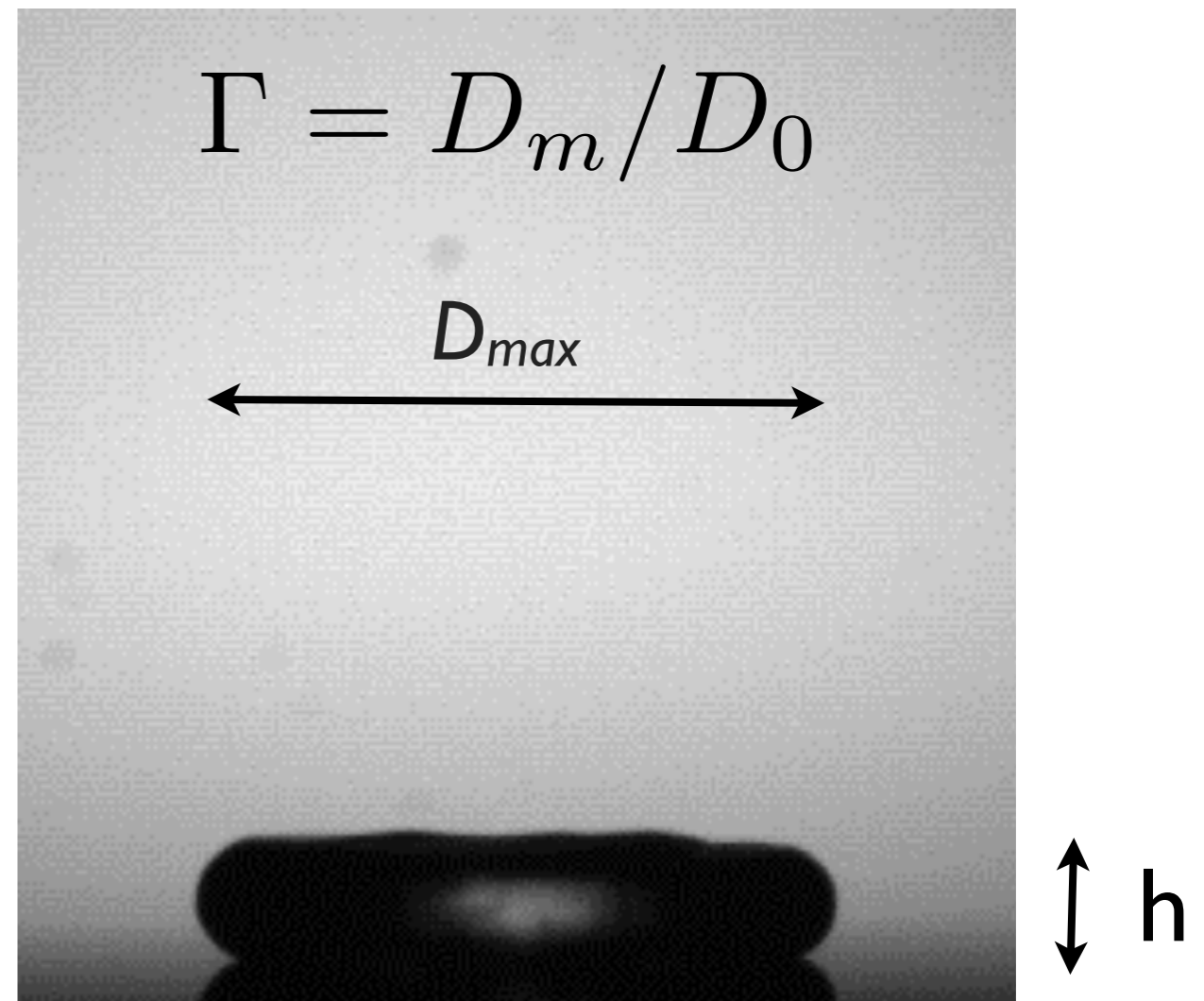
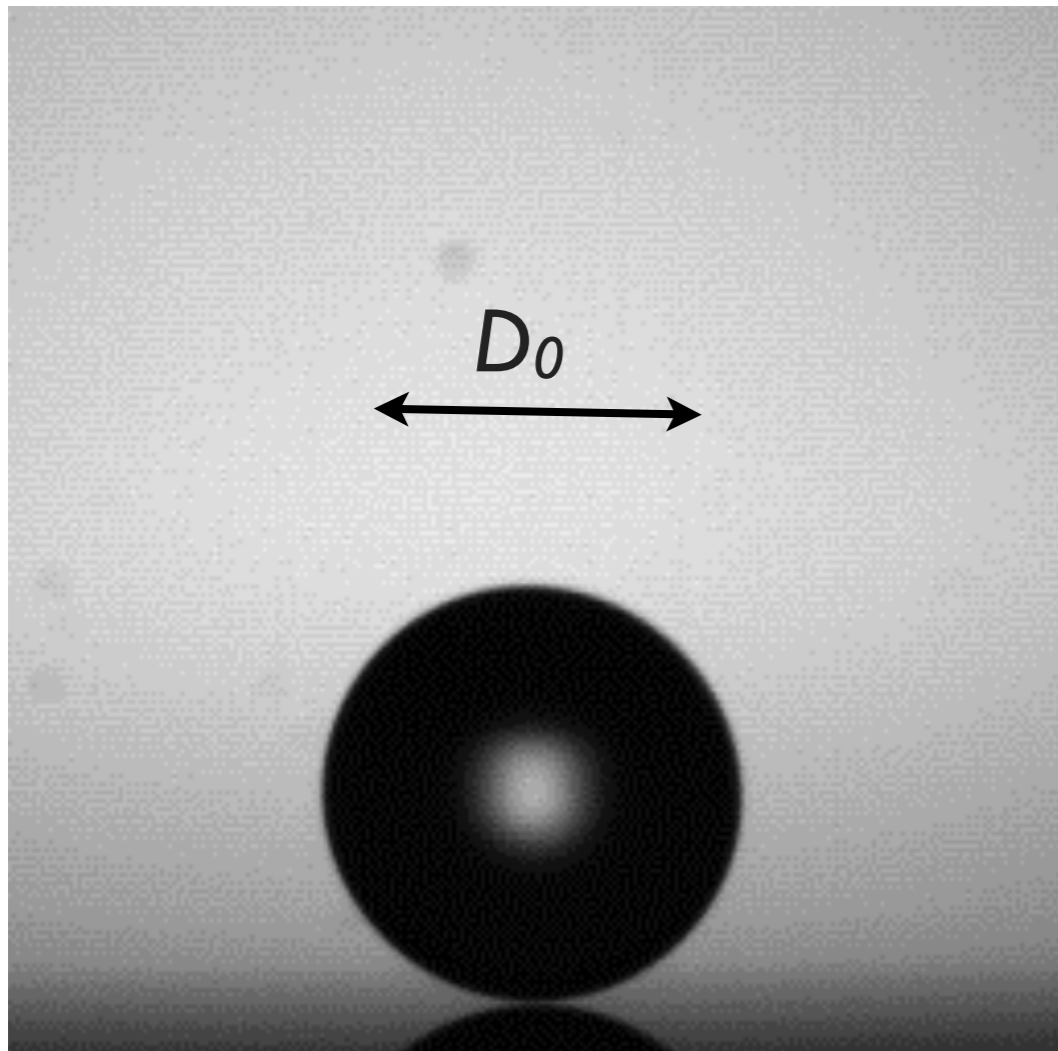
$We = 32$

Clanet *et al.*, J Fluid Mech. 517, 199 (2004)

**Looks identical!**

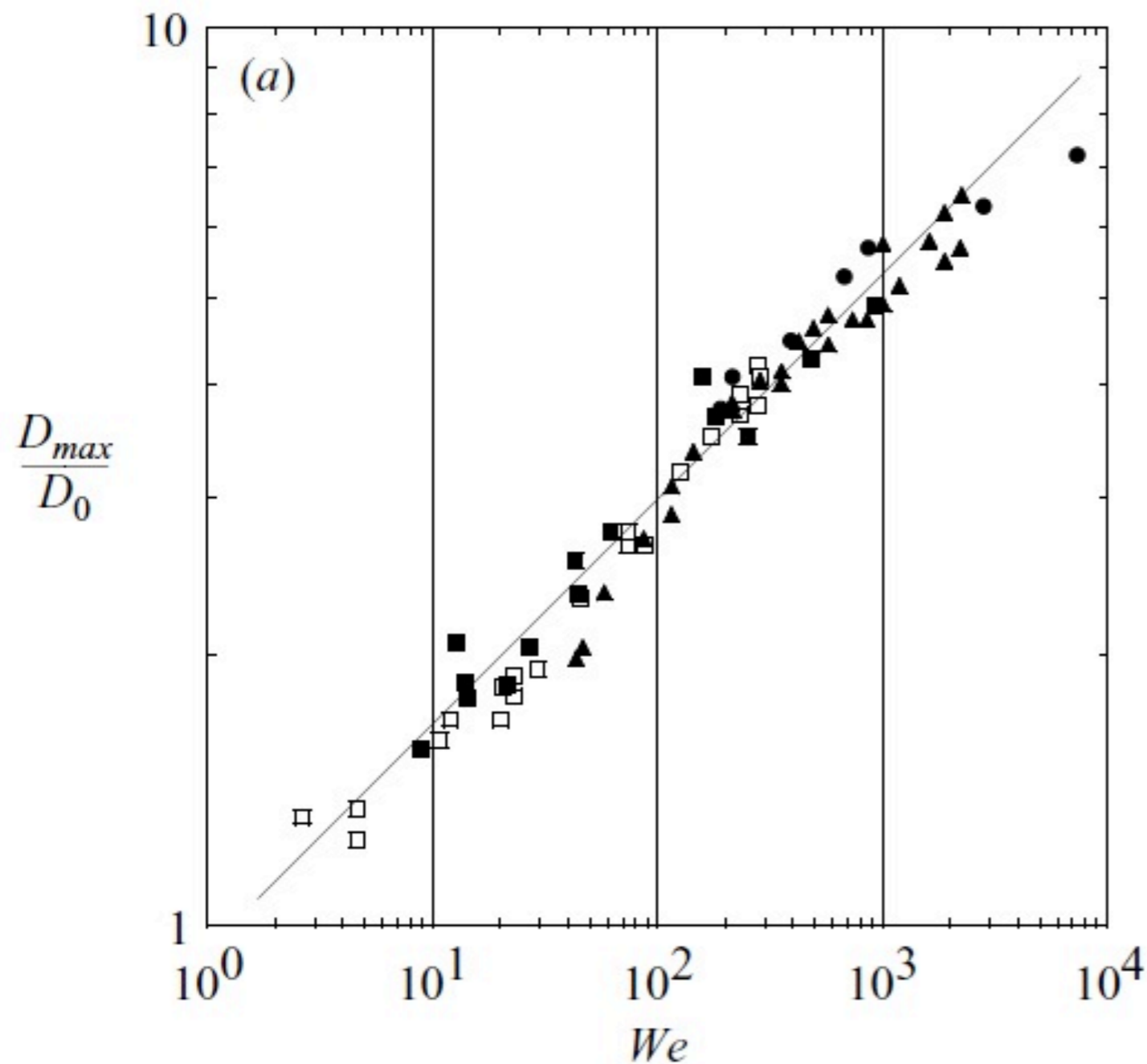
**What is the spreading of  
impacting droplets on  
standard and on  
superheated surfaces?**

# Maximal deformation of droplets at impact



$$D_0^3 \sim h D_{max}^2$$

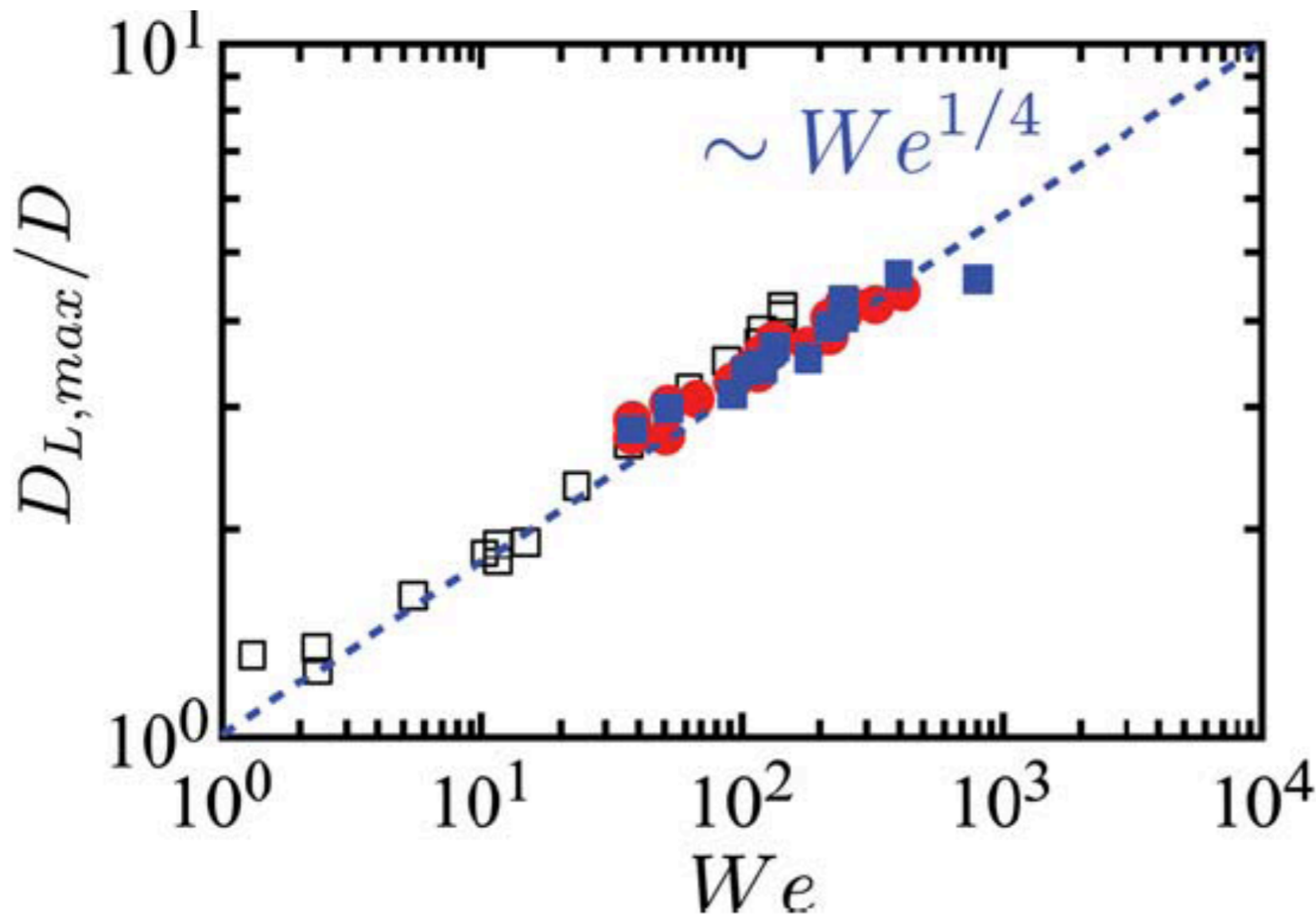
$$D_{max}/D_0 \sim We^{1/4}$$



$$F_{cap} \sim F_{inertial}$$

$$We = \frac{\rho D_o U^2}{\gamma}$$

# 1/4-scaling extremely robust

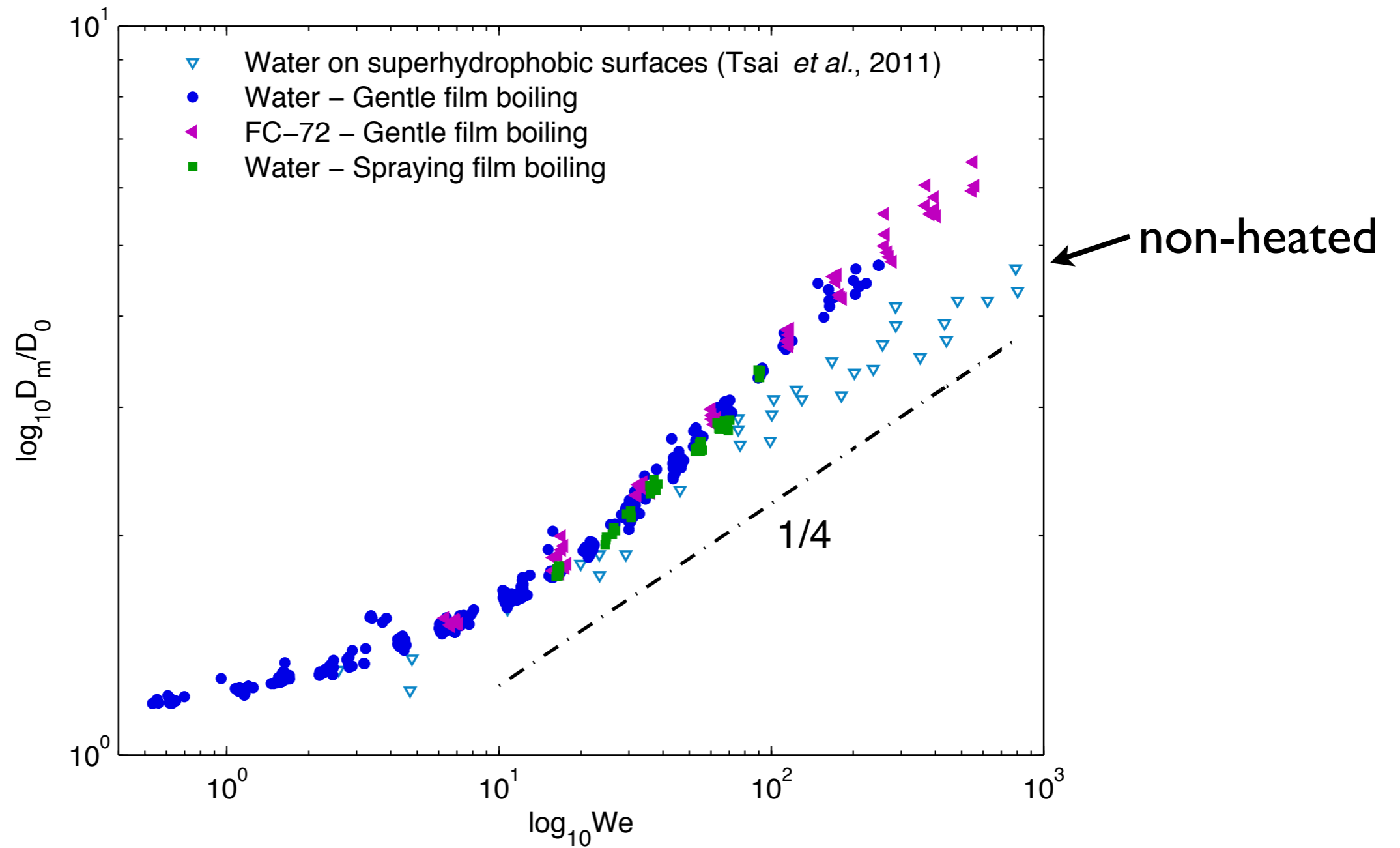


Impact on micro-structured surfaces

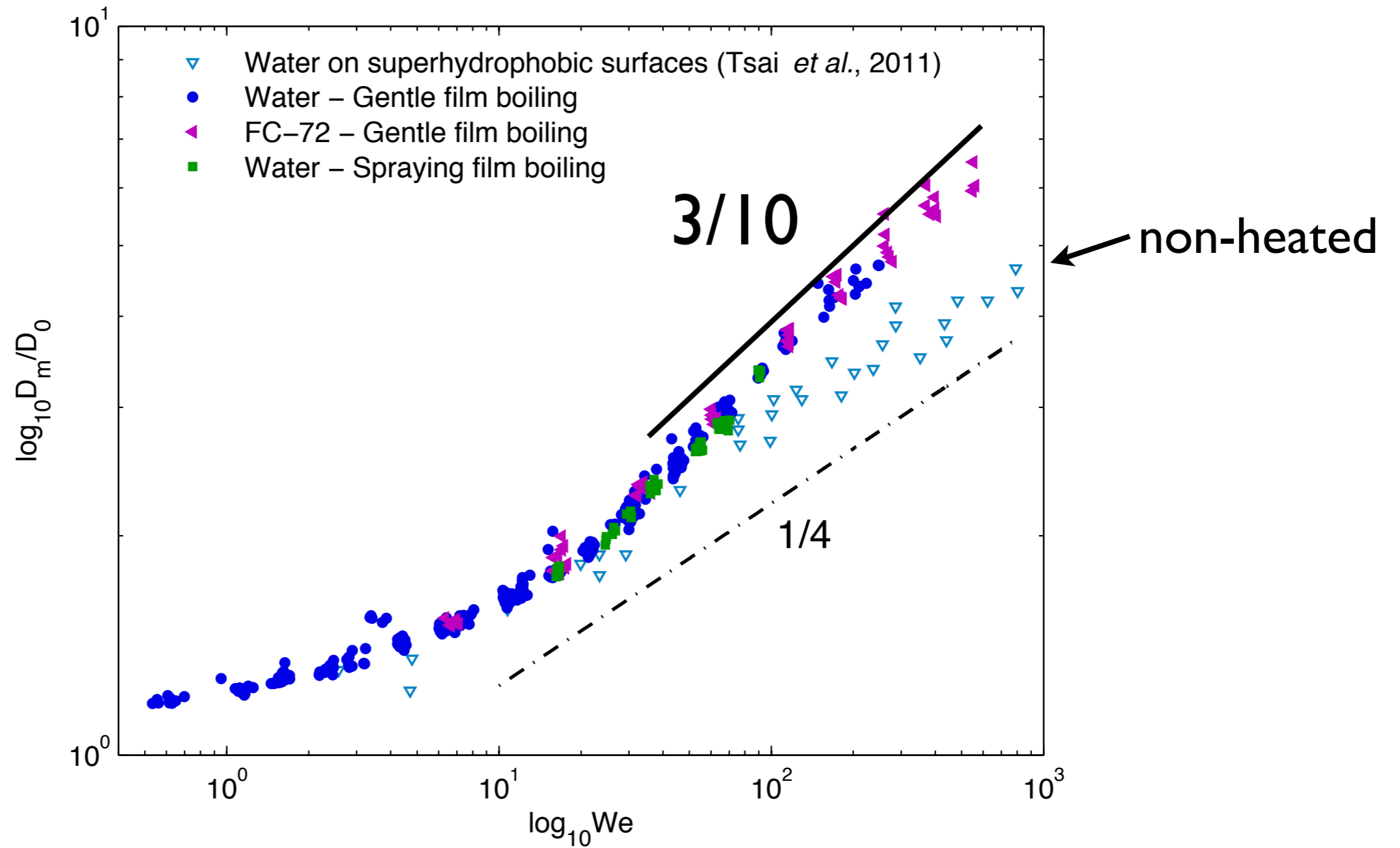
Scaling also independent of contact angle  
→ gas layer!



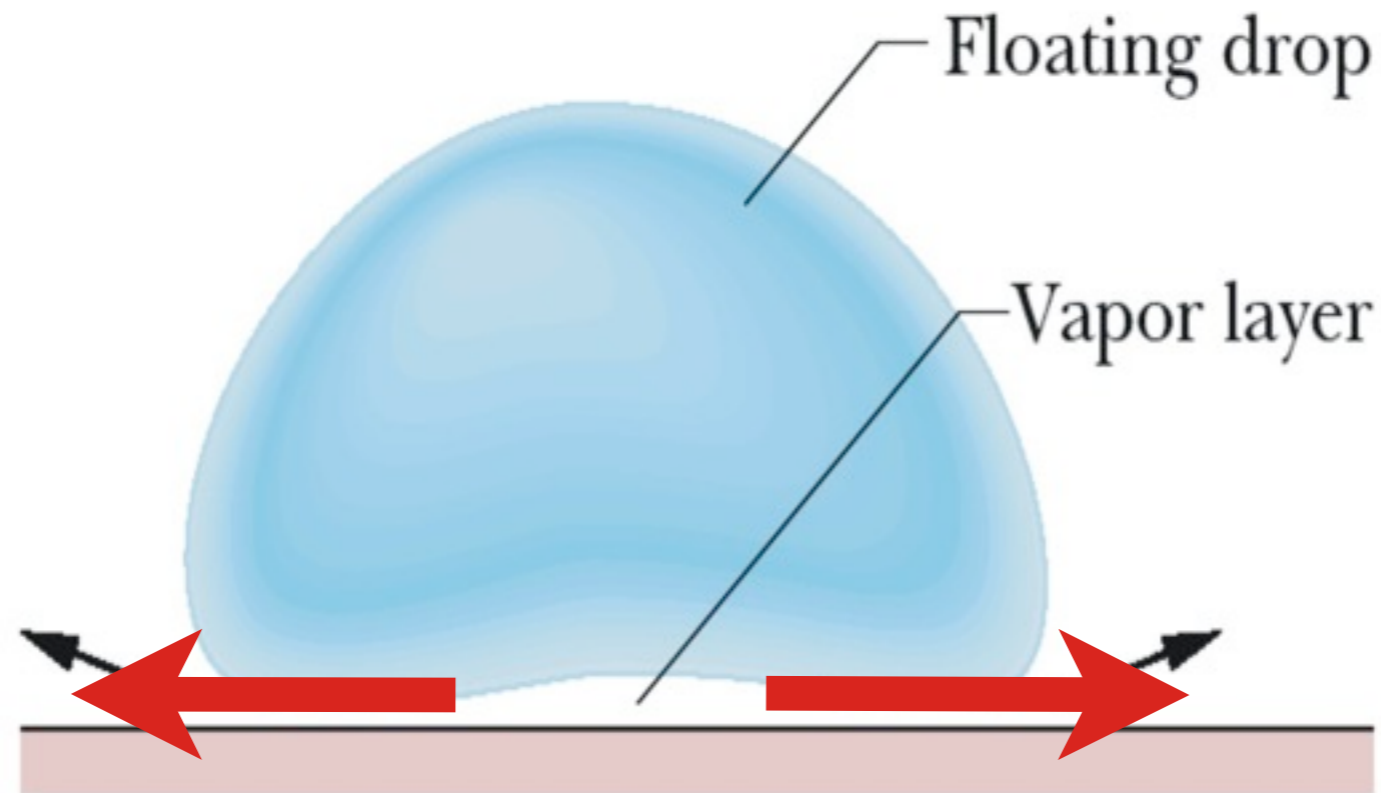
# Maximum spreading in film boiling regime



# Maximum spreading in gentle & spraying film boiling regimes

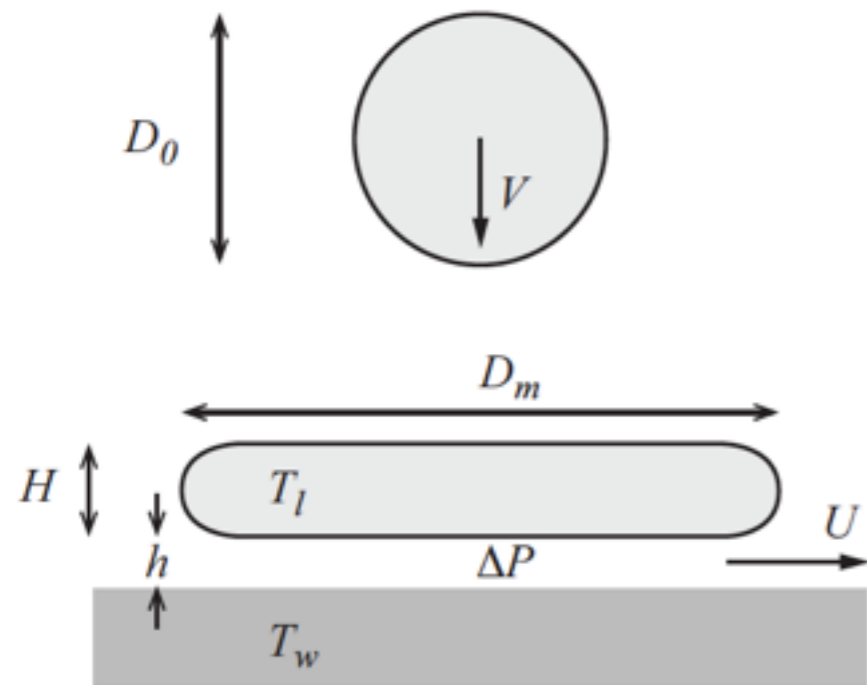


# Physical mechanism of enhanced spreading



- Vapor shoots out and drags liquid along!
- The more, the larger  $U$ , as then more liquid evaporates

# Derivation of the 3/10 scaling law for impact on superheated surfaces



$$\Delta P \sim \rho V^2$$

1. viscous driving force vs capillarity

$$\frac{\gamma}{H} \sim \mu_v \frac{U}{h}$$

2. Pressure buildup drives viscous vapor flow

$$\frac{\Delta P}{D_m} \sim \mu_v \frac{U}{h^2}$$

3. mass evaporation rate  $\sim$  flow rate

$$\frac{k\Delta T}{Lh} D_m^2 \sim \frac{\rho_v}{\mu_v} h^3 \Delta P.$$

# Non-dimensionalize

$$\Gamma = D_m/D_0$$

$$\tilde{U} = U/V$$

$$\tilde{h} = h/D_0$$

$$We = \frac{\rho_l D_0 V^2}{\gamma}$$

$$St = \frac{\rho_l D_0 V}{\mu_v}$$

$$Pe = \frac{V D_0 L \rho_v}{k \Delta T}$$

# Resulting scaling laws

$$\Gamma \sim \frac{We^{2/5}}{St^{1/10} Pe^{1/10}}$$

$$\Gamma \sim We^{3/10}$$

$$\tilde{h} \sim \frac{We^{1/5}}{St^{3/10} Pe^{3/10}}$$

$$\tilde{h} \sim We^{-1/10}$$

$$\tilde{U} \sim \frac{St^{1/2}}{Pe^{1/2}}$$

$$\tilde{U} \sim We^0$$

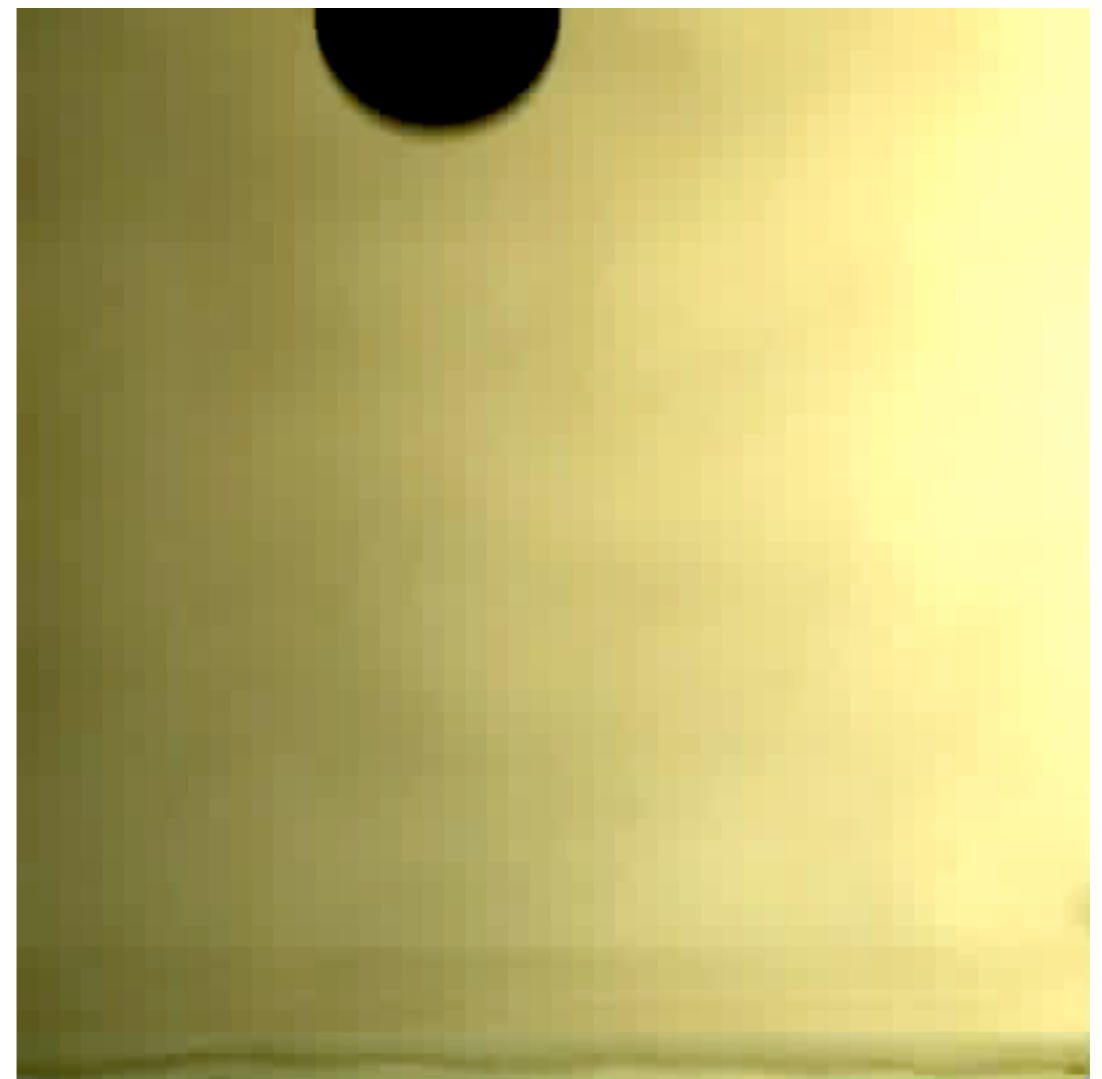
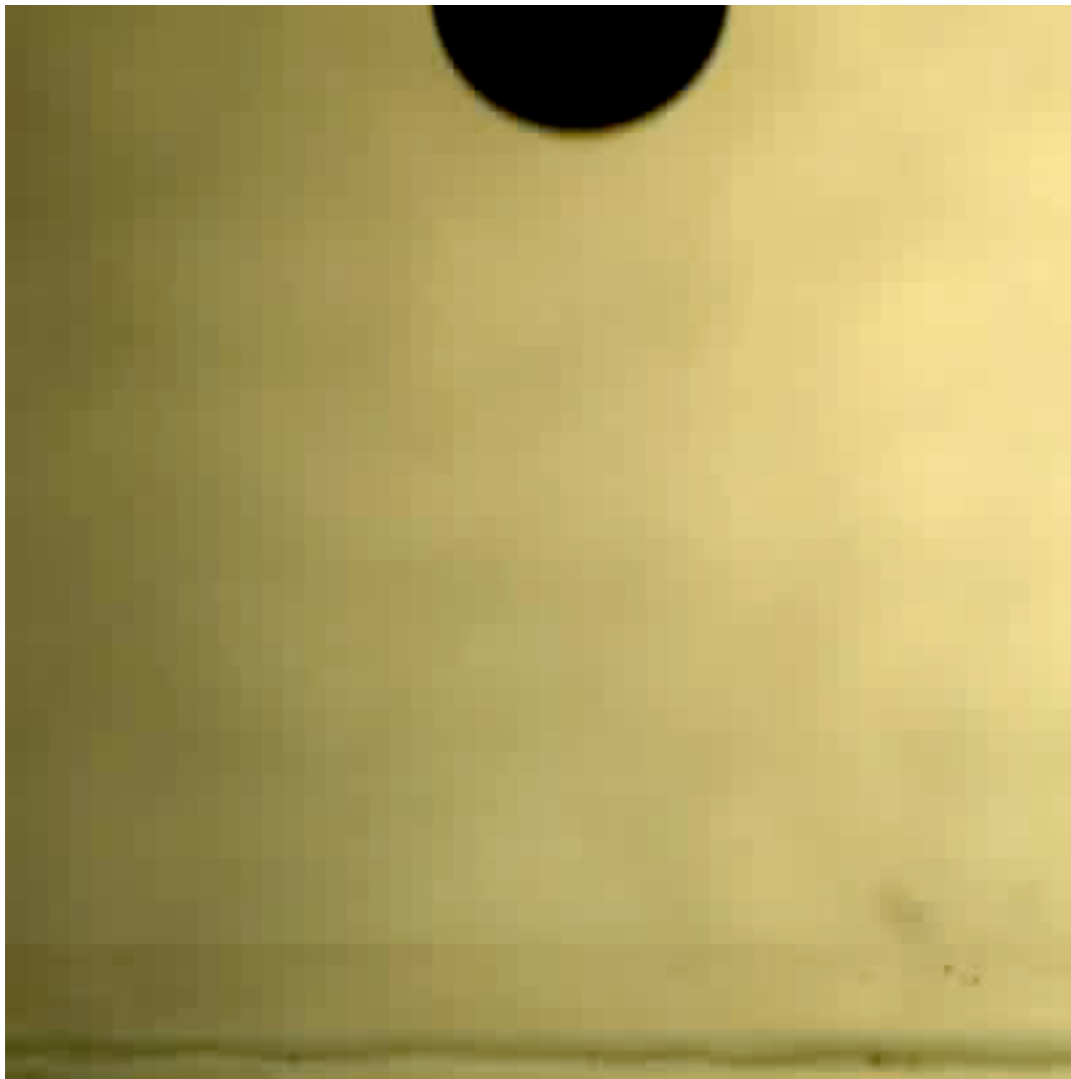
**Predictions!**

... and when all velocity dependences are put into We:

# Impact of suspension droplets

e.g.: paint (pigments!)

# Impact of 39%-suspension droplet



shear-thickening

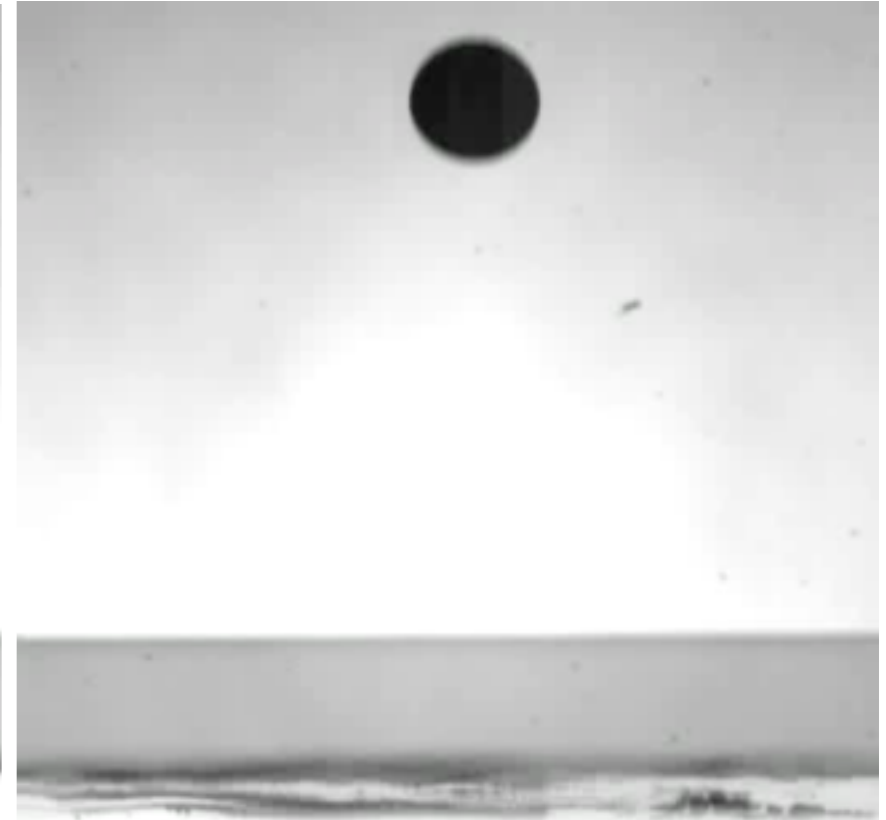
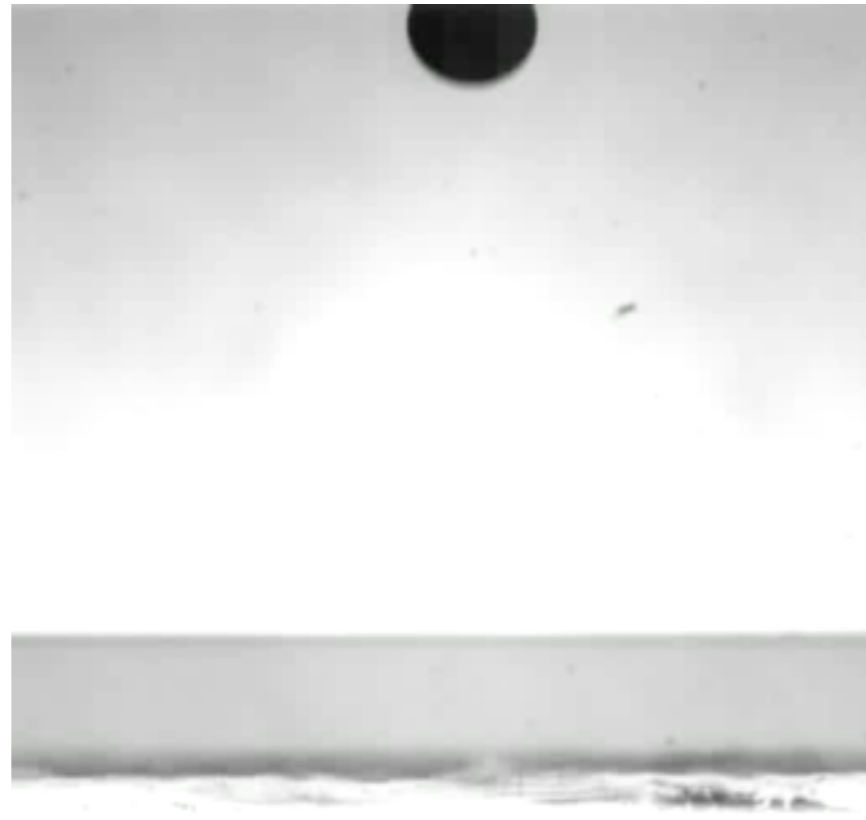
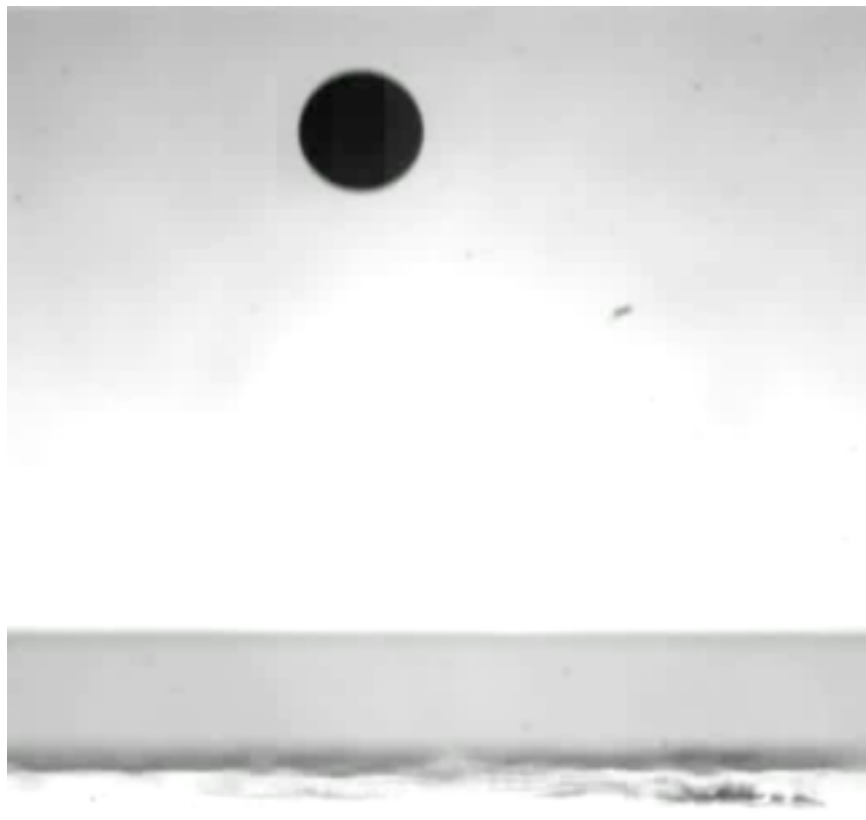


# Is there a splash at impact?

From spreading....

to splashing...

and spreading again!

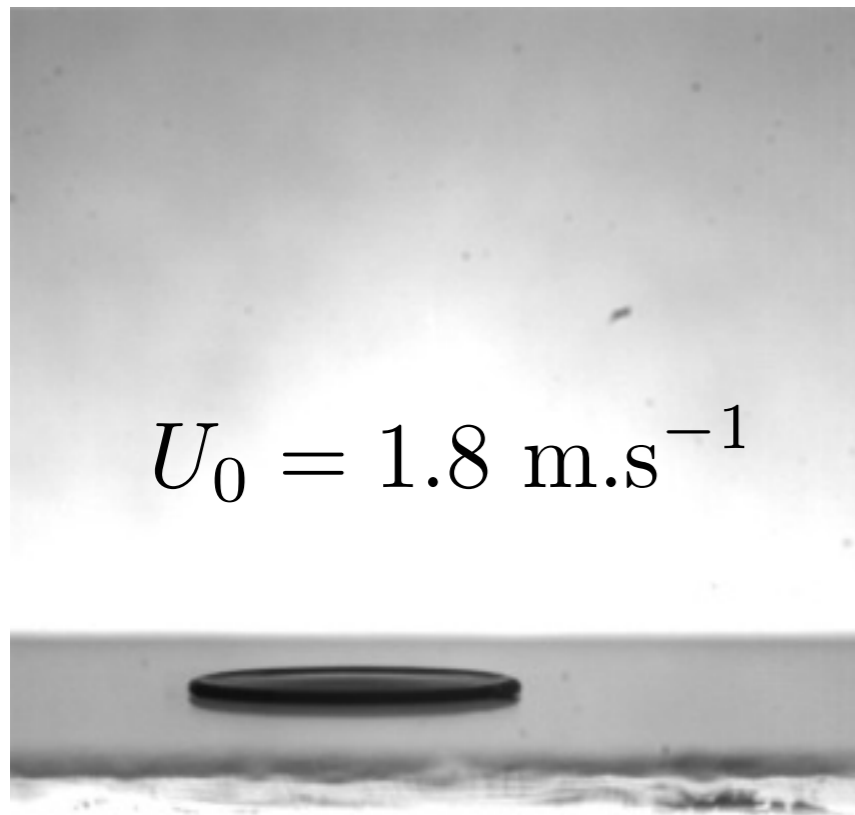


Increasing impact speed

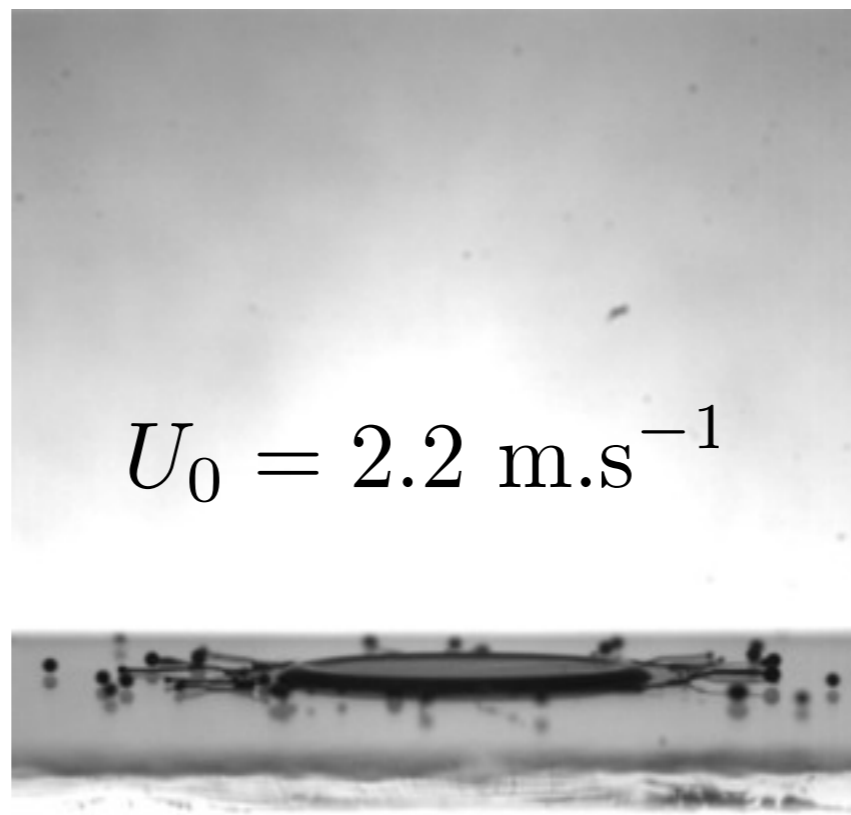


# Is there a splash at impact?

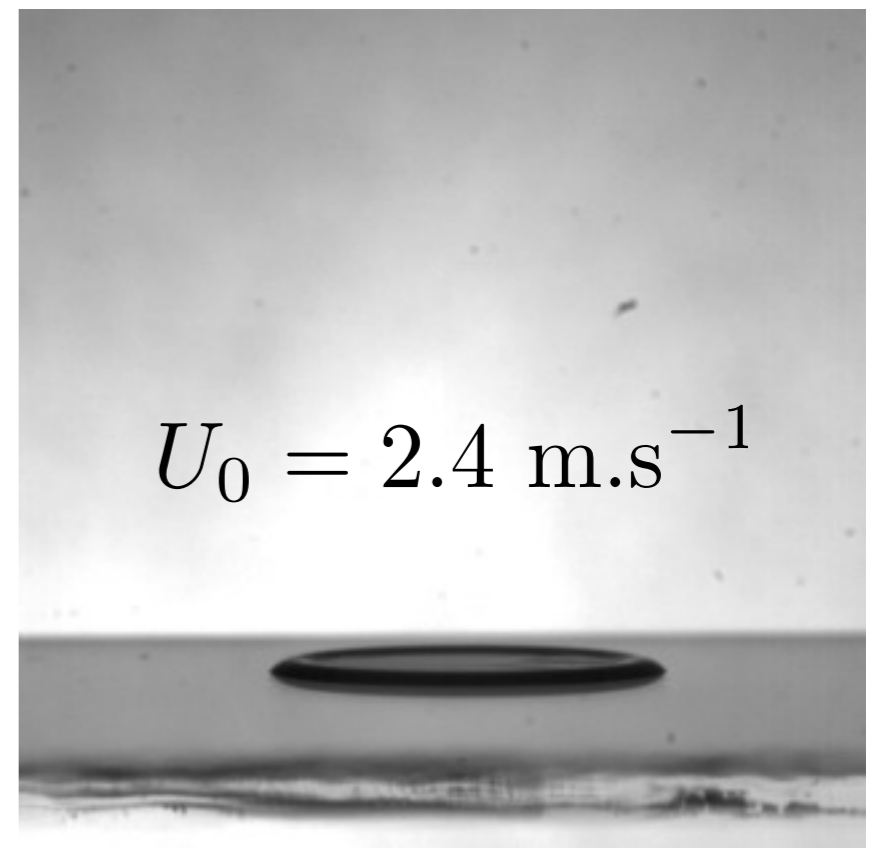
spreading



splashing



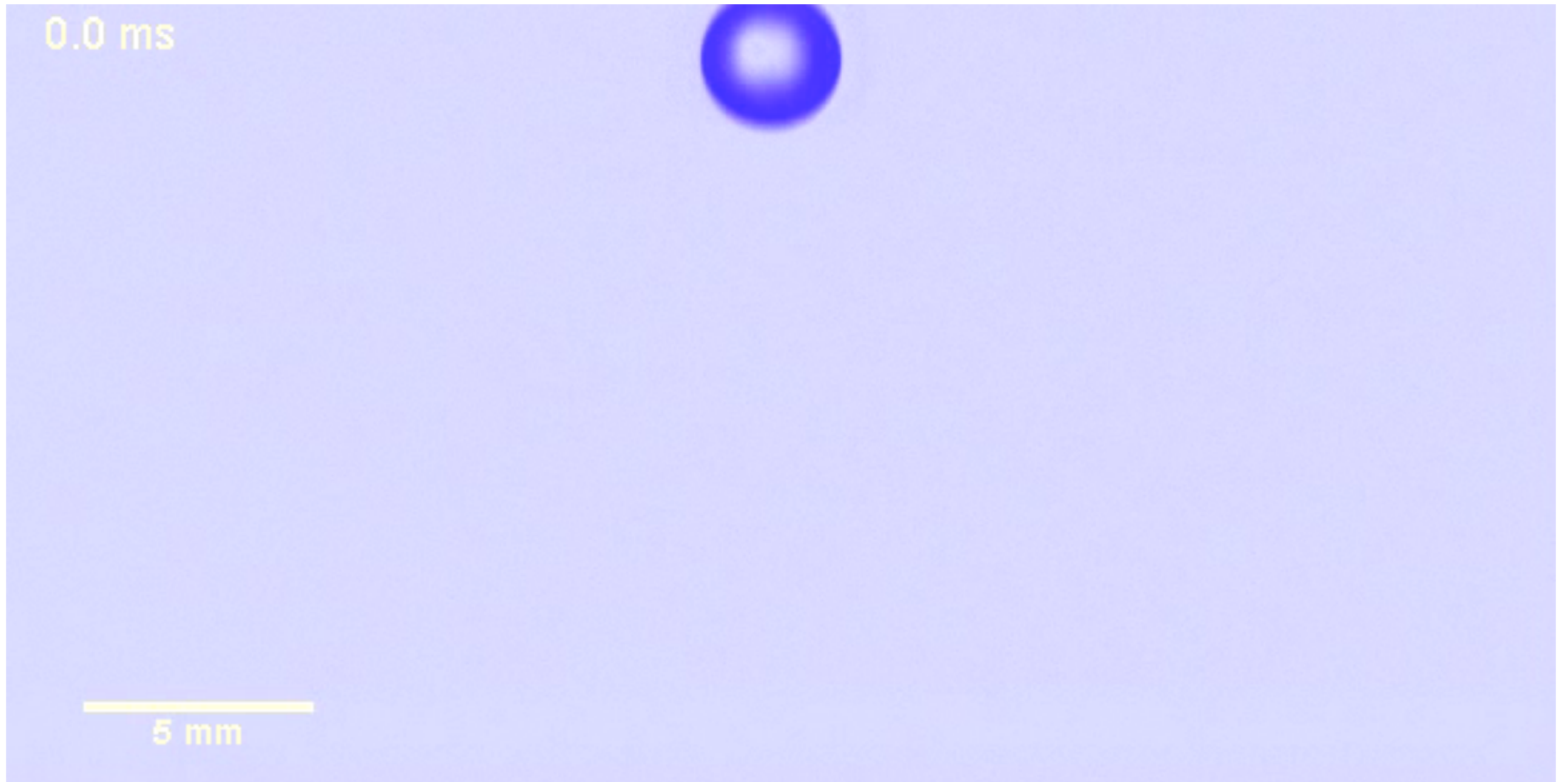
spreading



Increasing impact speed

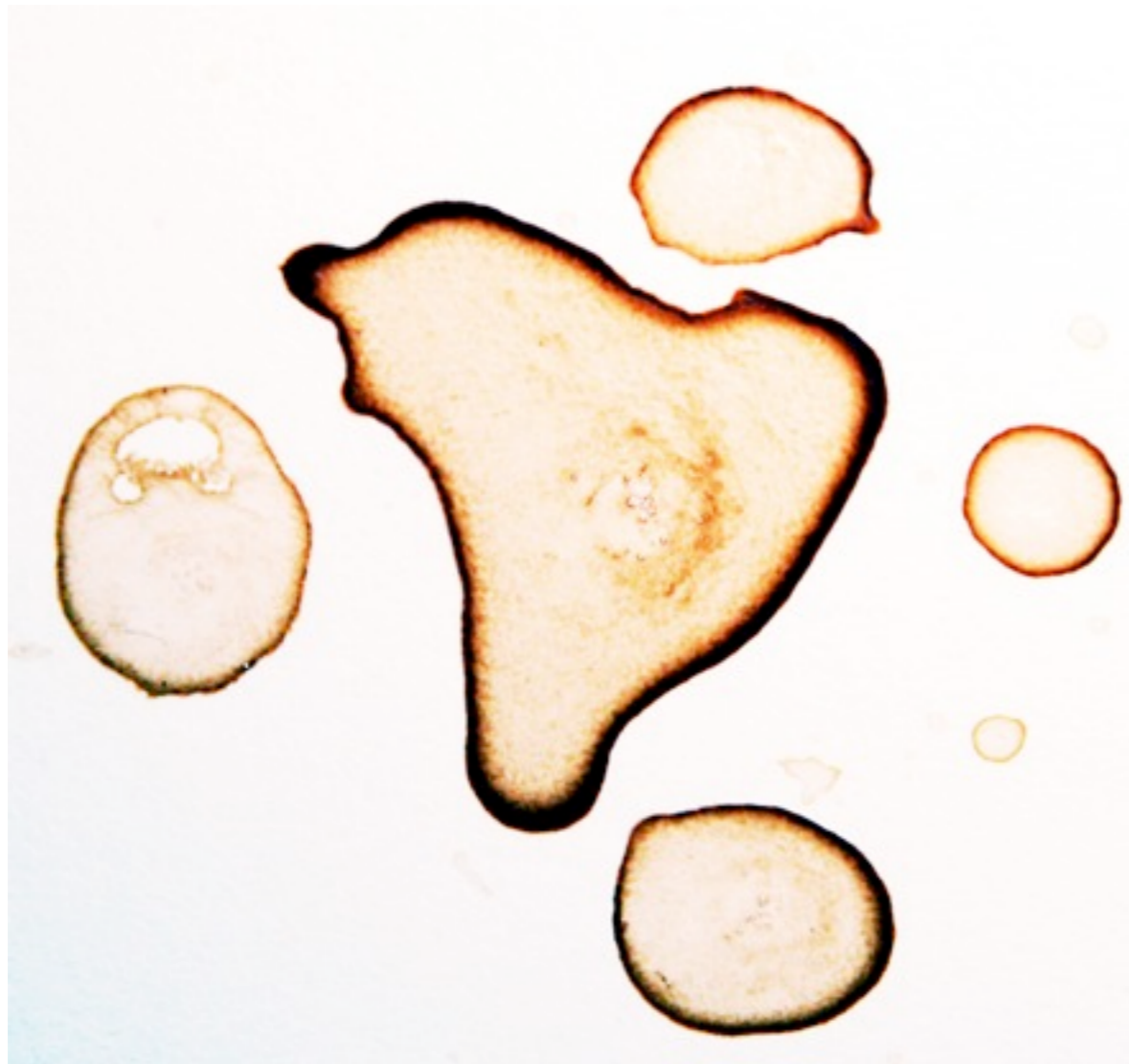
shear-thickening!

# Impact of shear-thinning liquid



# **How do suspension droplets dry?**

# How do such droplets dry?



“coffee-stain  
problem”

Marin, Gelderblom, Lohse, Snoeijer,  
Phys. Rev. Lett. 107, 085502 (2011)

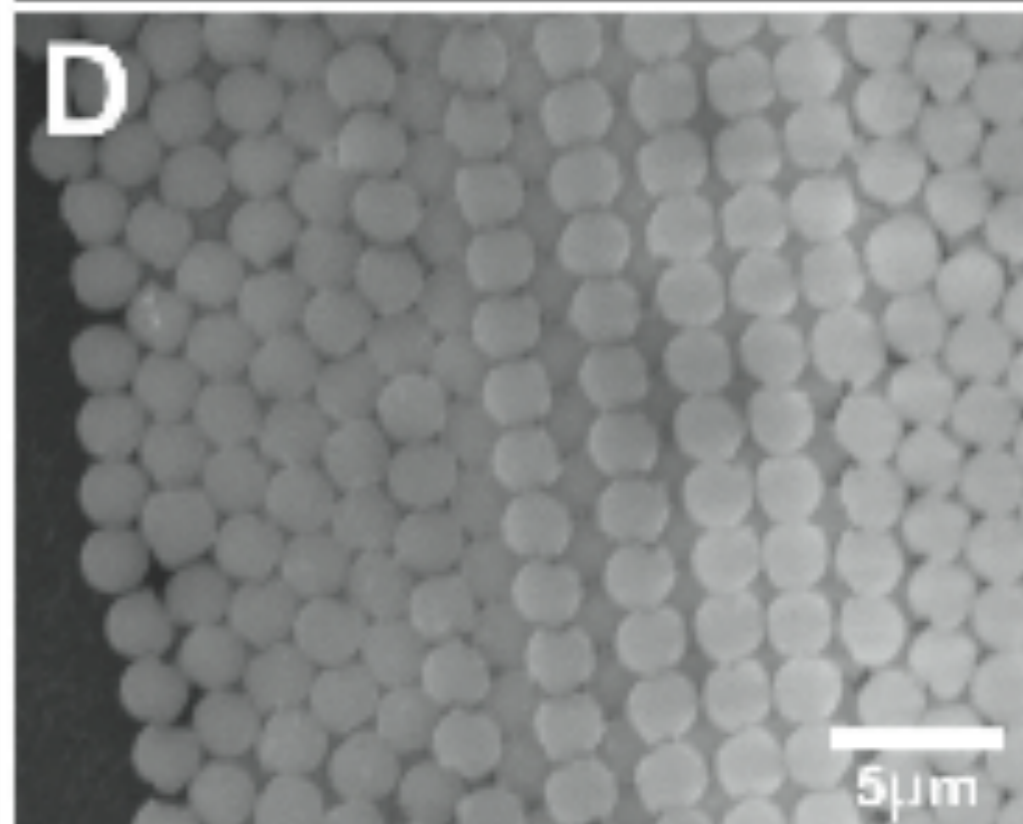
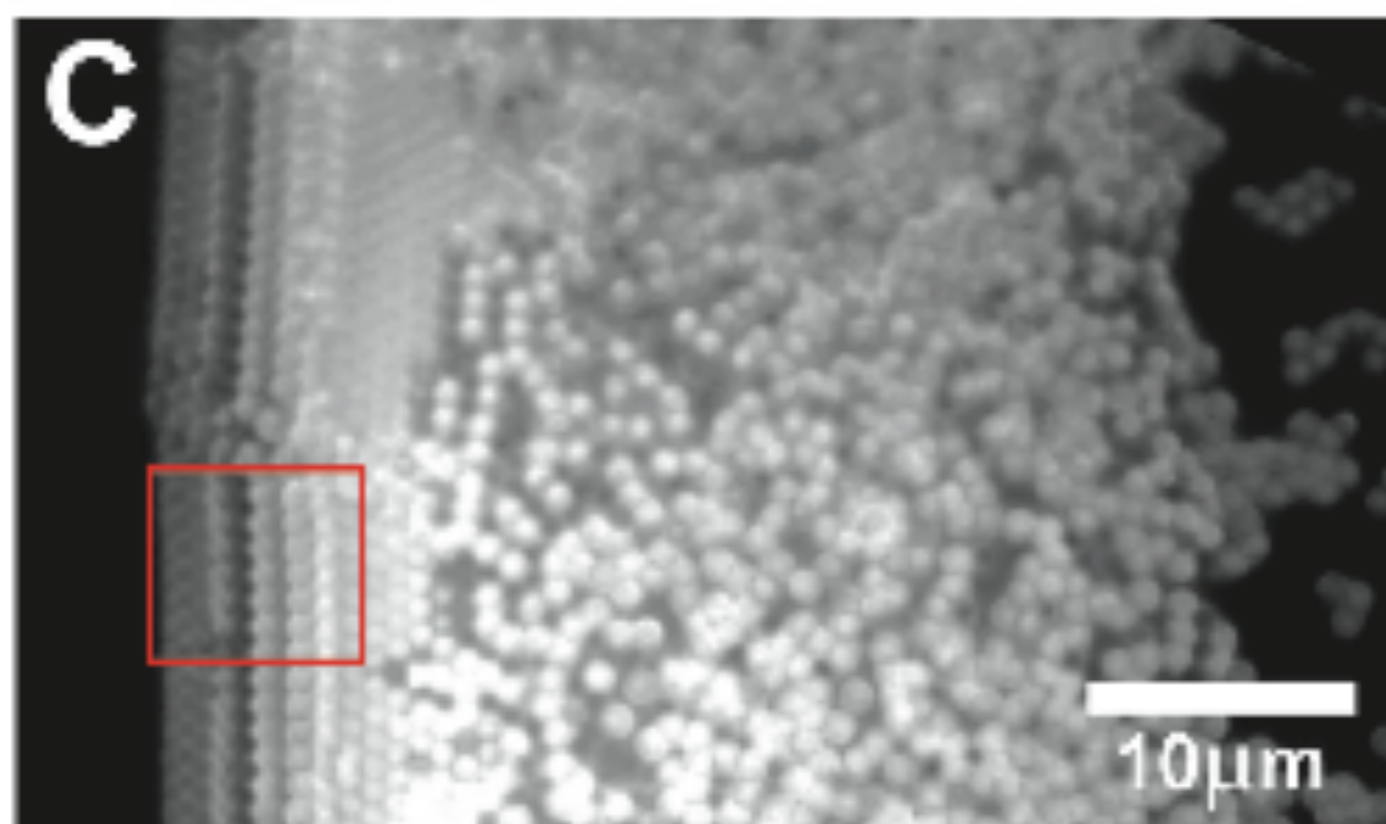
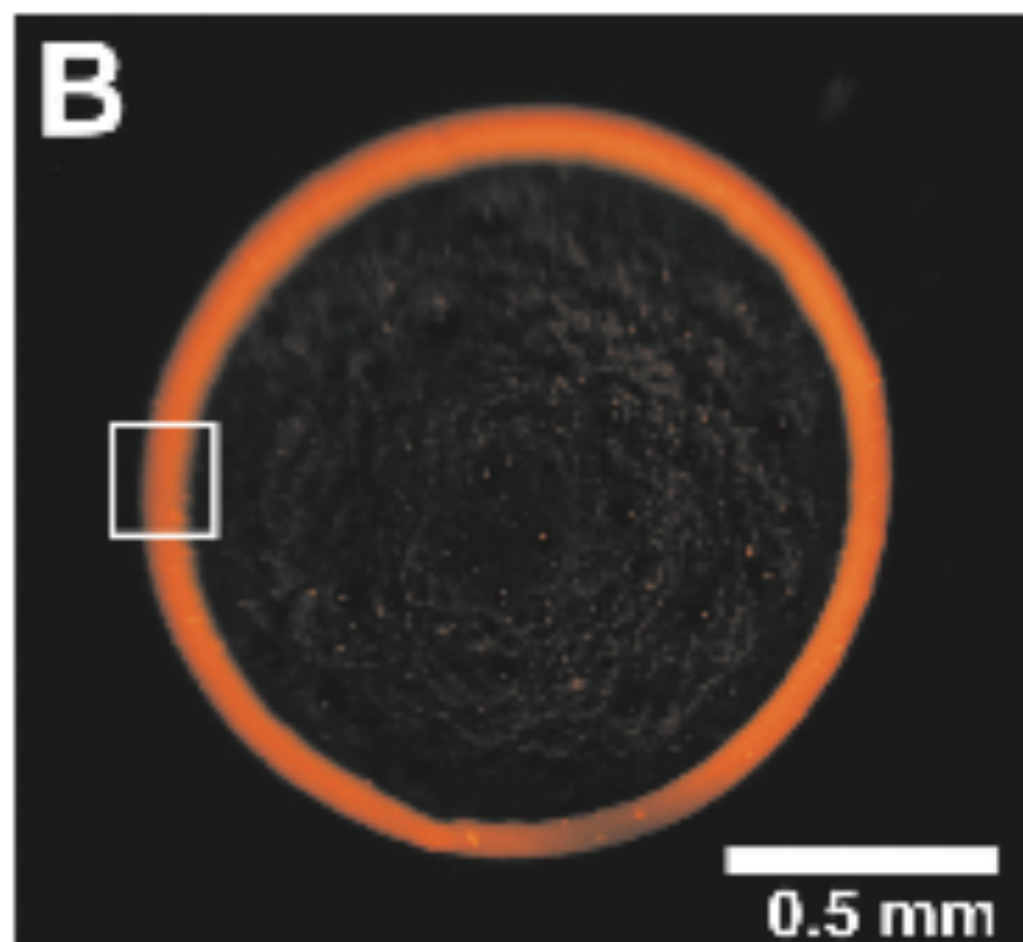
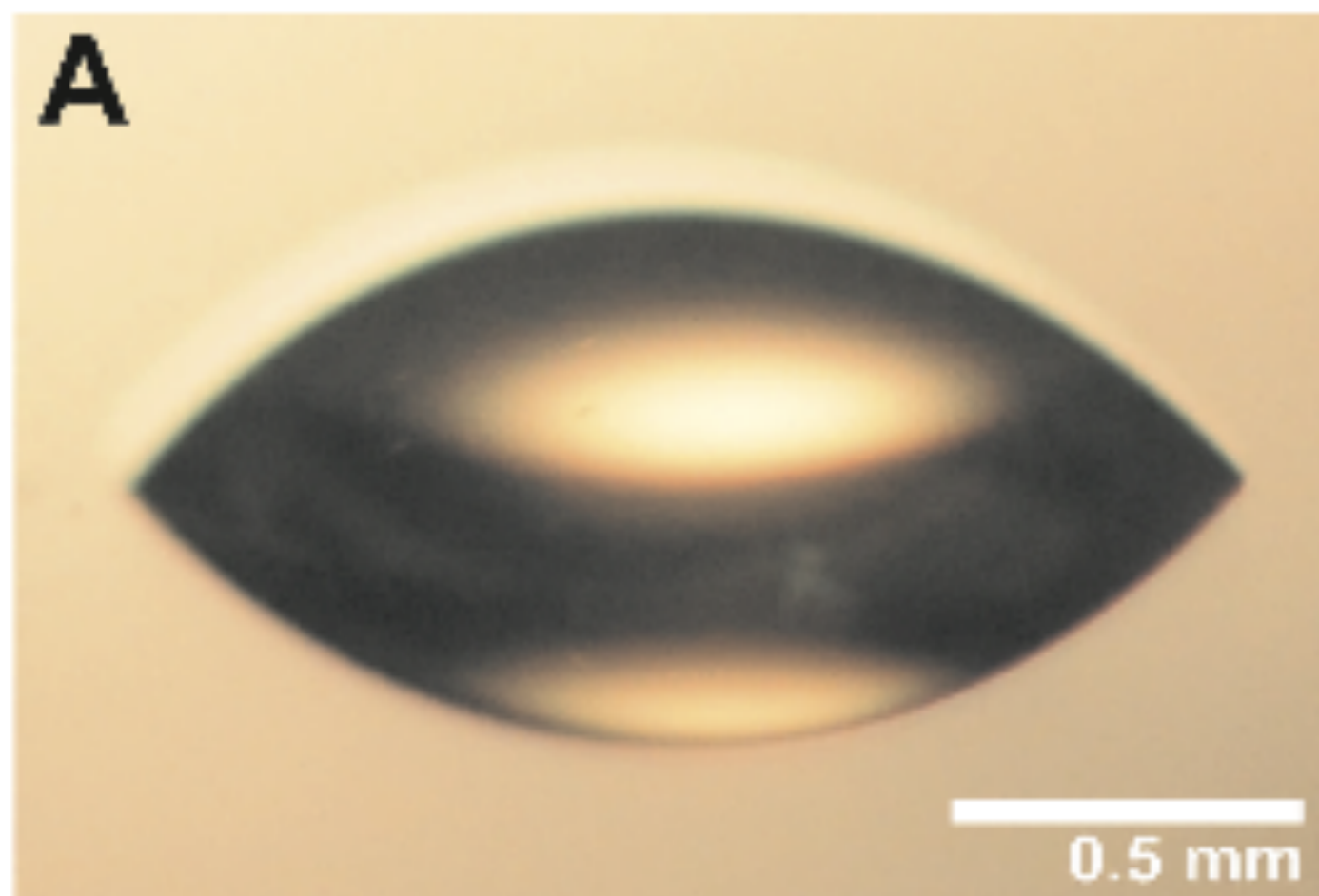
# Avalanche of Particles in Evaporating Drops

A. G. Marin, H. Gelderblom,

J. Snoeijer, D. Lohse

Physics of Fluids, Univ. Twente

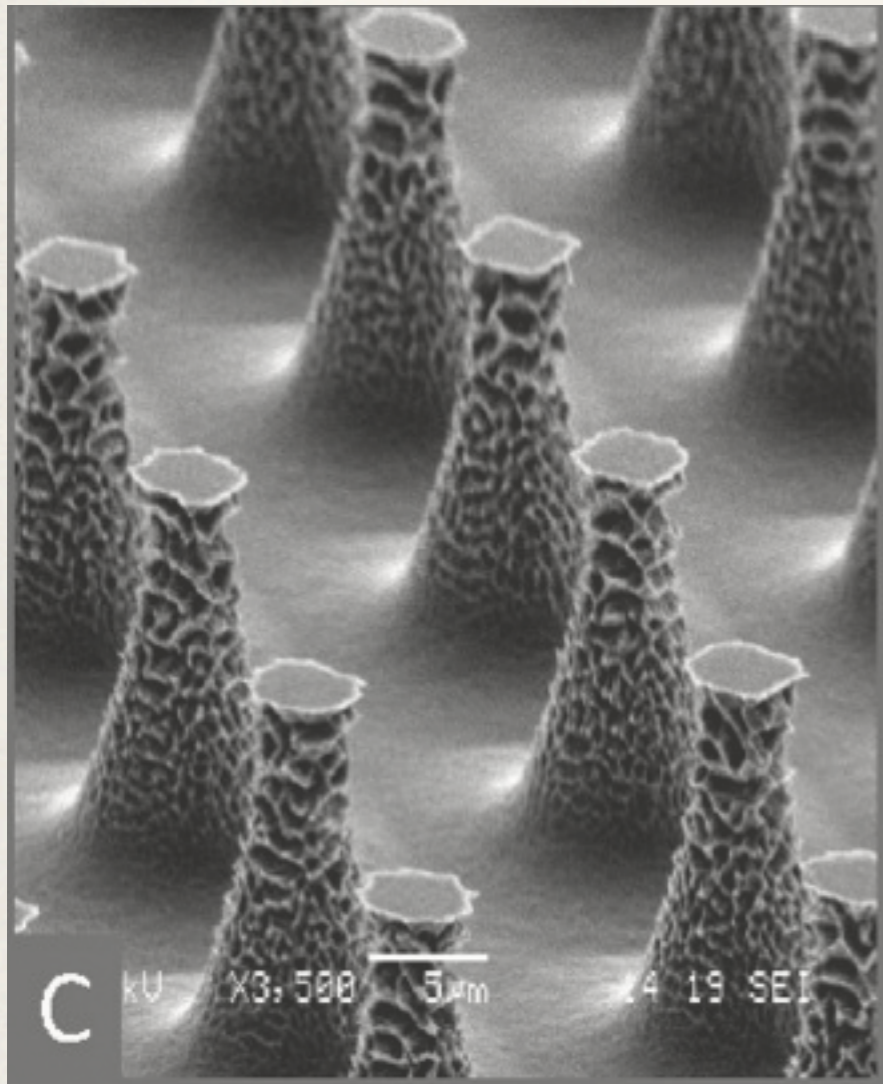






# Microstructured substrates

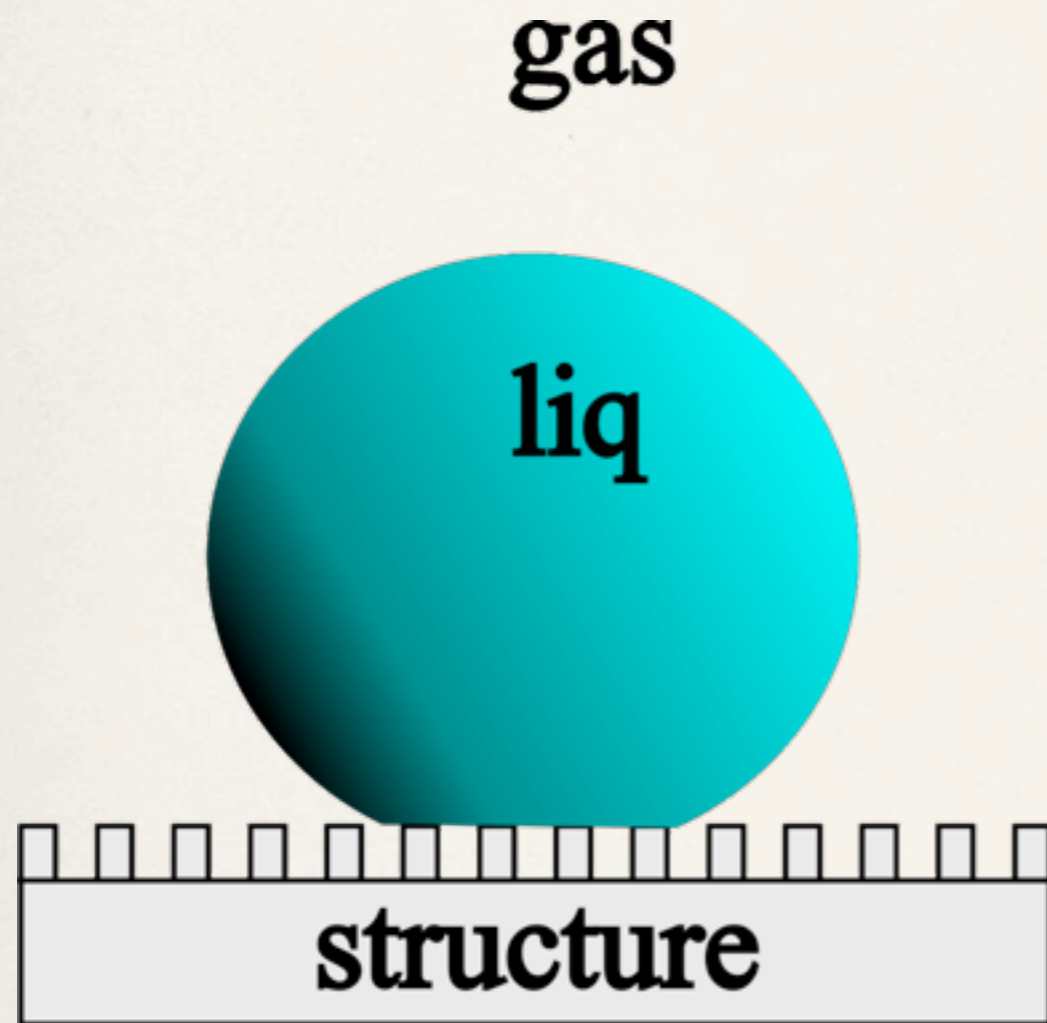
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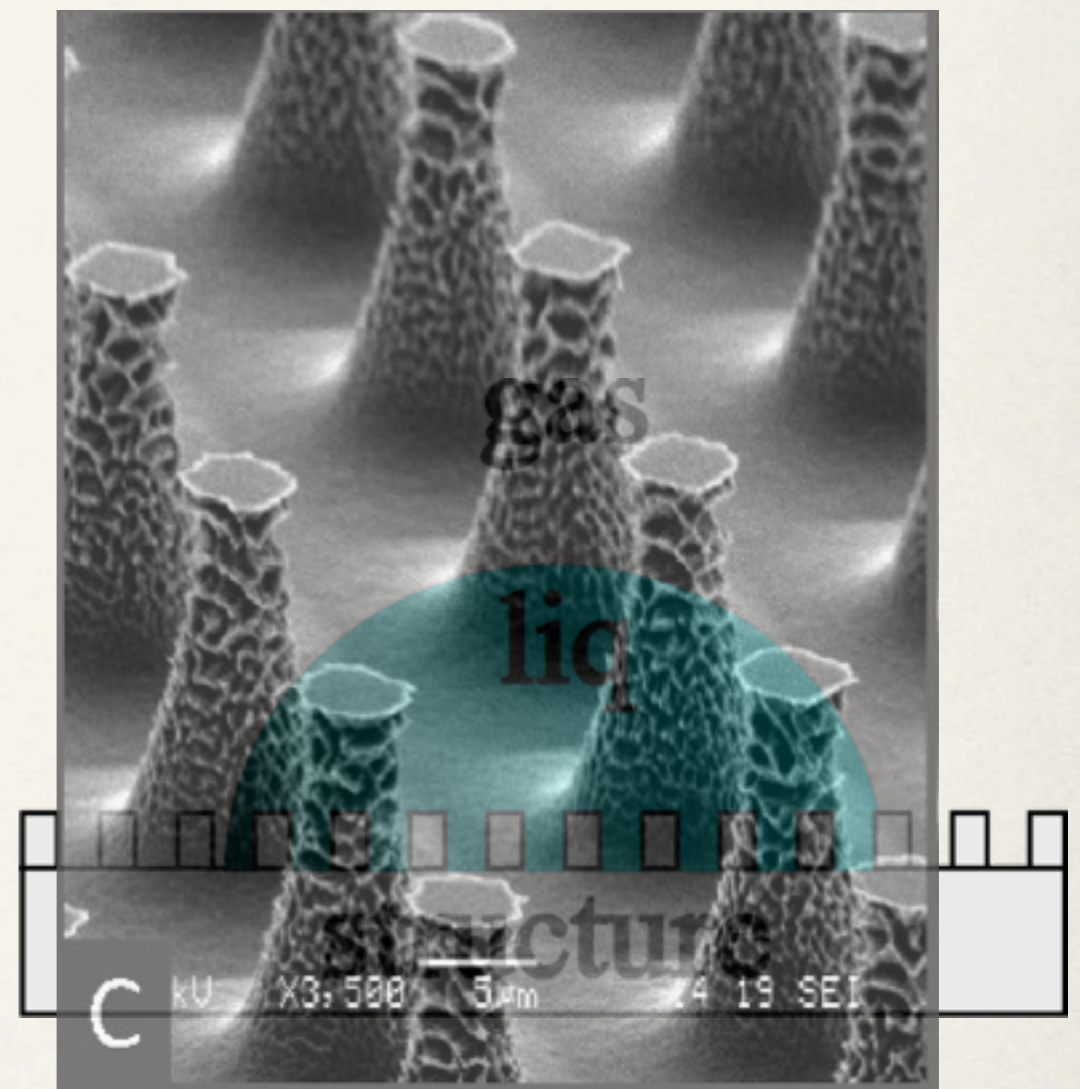
*Absence of an evaporation-driven wetting transition on omniphobic surfaces,*  
Susarrey-Arce, Marín, Nair, Lefferts, Gardeniers, Lohse, van Houselt, *Soft Matter* 8 (2012)  
Susarrey-Arce et al. *J. Micromech. Microeng.* 23 (2013)



# Diehard superhydrophobic substrates

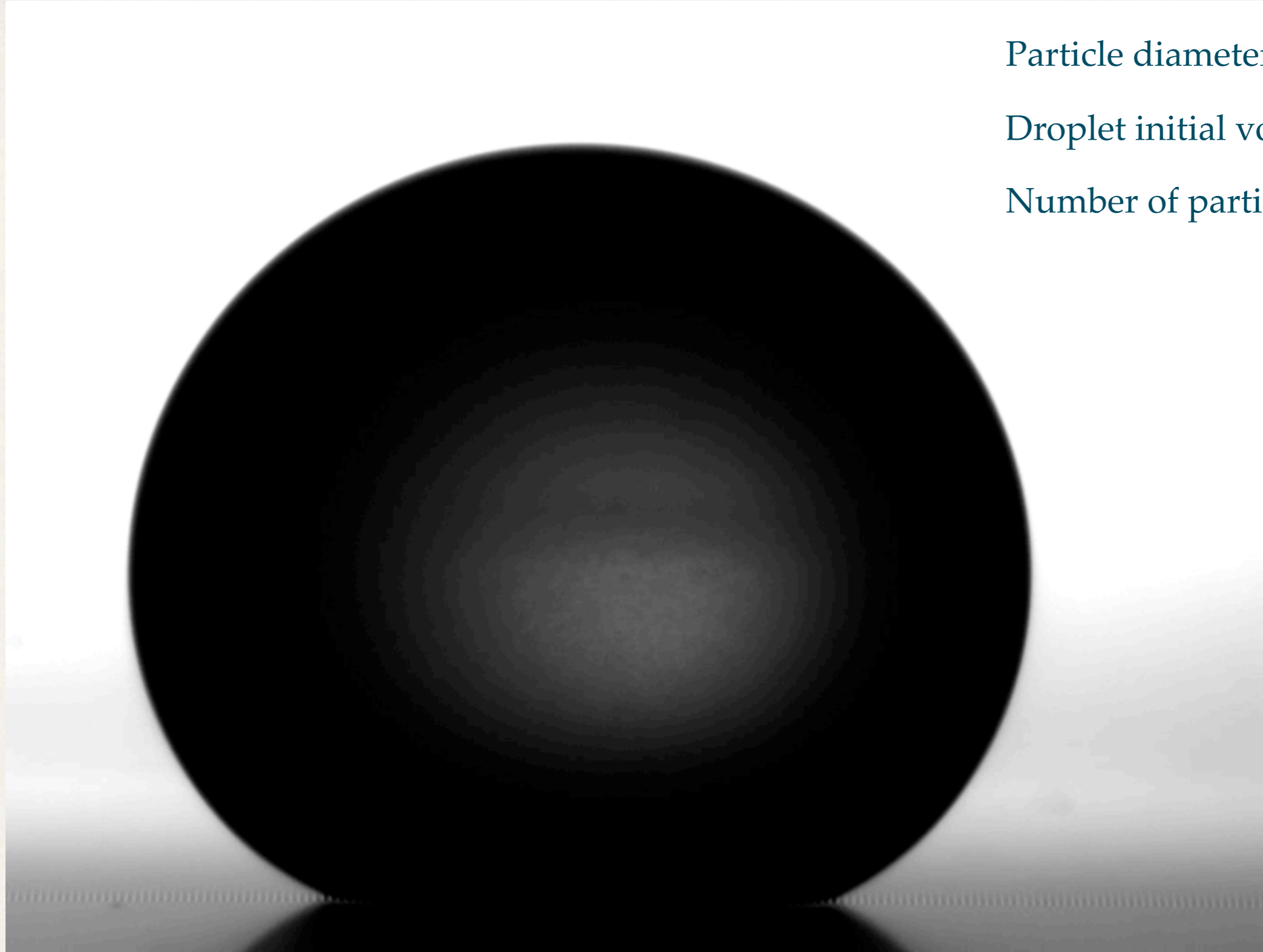


Cassie-Baxter or "fakir" state



Wenzel or "impaled" state

# Drying a colloidal suspension on a die-hard surface



Particle diameter  $1\mu\text{m}$

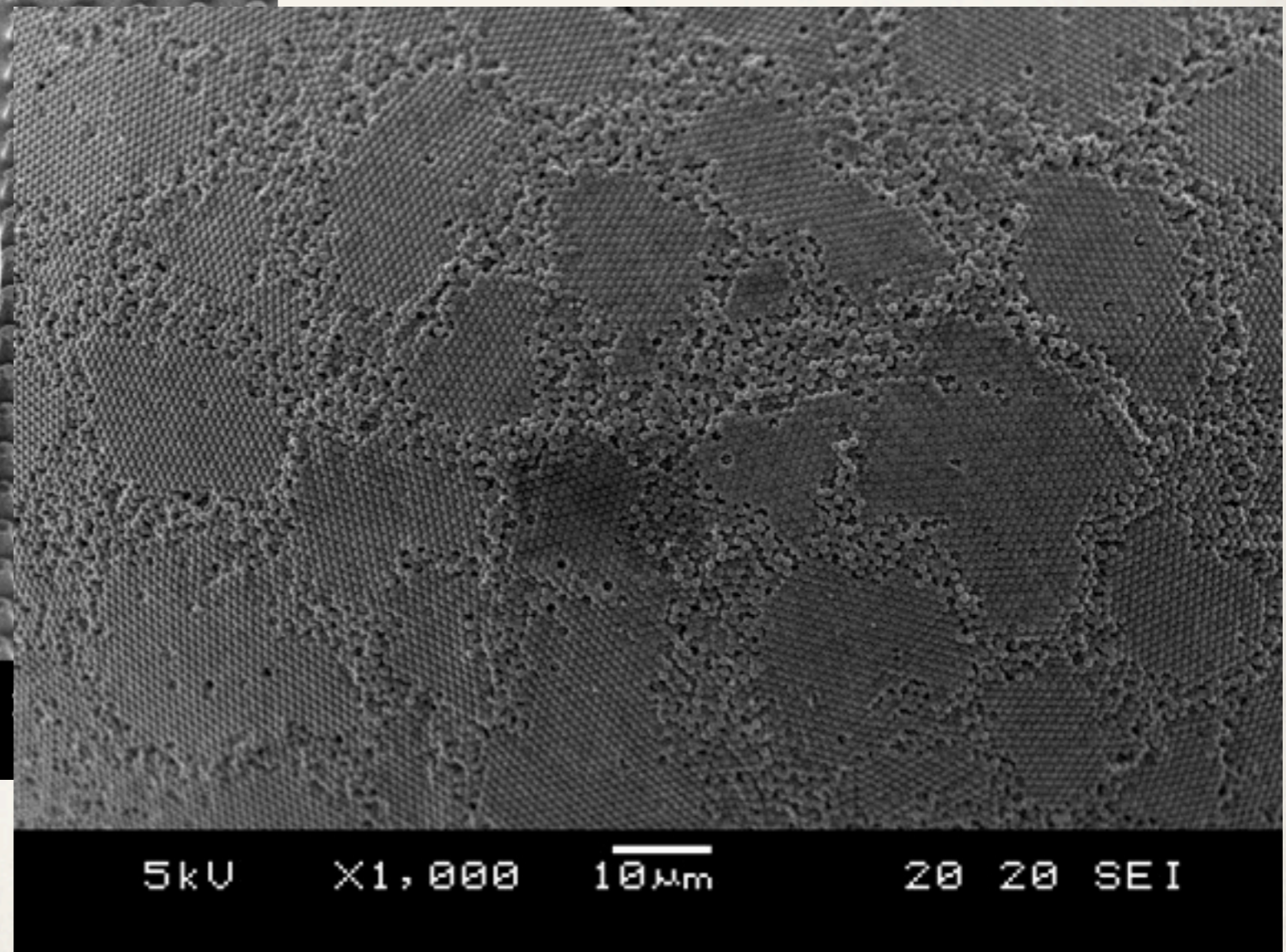
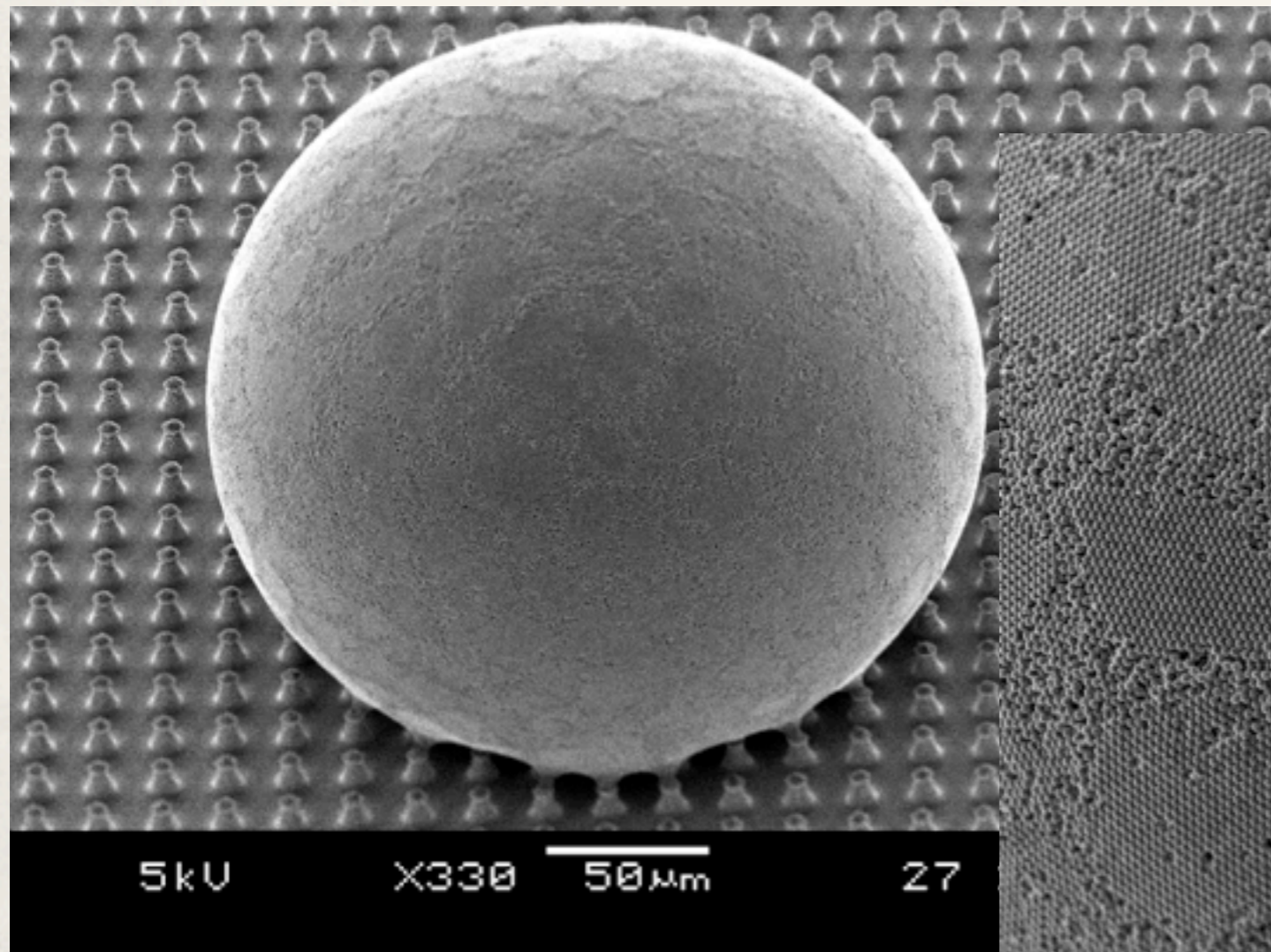
Droplet initial volume:  $10\mu\text{l}$

Number of particles  $\sim 10^7$



# Microscopic soccer balls!

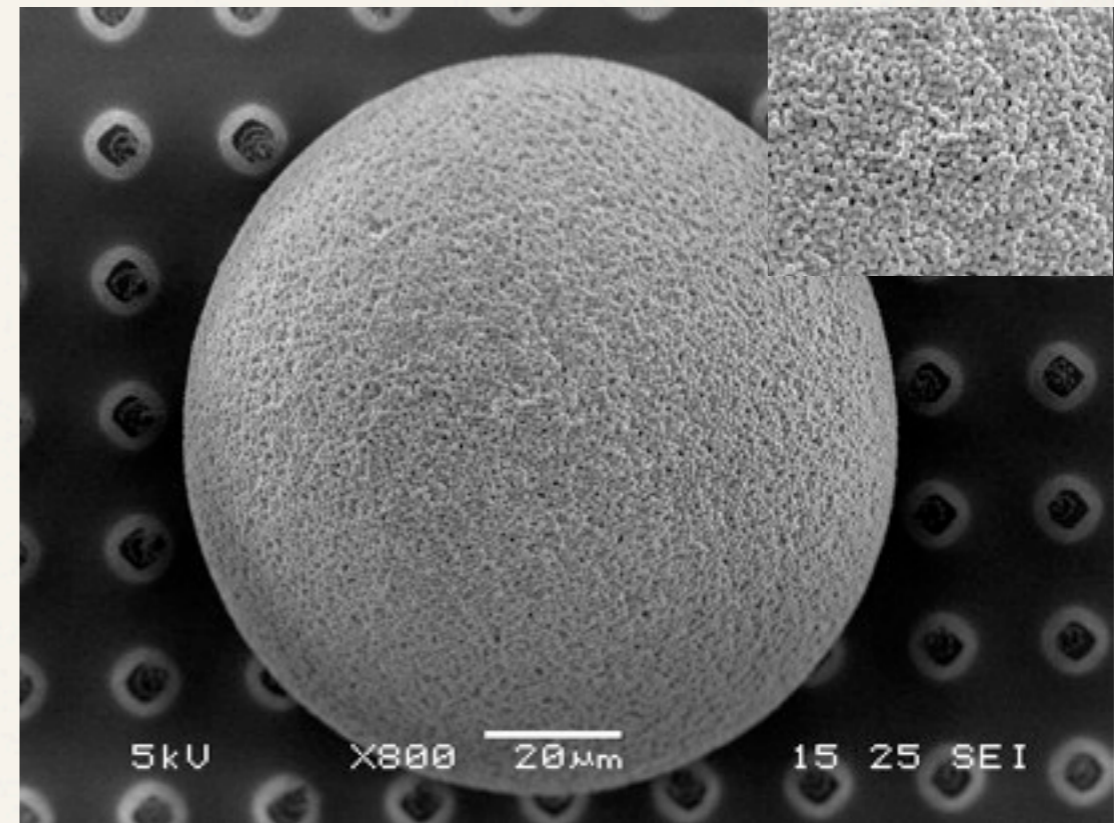
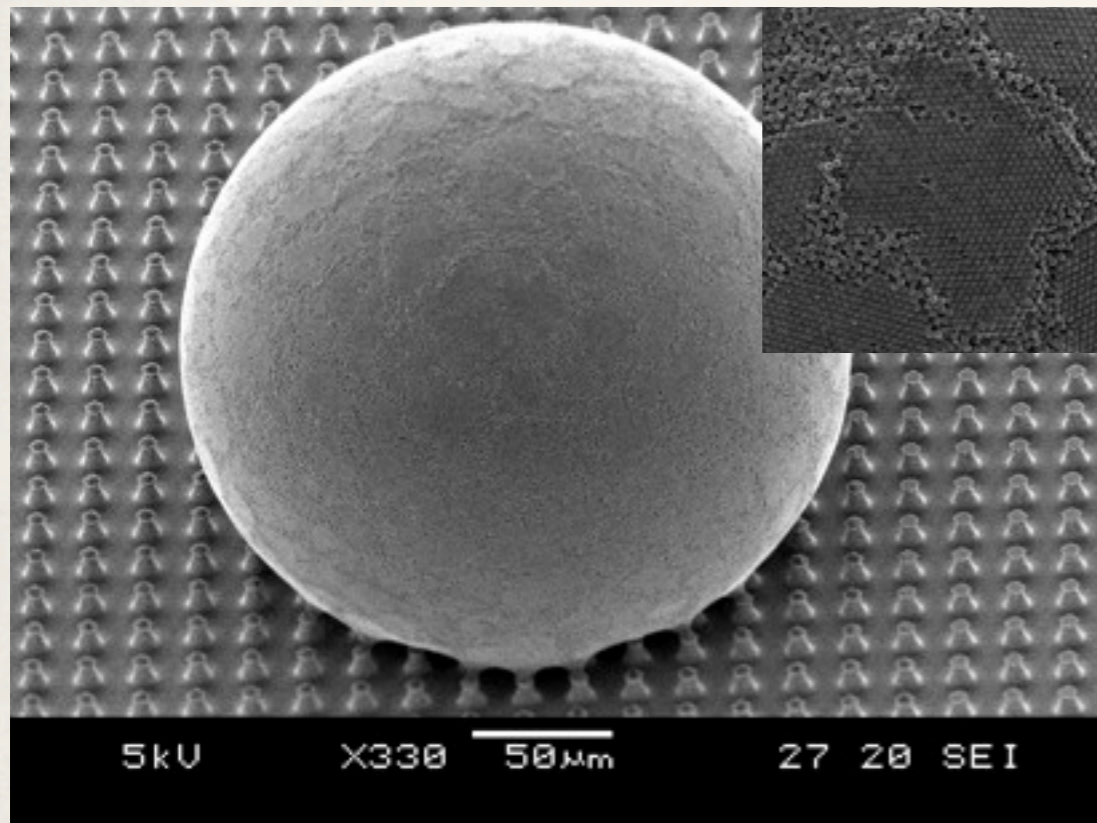
---



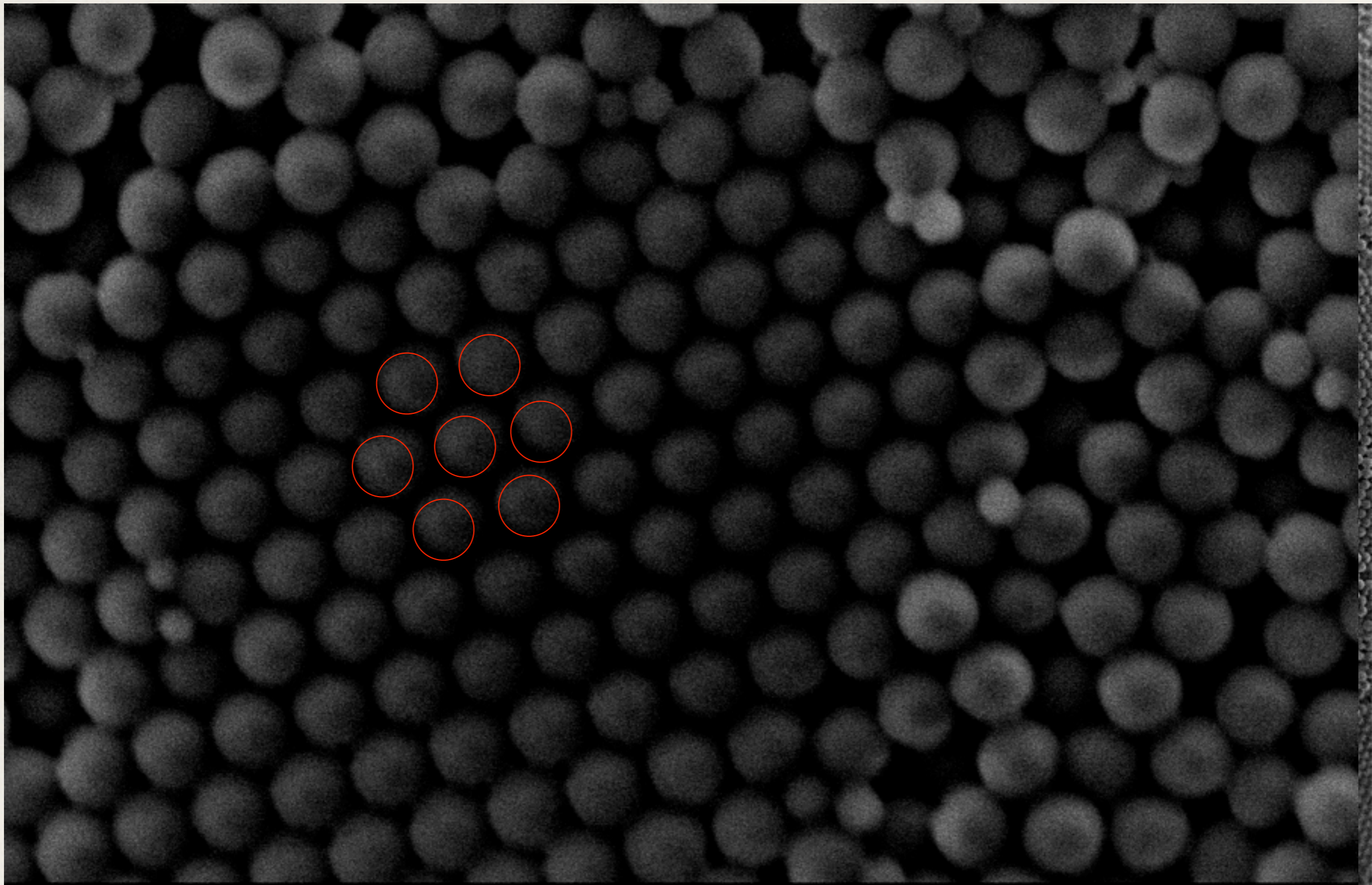


# Different structures?

---







5kV

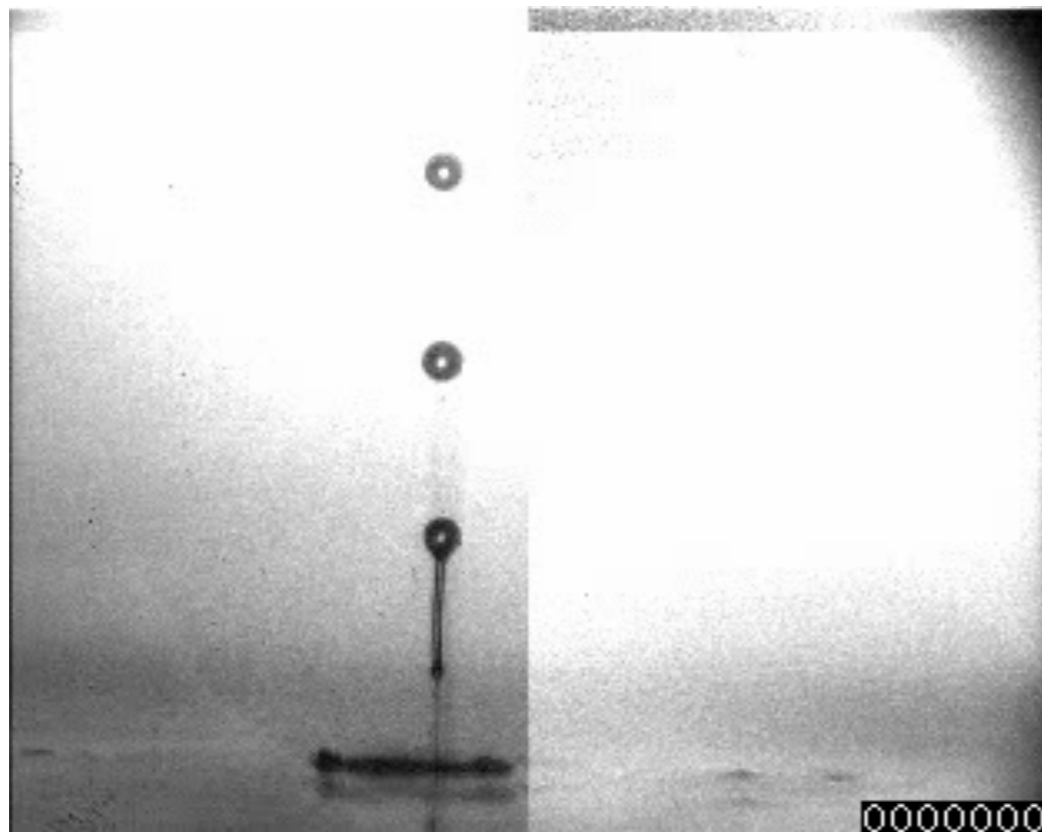
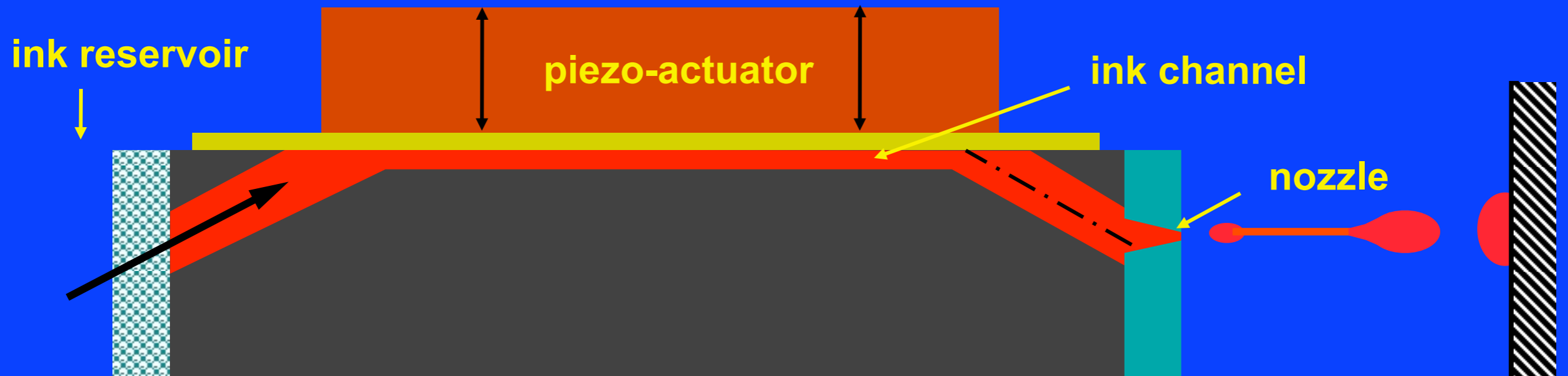
X8,000

2µm

20 20 SEI

# **Making of extremely controlled droplets: inkjet printing**

# Piezoacoustic inkjet printing

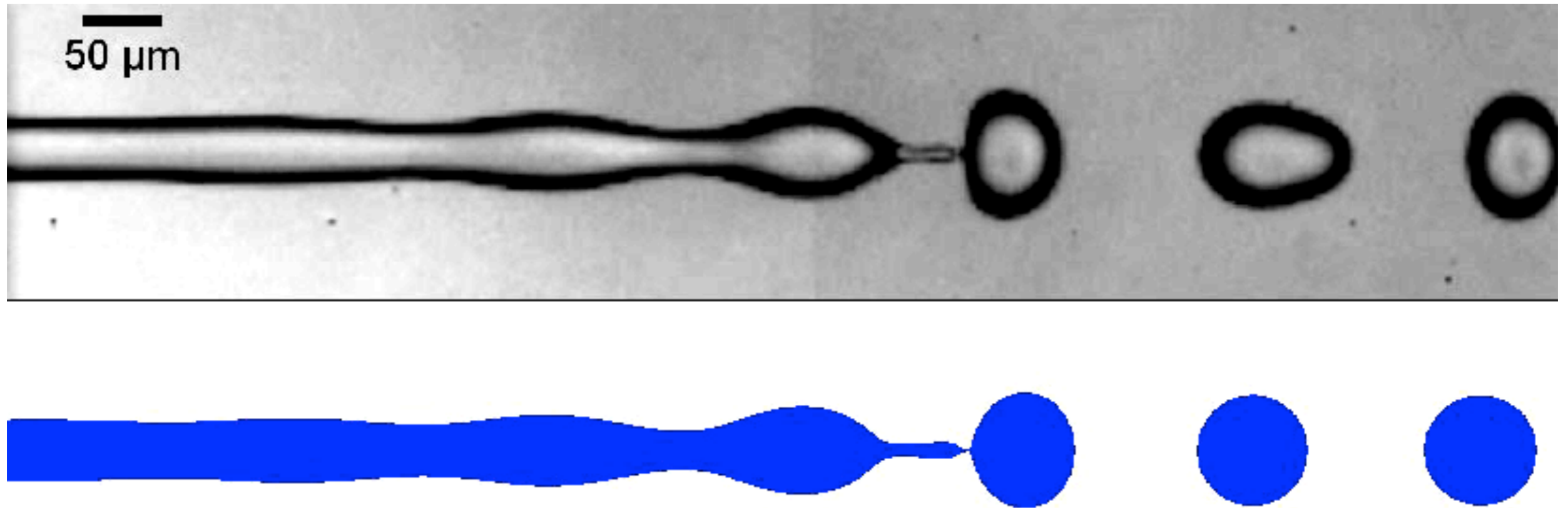


... or even MEMS-based ...



High speed imaging:  
1 MHz

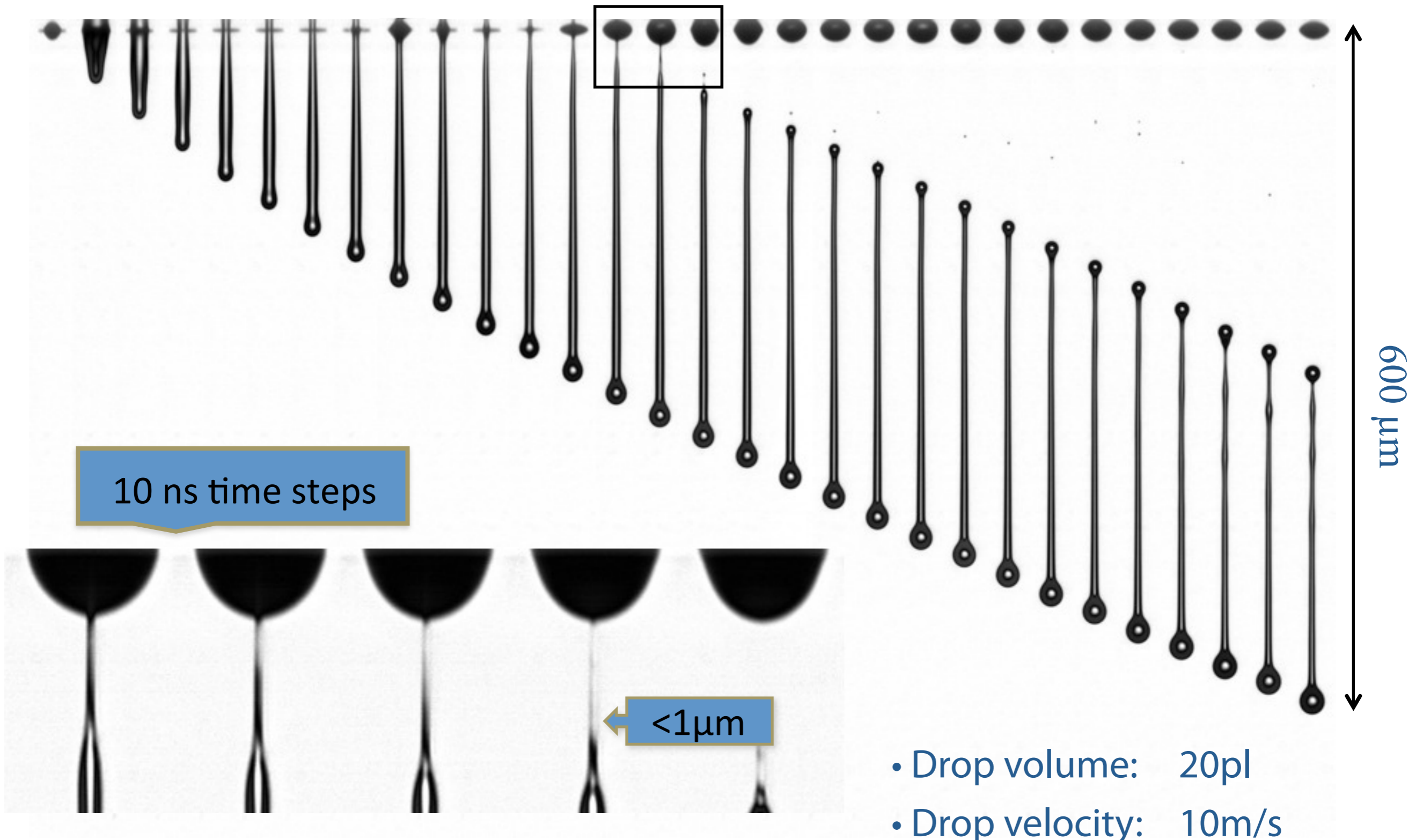
# Comparison experiment vs lubrication model



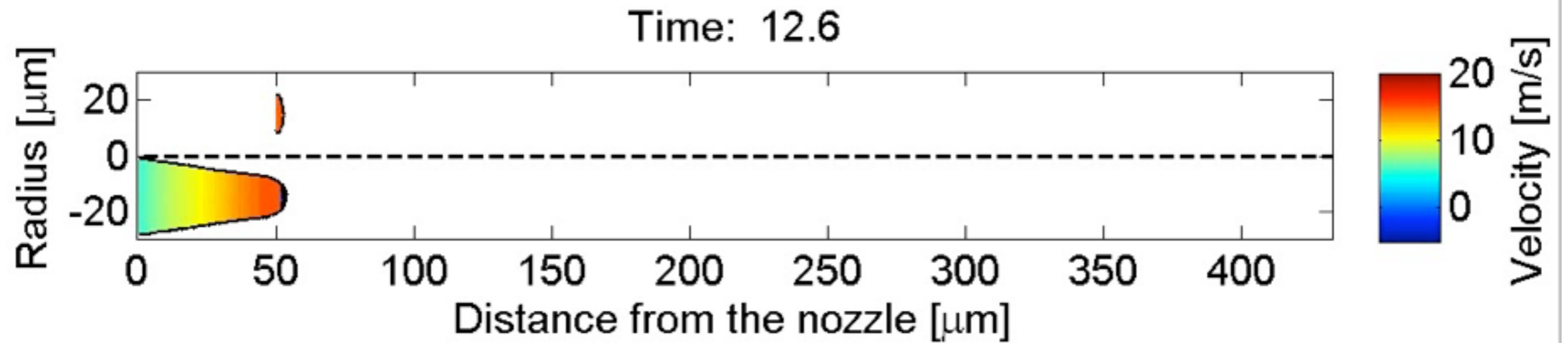
Satellite droplets



# Stroboscopic imaging: iLIF



# Comparison with lubrication model



Num.



Exp.



Difference due to air friction!

# Predictions from lubrication model for inks with different viscosity

Viscosity = 5.5mPas

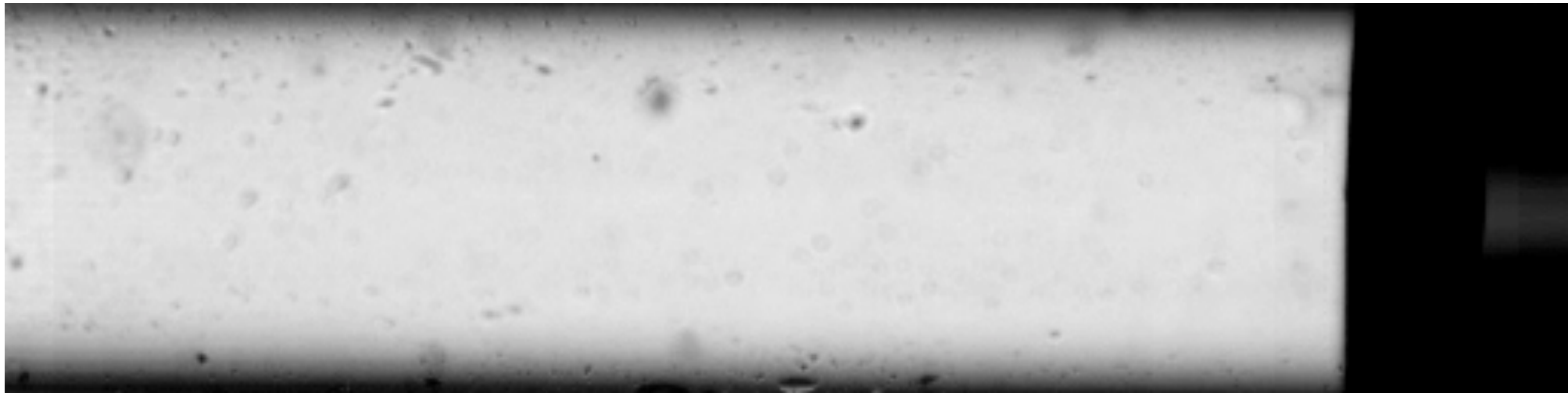
Viscosity = 8.25mPas

Viscosity = 11mPas

Viscosity = 16.5mPas

Viscosity = 22mPas

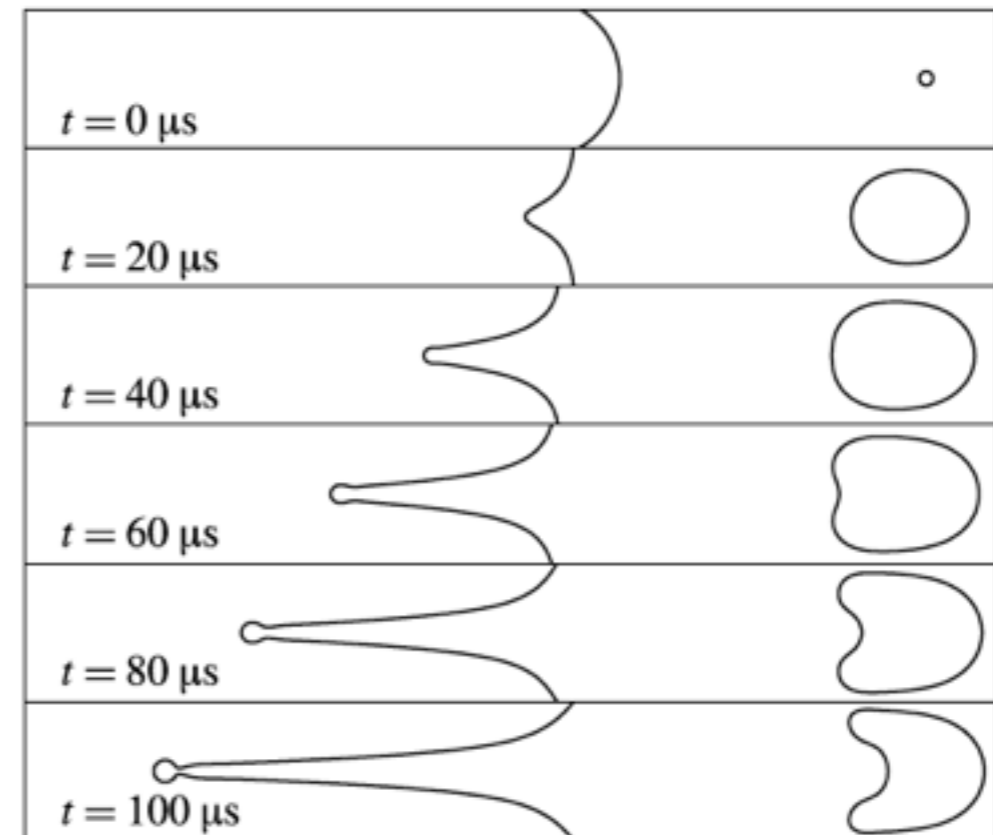
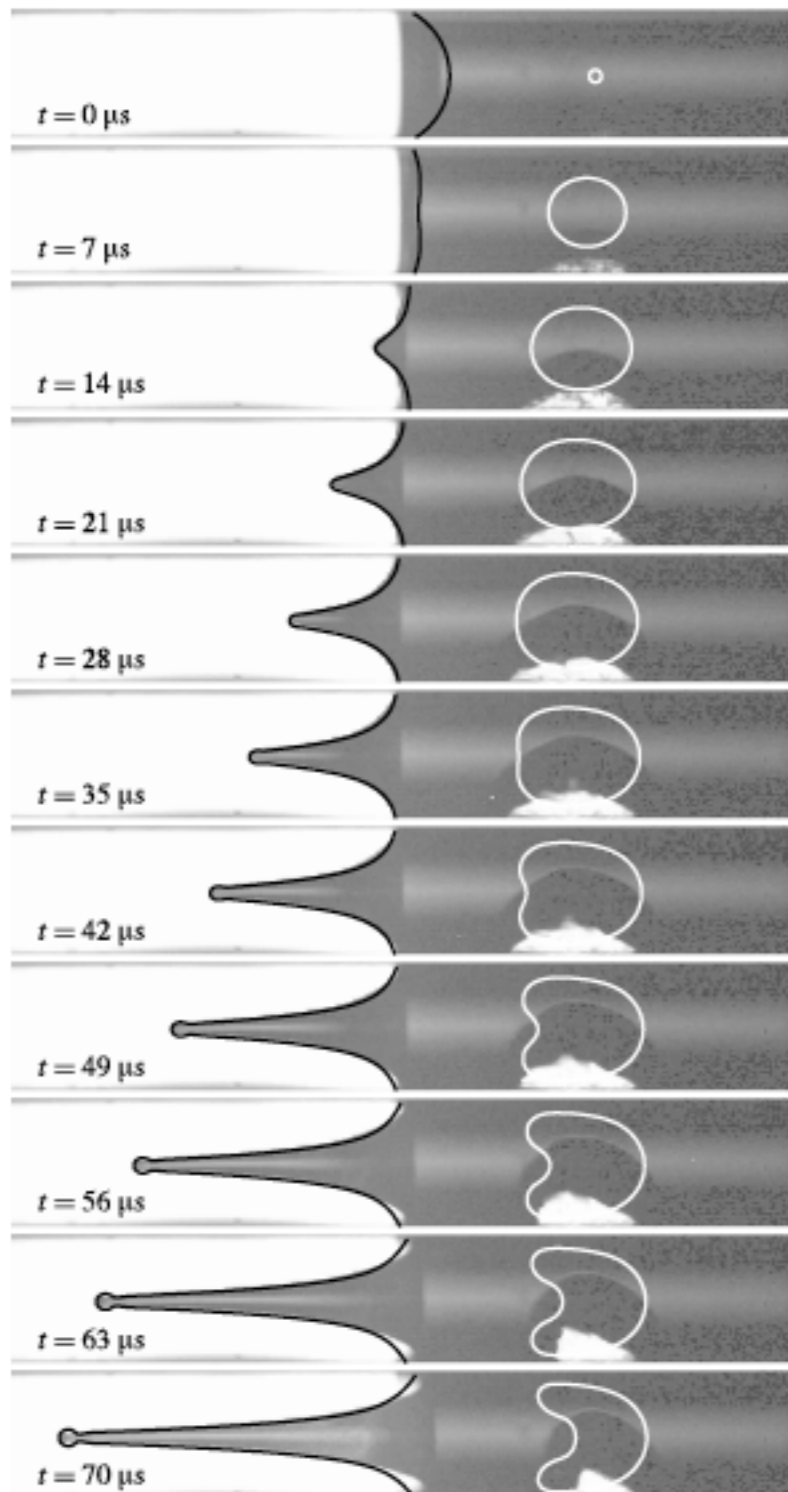
# Supersonic microjets through flow focusing!



**$v \sim 1000 \text{ m/s}$      $D \sim 10 \mu\text{m}$**

I. R. Peters *et al.*, JFM 719, 587 (2013)

# Mechanism: flow focusing



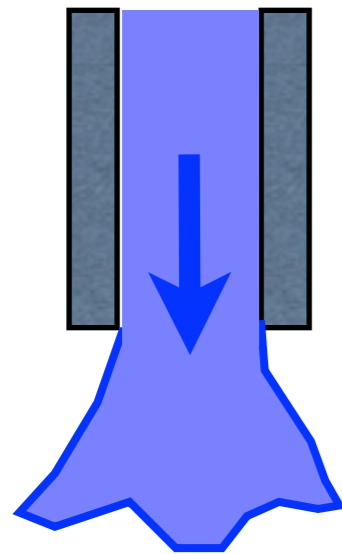
boundary integral

experiment

# Application: Needle-free injection

## Existing methods

Piston-syringe system

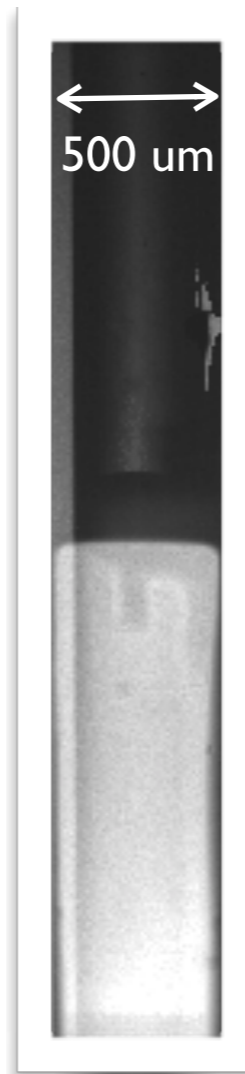


Diffusive jets

- Severe deceleration
- Scattered pattern
- ➔ Causes pain
- ➔ Insufficient penetration
- ➔ Little volume control

## Novel method

### Highly-focused supersonic microjets

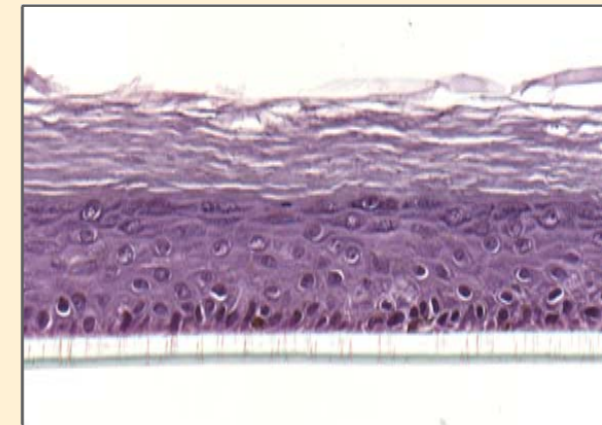


Target materials

Tagawa et al., PRX 2, 031002 (2012)

- **Highly-focused** shape
- **Ultra-high speed** (up to 850 m/s)
- Good controllability

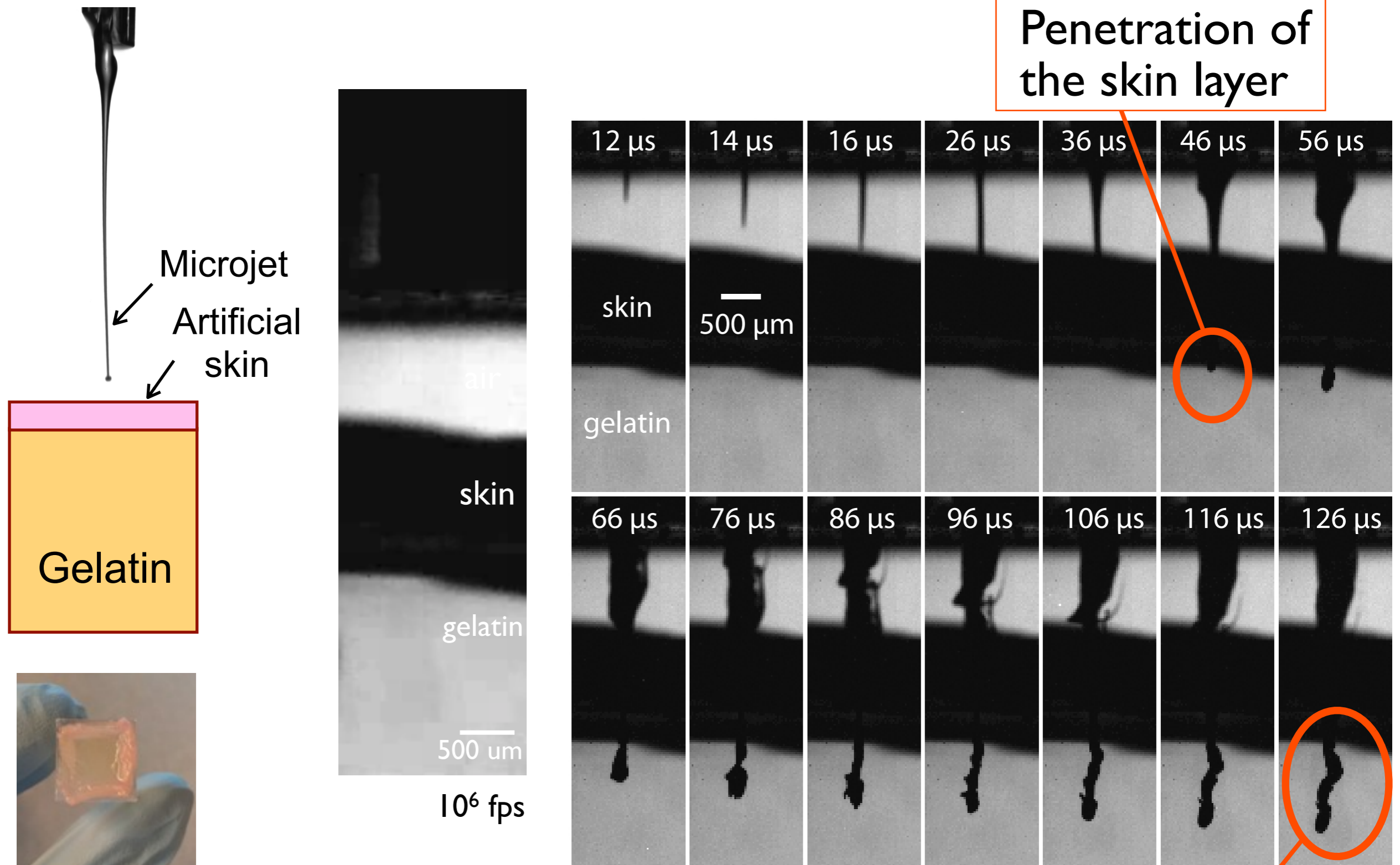
#### ◆ Artificial human skin



*Leiden epidermal skin model*

- ◆ Gelatin 5wt%  
(Human soft tissue equivalent)

# Application: Needle-free injection

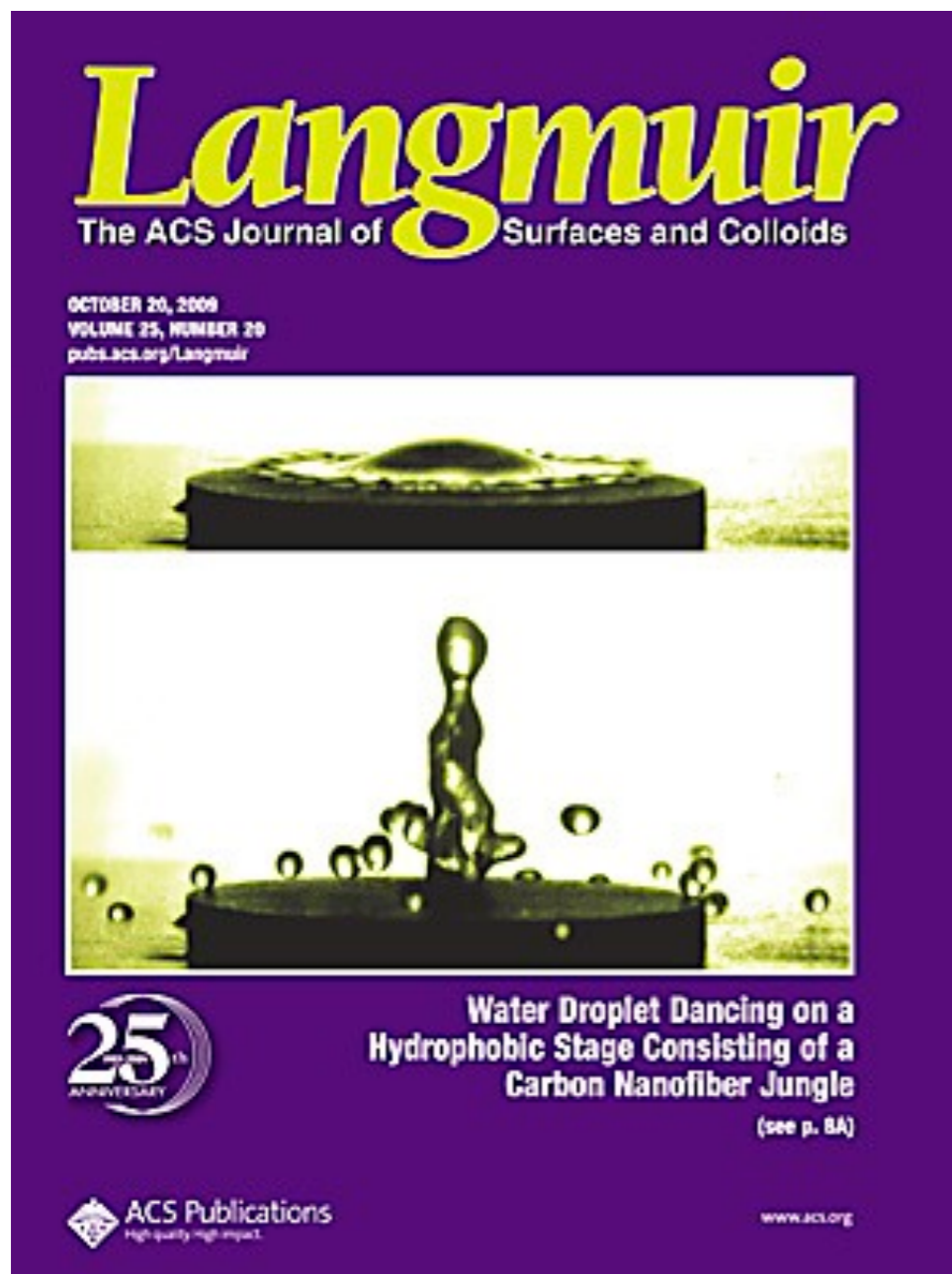


Equivalent to human body

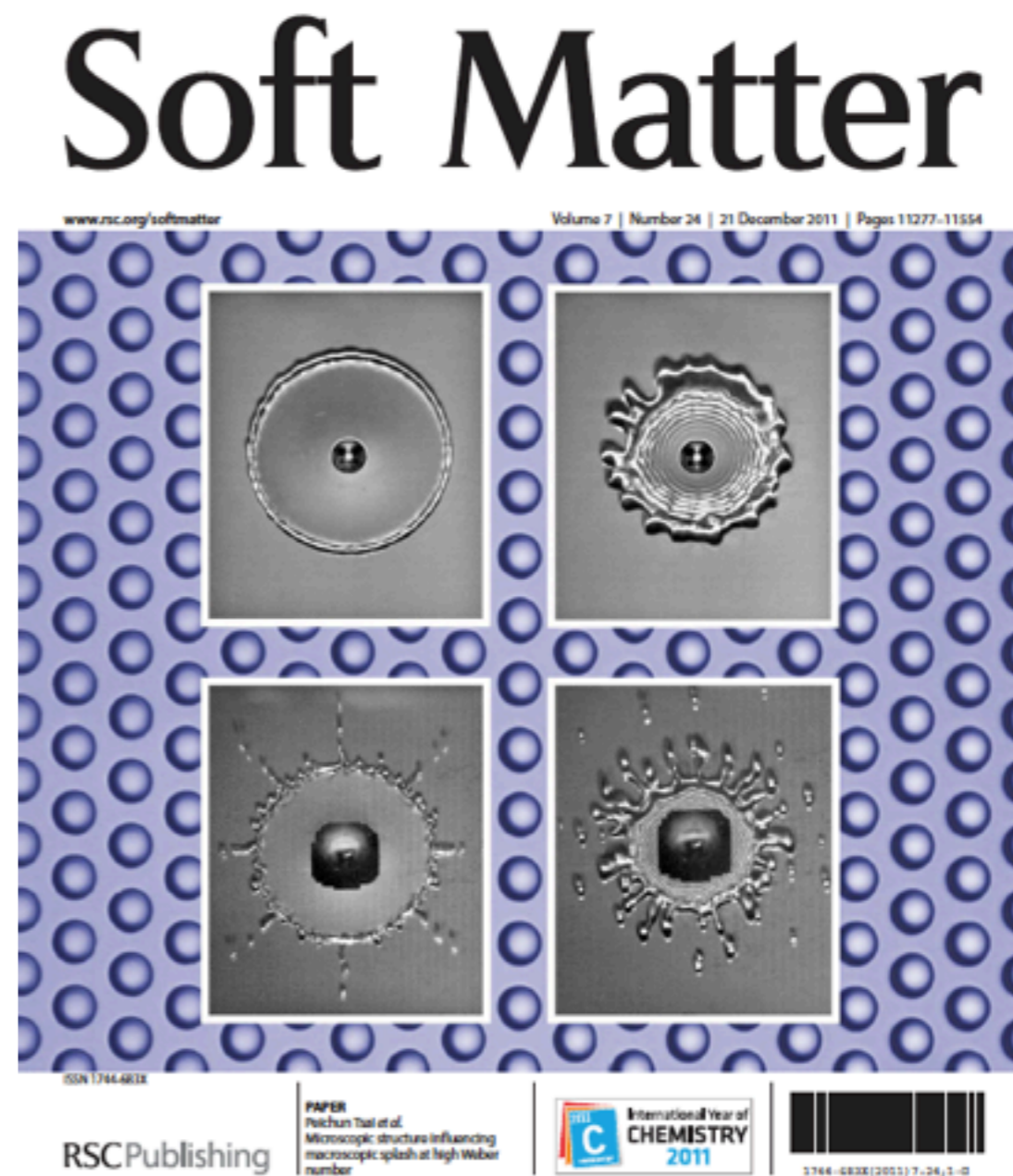
Tagawa *et al*, Lab on Chip (2013)

# Impact on **structured** surfaces



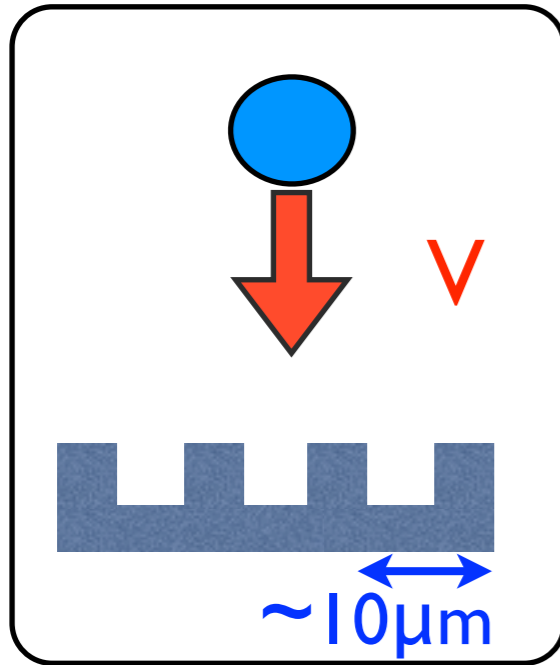


P. Tsai, S. Pacheco, C. Pirat, L. Lefferts, & D. Lohse,  
**Drop impact upon micro- and nanostructured superhydrophobic surfaces,**  
Langmuir **25**, 12293 (2009).



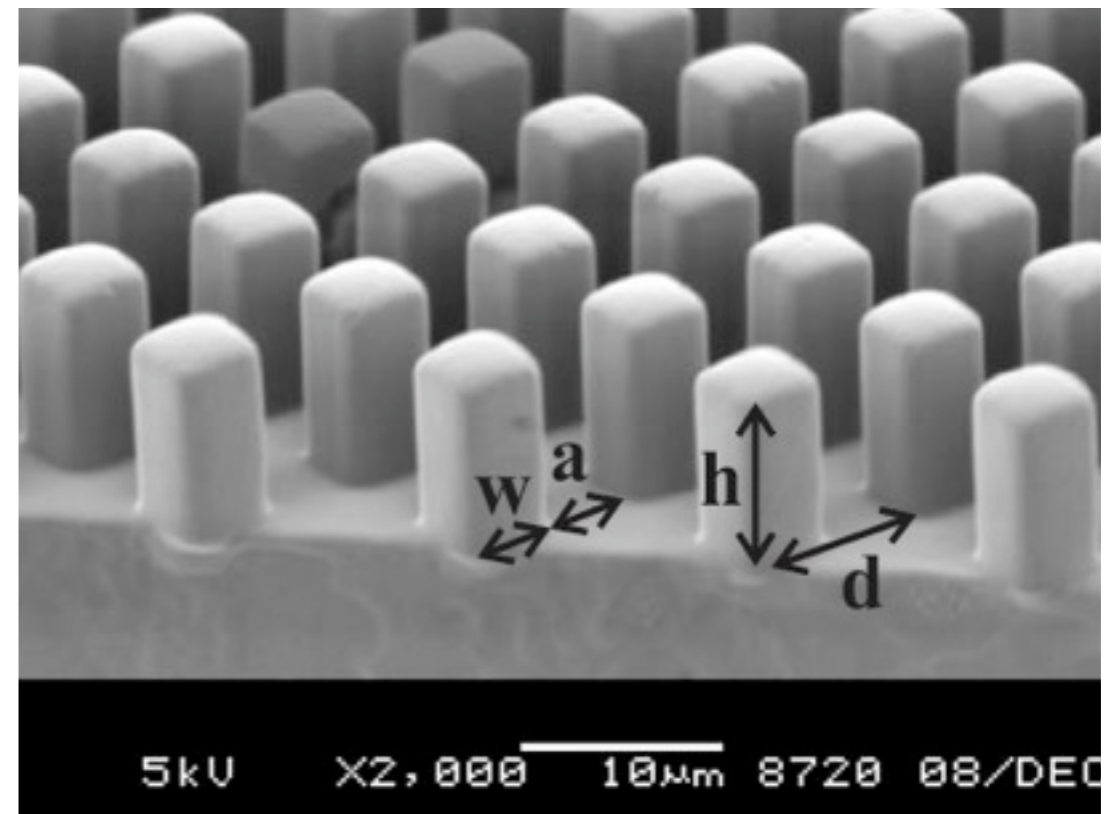
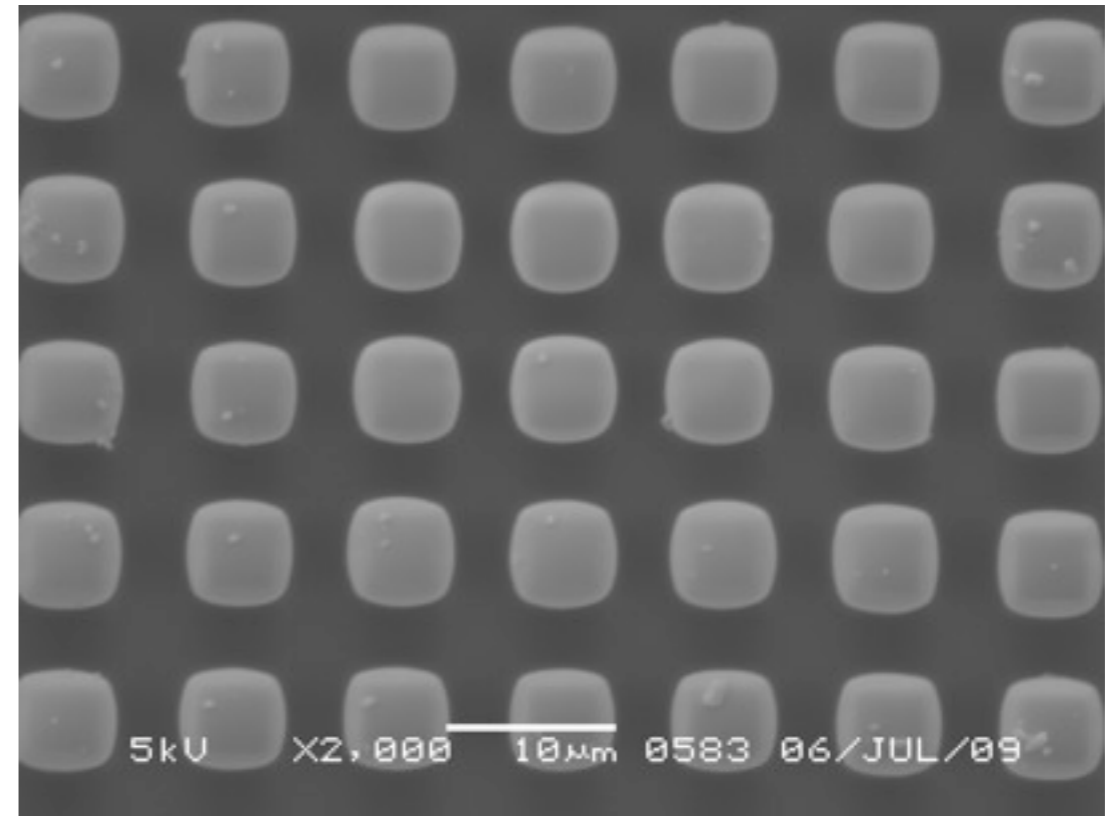
P. Tsai, M. Hendrix, R. Dijkstra, L. Shui & D. Lohse,  
**Microscopic structure influences macroscopic splash at large  $We$ ,**  
Soft Matter **7**, 11325 (2011).

# Structure of the surfaces

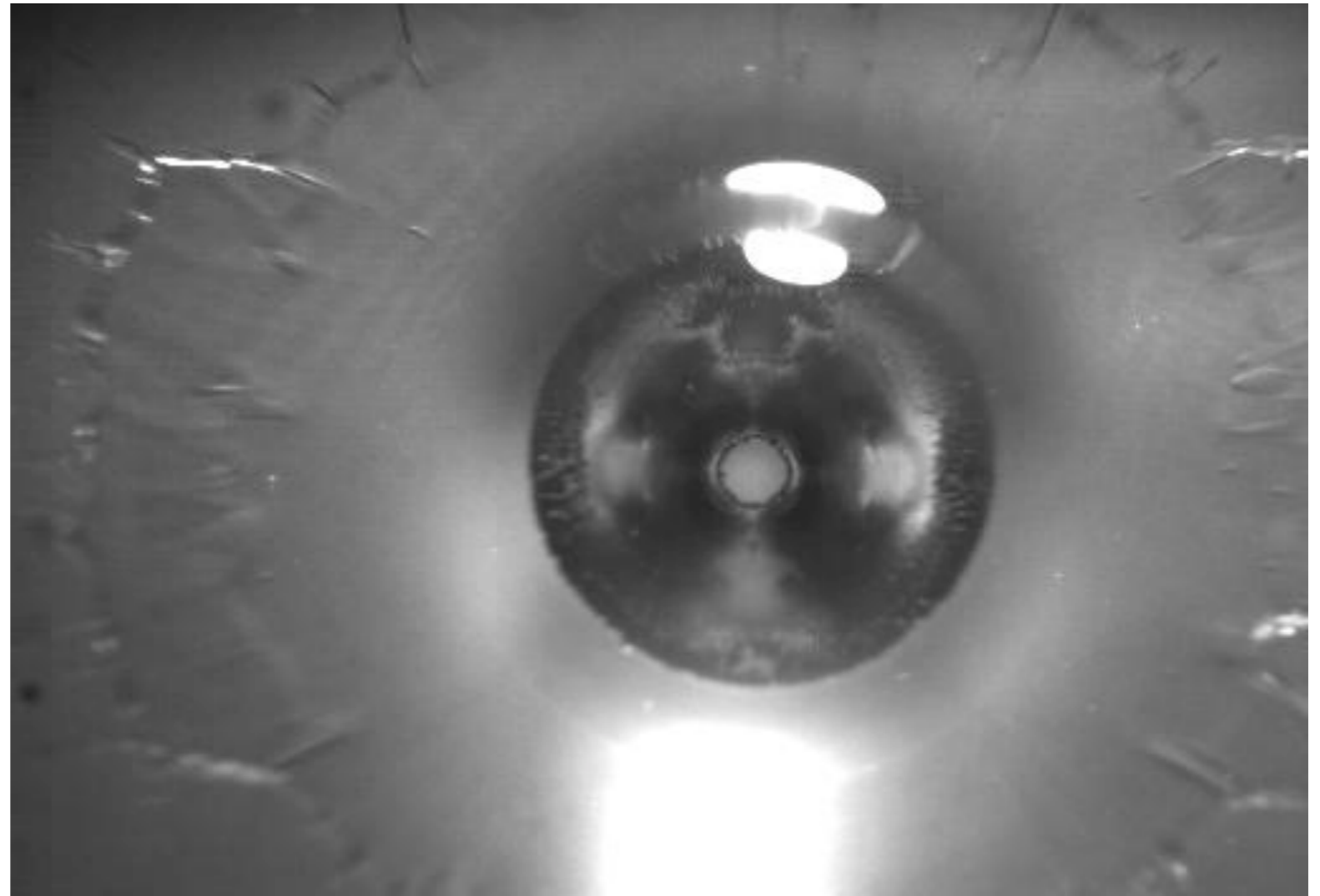
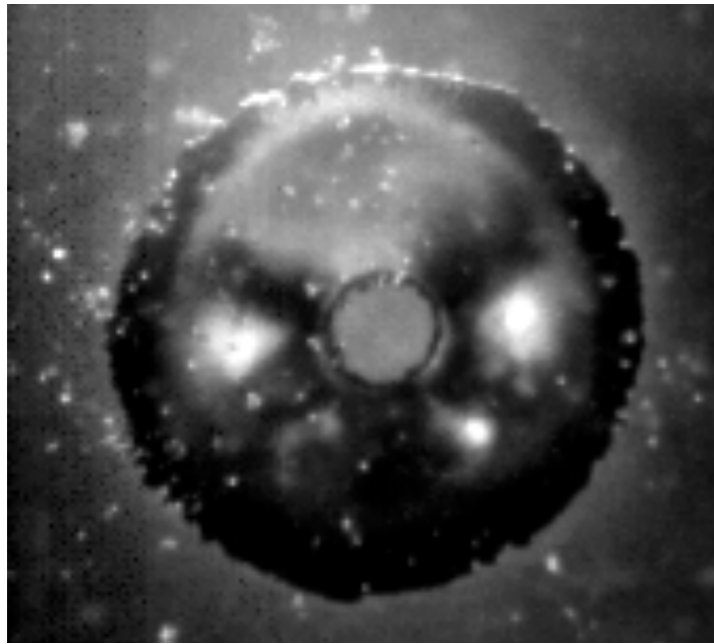


Transparent PDMS  
(polydimethylsiloxane)

$h = 6 \mu\text{m}$   
 $w = 5 \mu\text{m}$



# Entrapped air bubbles & wetted central region

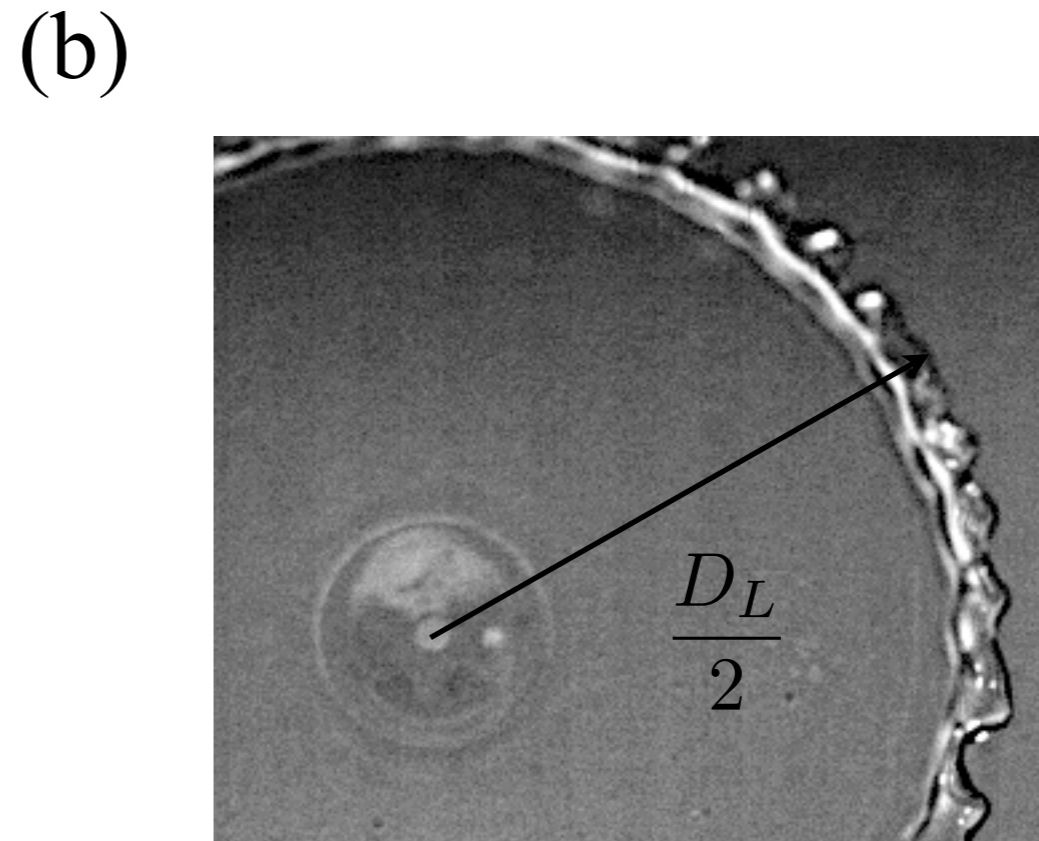
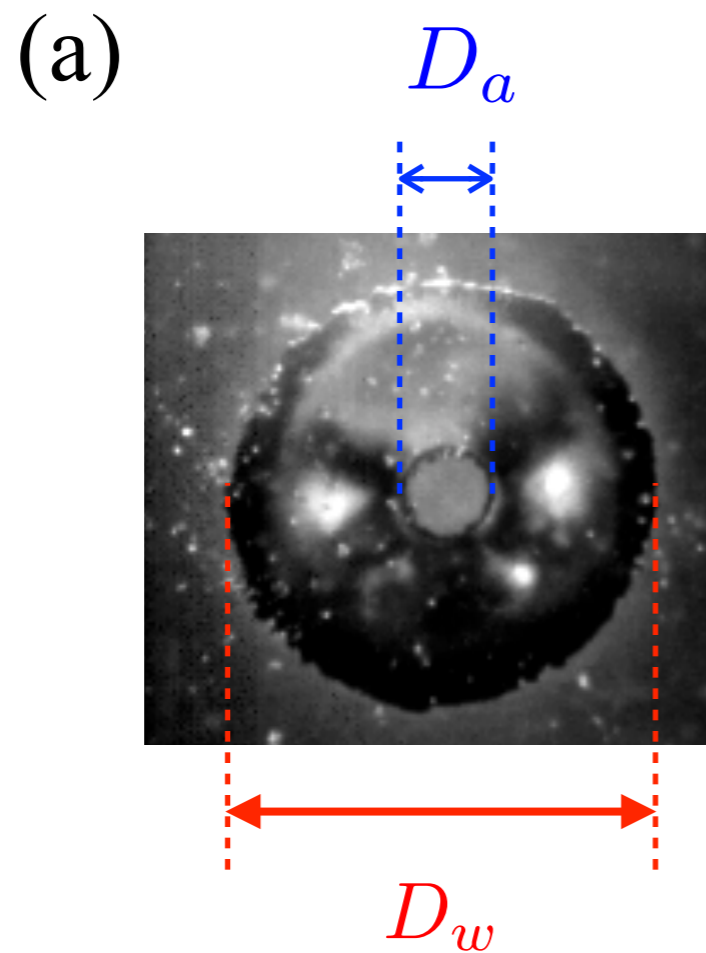




$D_a$  = diameter of entrapped air disk

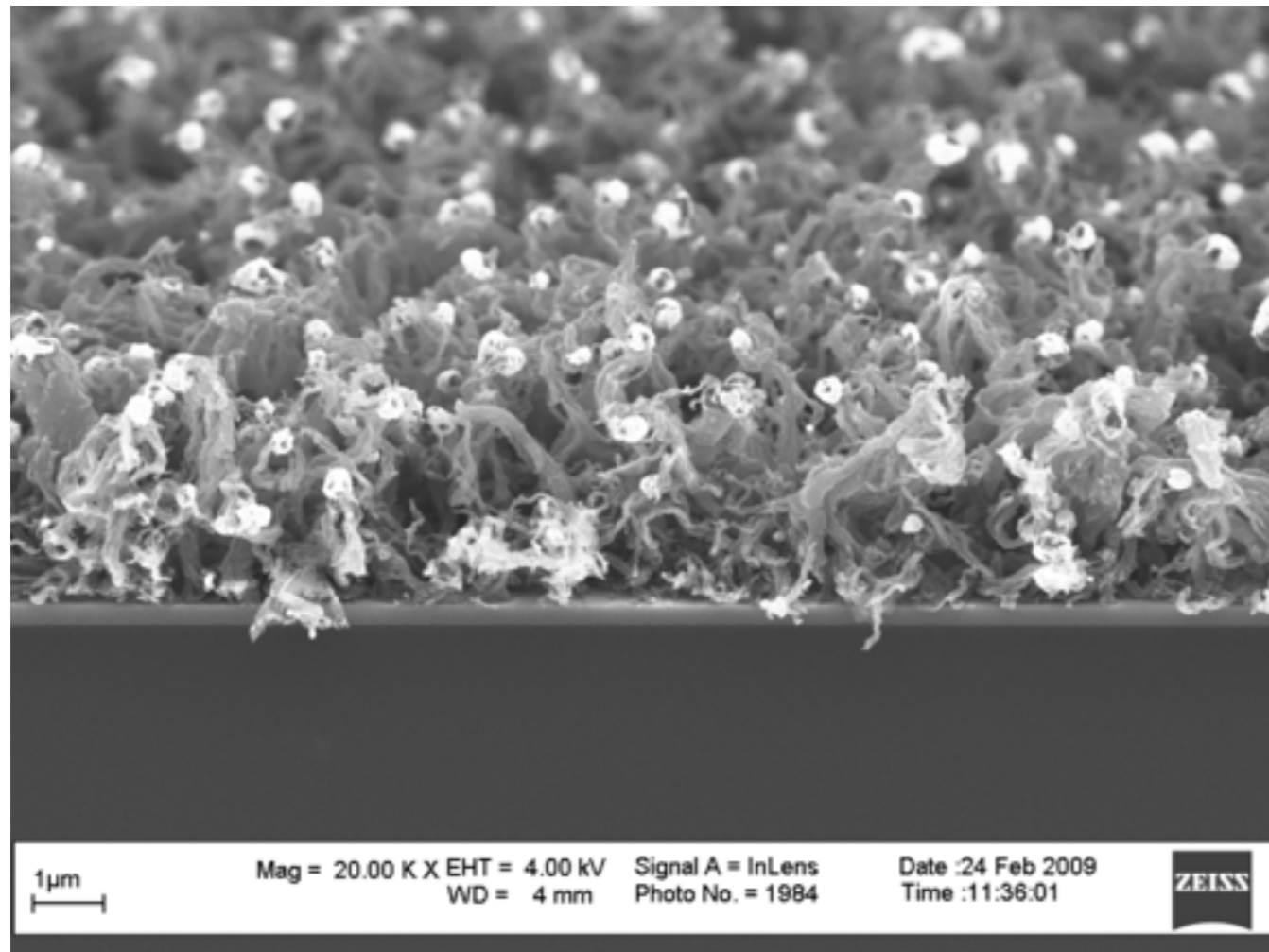
$D_w$  = diameter of wetted area

$D_L$  = diameter of max. spreading lamella



$$w = 5\mu m, h = 6\mu m, d = 10\mu m$$

# Impact on superheated carbon nanofiber jungle surface



$H \sim 4 \mu\text{m};$   
 $d_{\text{CNF}} \sim (80-200) \text{ nm}$

$D \sim 2.27 \text{ mm}$

$U \sim 0.4 \text{ m/s}$

SEM

**Photron**

FASTCAM SA2 mode... 10000 fps

1/30000 sec

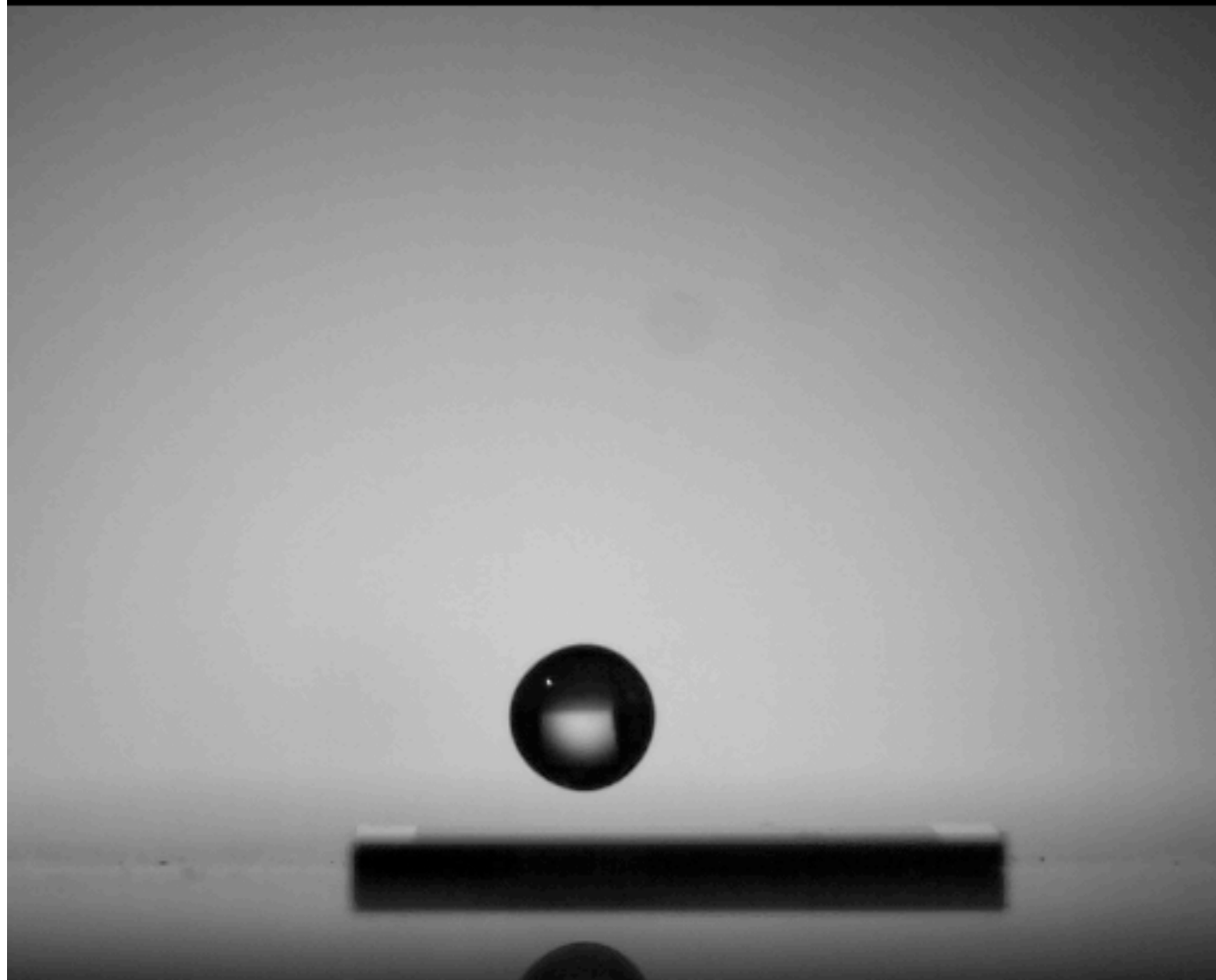
512 x 416

frame : -2635

-00:00:00.2635

Date : 2011/11/15

Time : 14:28



T=350°C

**Photron**

FASTCAM SA2 mode... 10000 fps

1/30000 sec

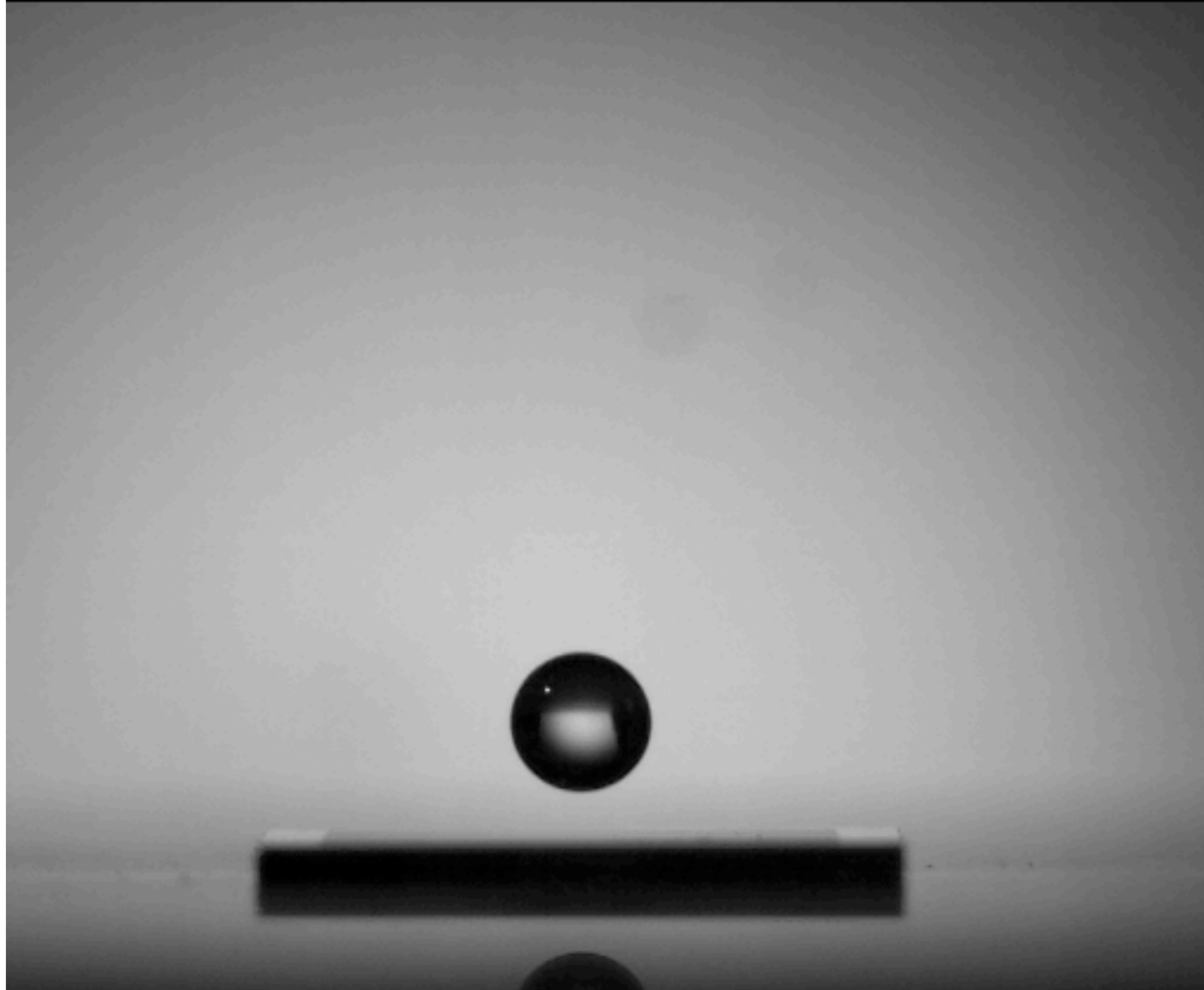
512 x 416

frame : -3224

-00:00:00.3224

Date : 2011/11/15

Time : 15:12



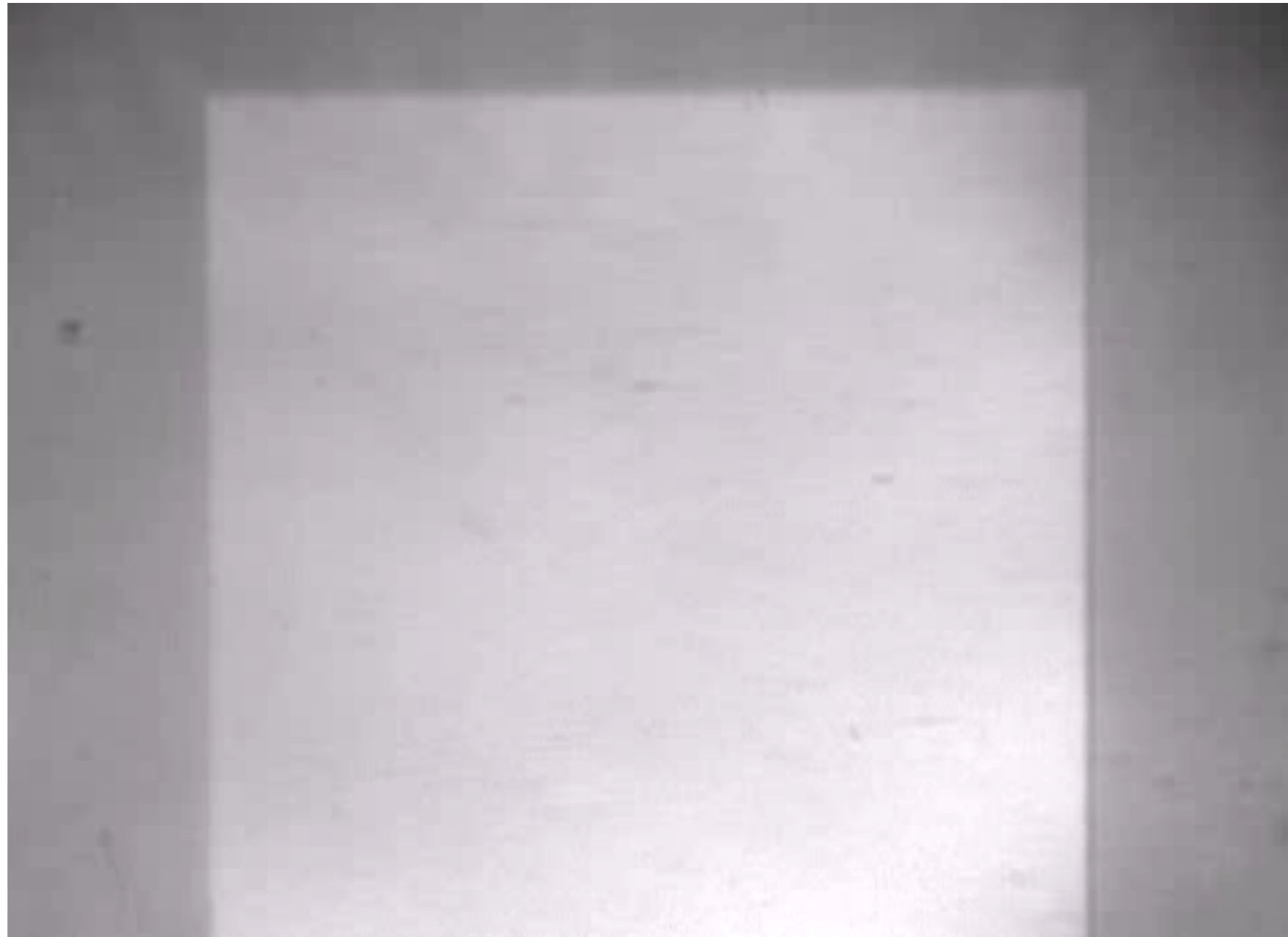
T=440°C

# Surface manipulation and **controlled cavitation**

Bremond, Arora, Ohl, Lohse, Phys. Rev. Lett. 96, 224501 (2006);  
Borkent, Gekle, Prosperetti, Lohse, Phys. Fluids 21, 102003 (2009)

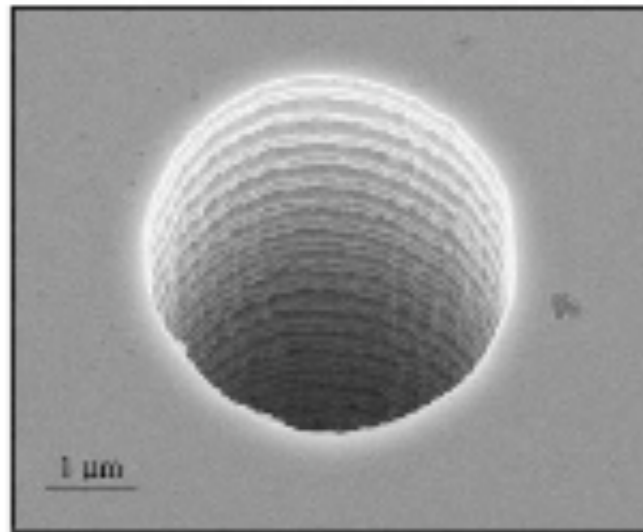


**Massive pressure reduction:**  
**Uncontrolled** cavitation & bubble nucleation

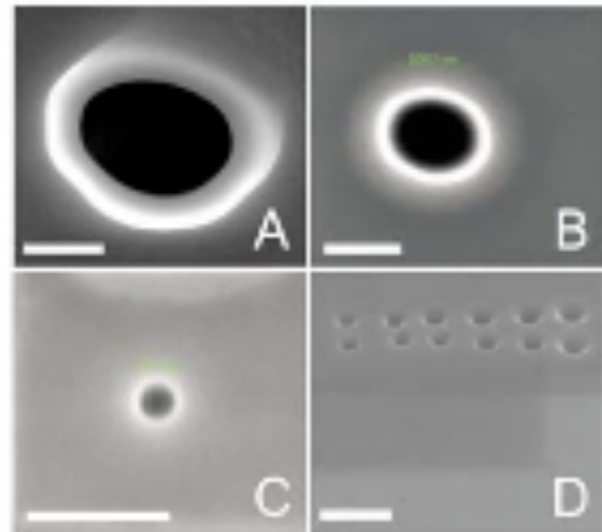


| Mfps movie, Si surface, nucleation at natural crevices

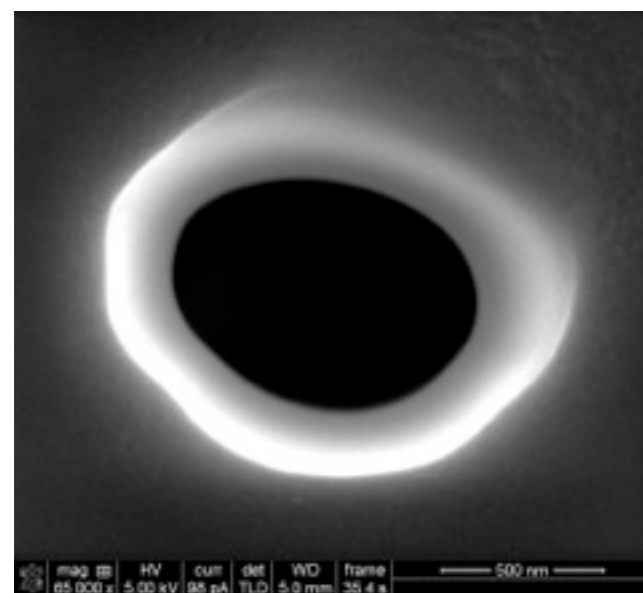
# Offer system **weak spots** through micro- & nano-machined “artificial crevices”



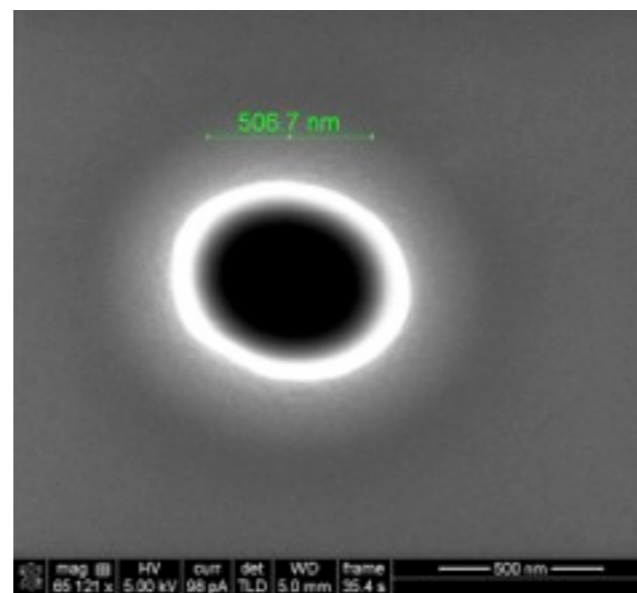
2000nm



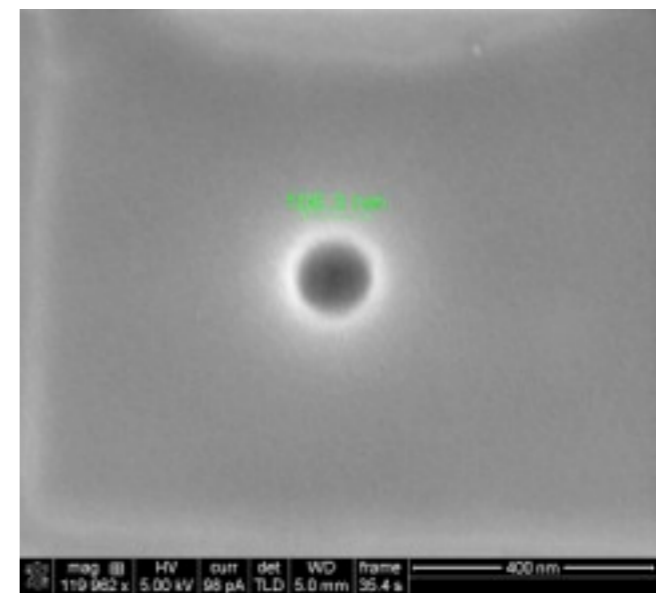
made by focused ion-beam



900nm

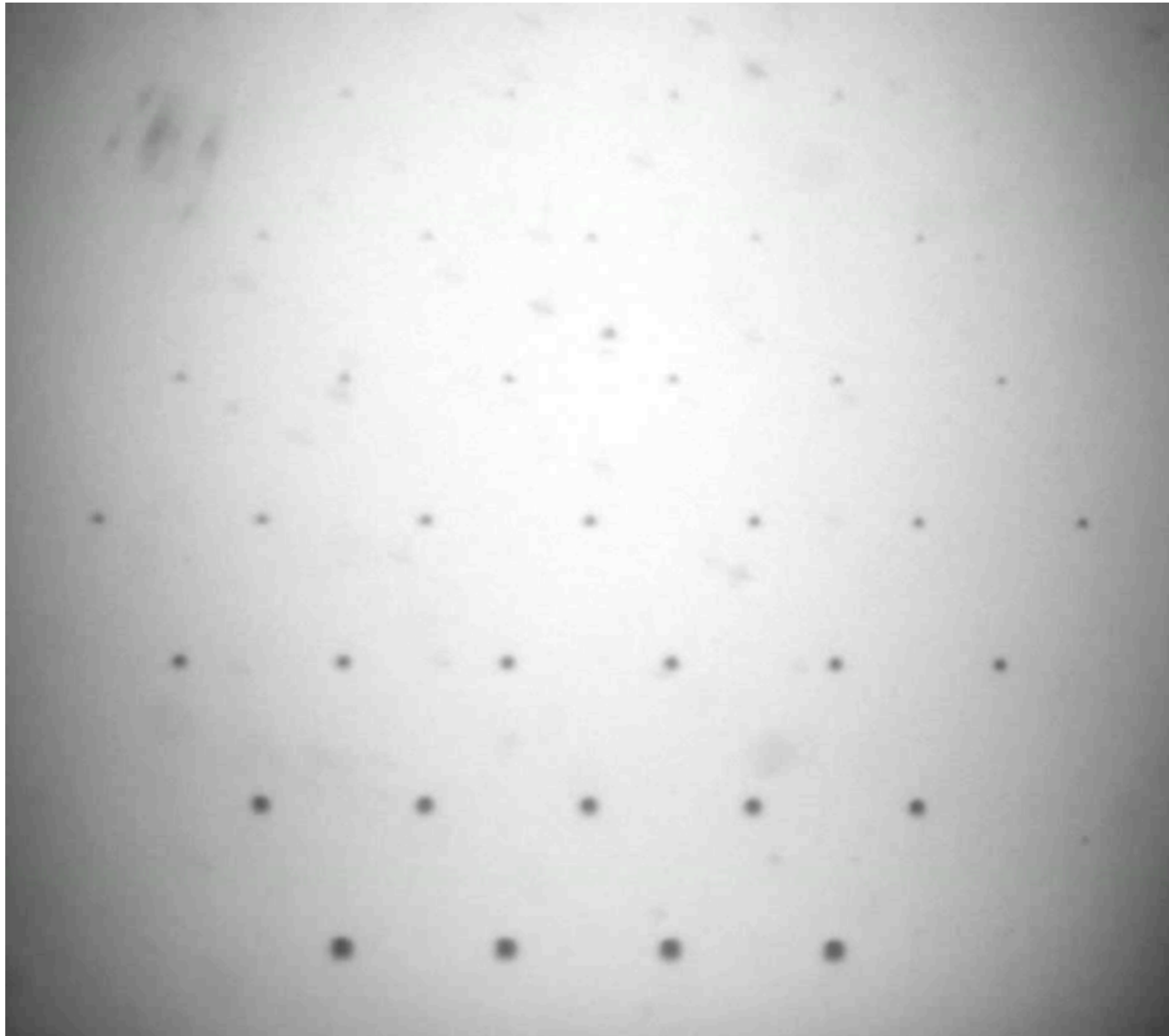


500nm

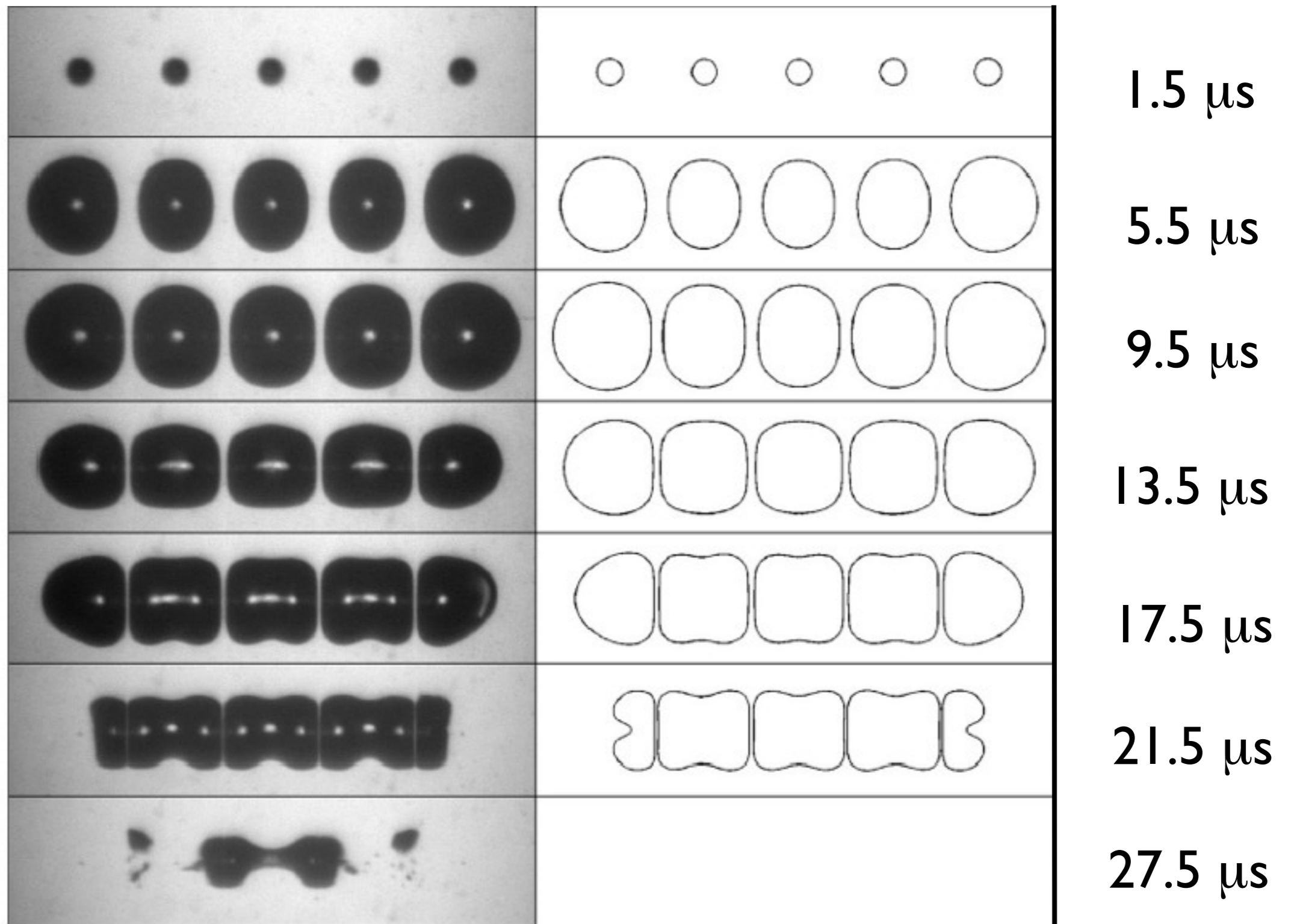


100nm

# Pattern of bubbles

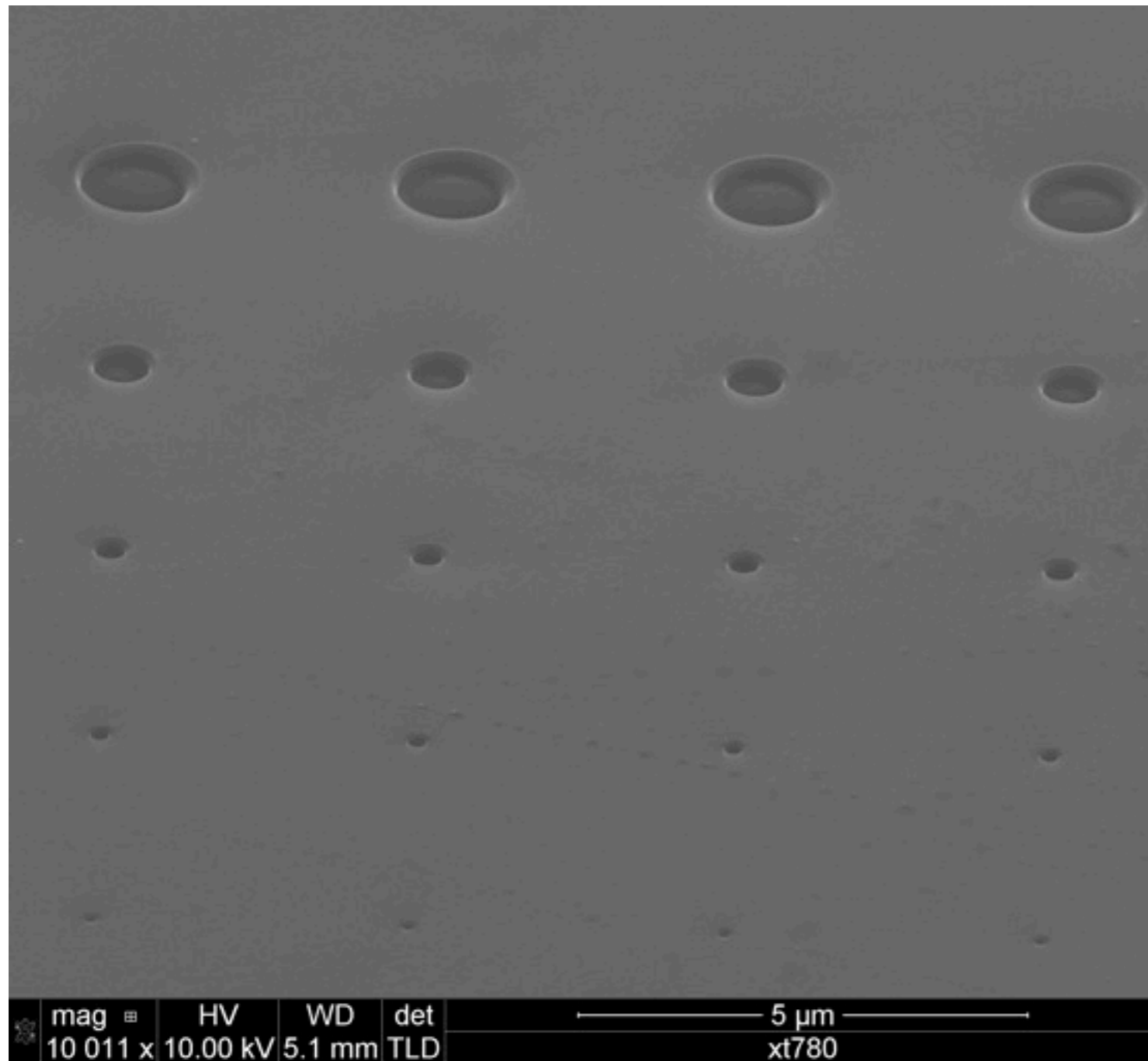


# Shielding effect of bubbles: experiment vs theory



# Cavitation from nanopits?

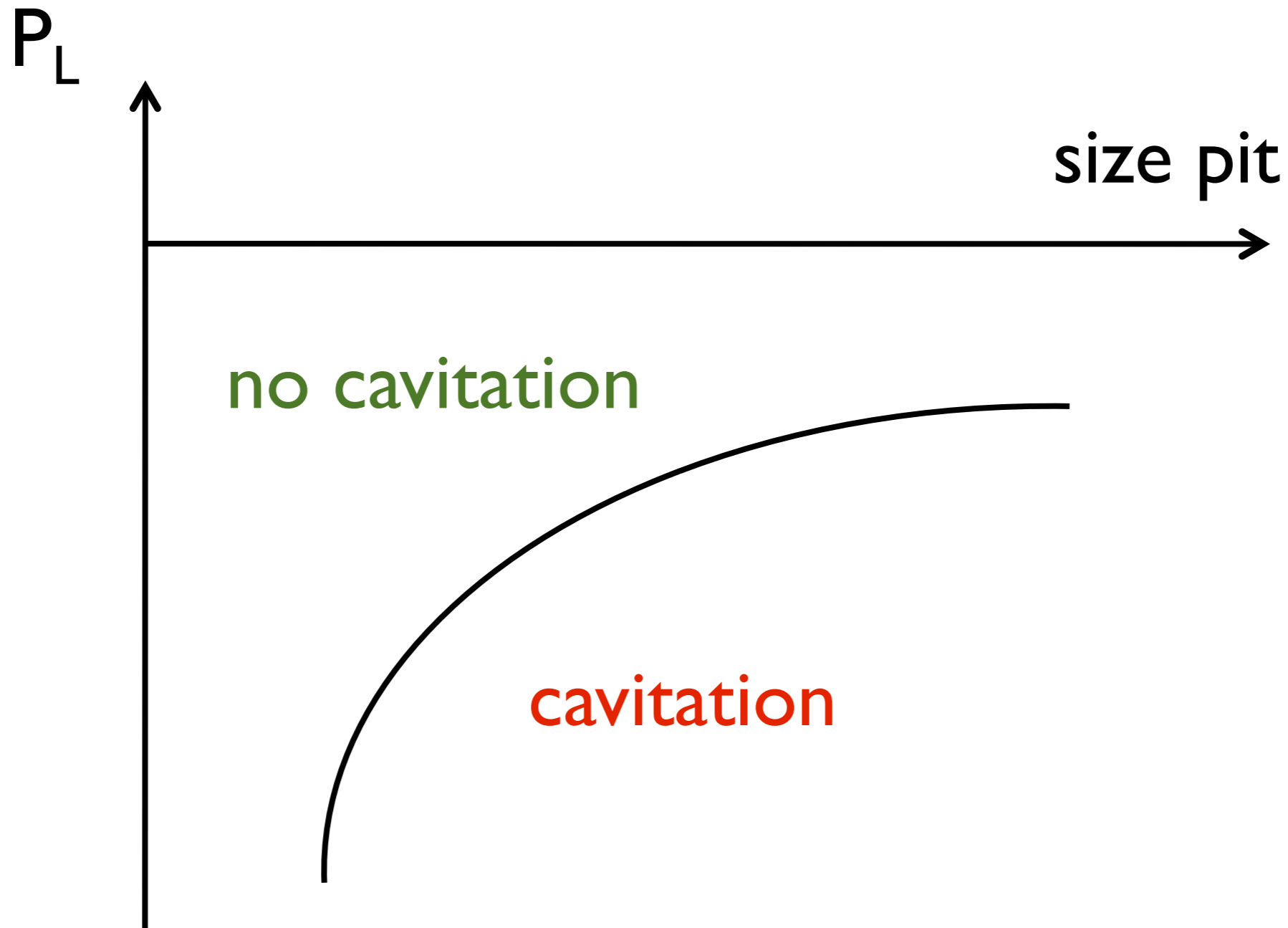
## How small can we go?



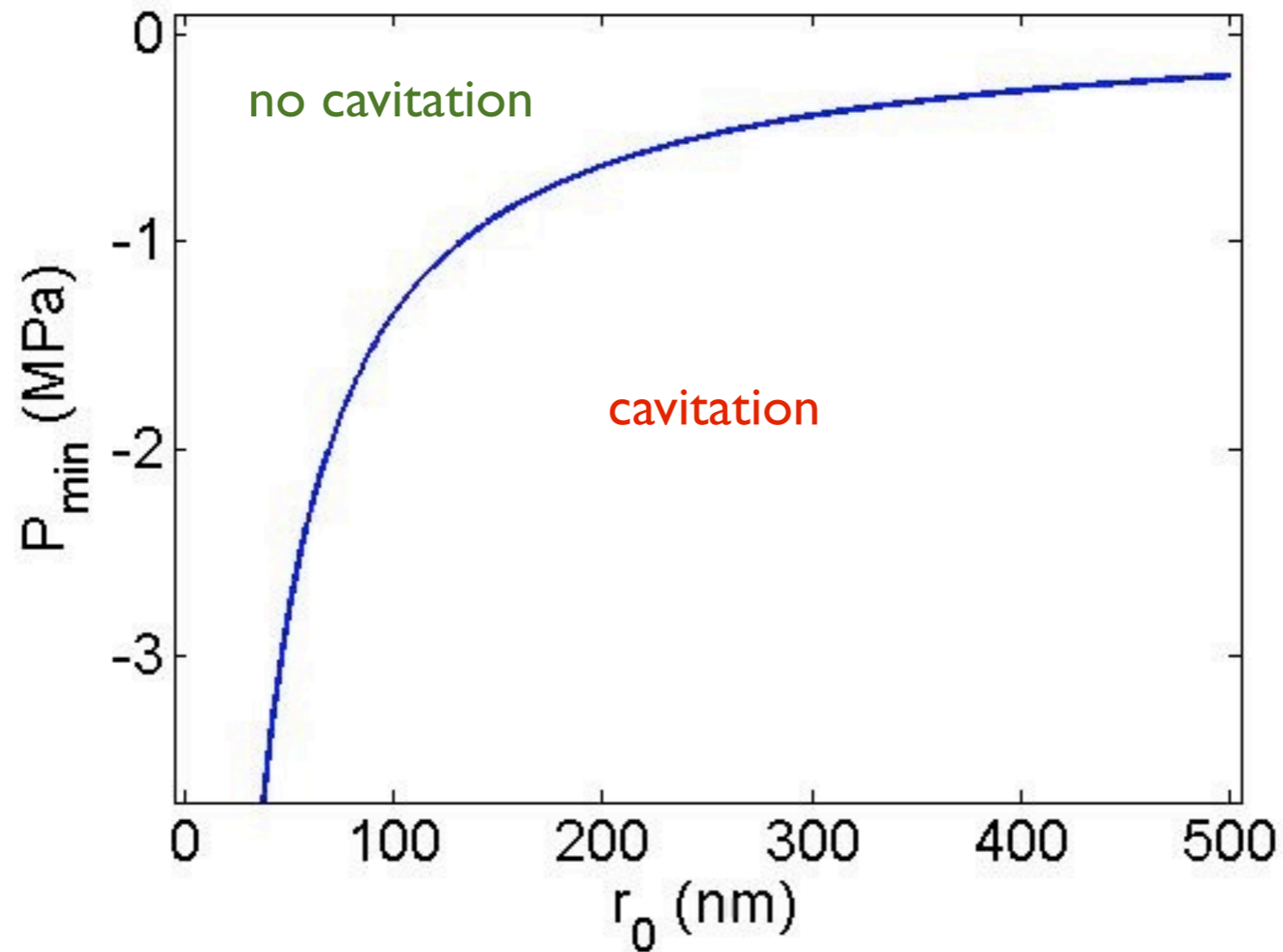
900nm

100nm

# Calculate & measure phase diagram of bubble nucleation



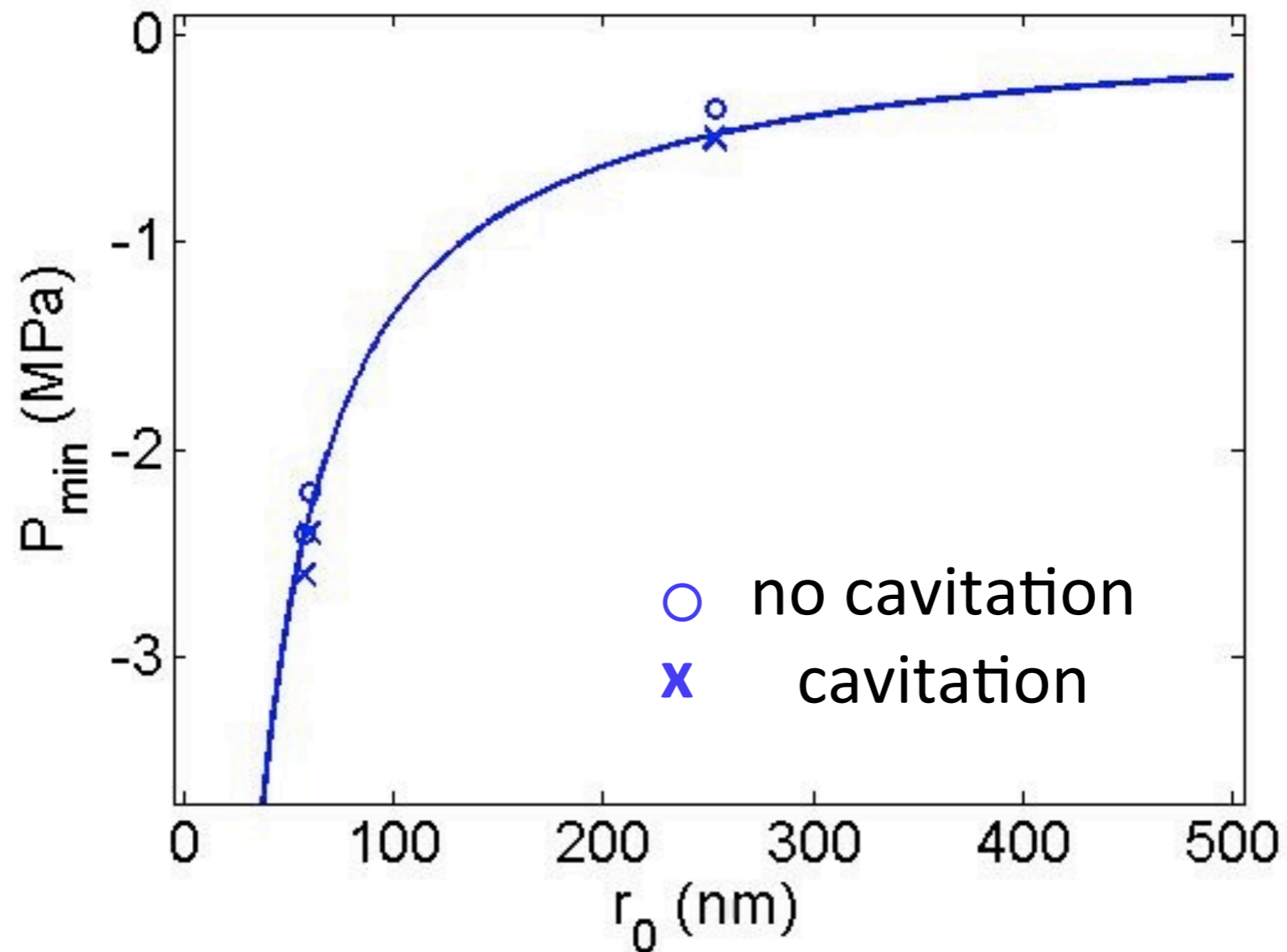
# Theoretical prediction



$$p_L = p_v + \frac{3p_{g,0}}{3 + r_0/d_0 f(\theta)} - \frac{2\sigma}{r_0}$$

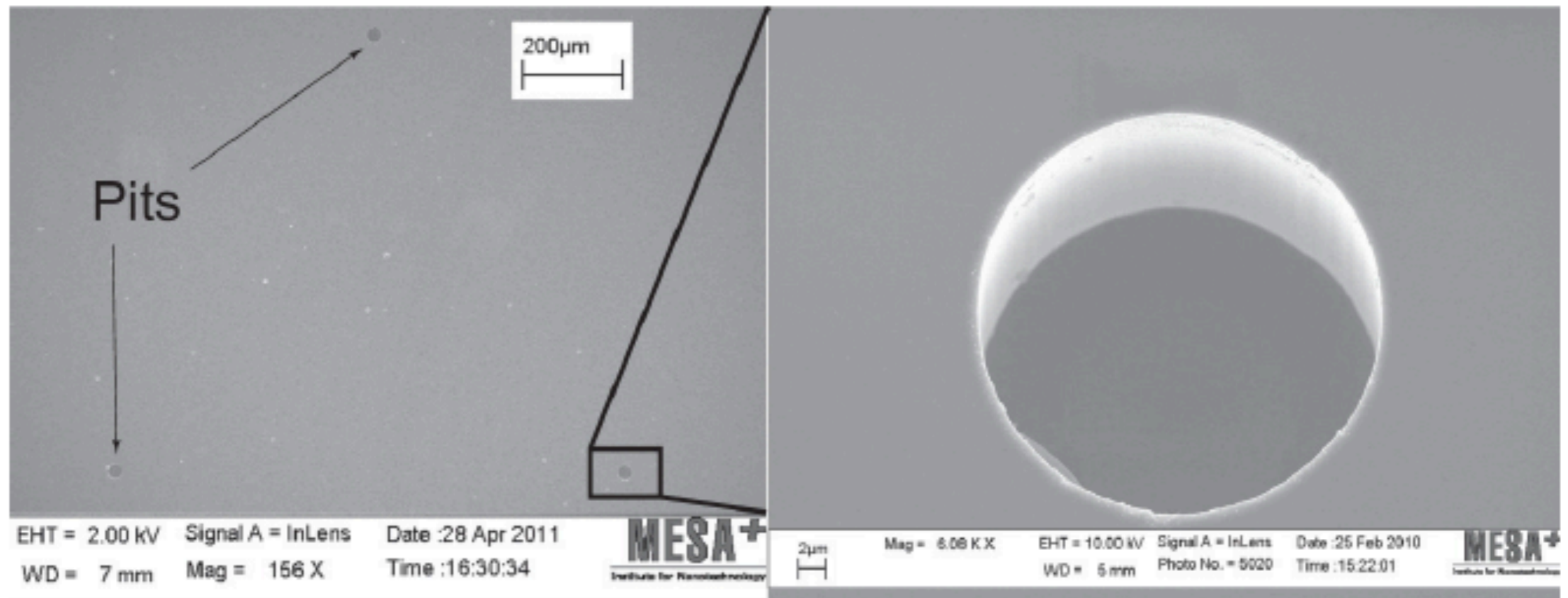


# Perfect agreement theory vs experiment



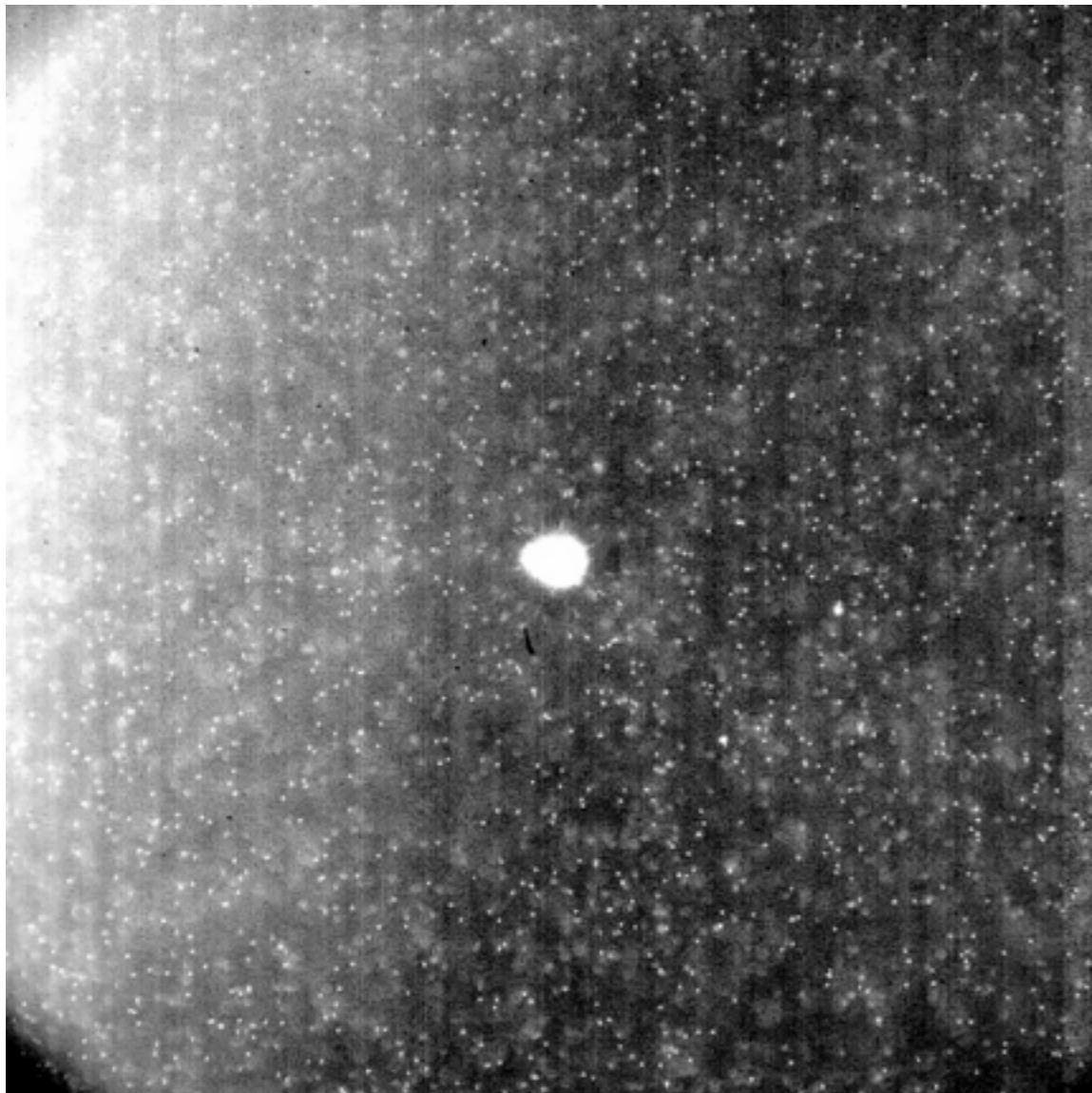
No free parameter!

# Use micropits to stimulate bubble production and thus to enhance chemical activity

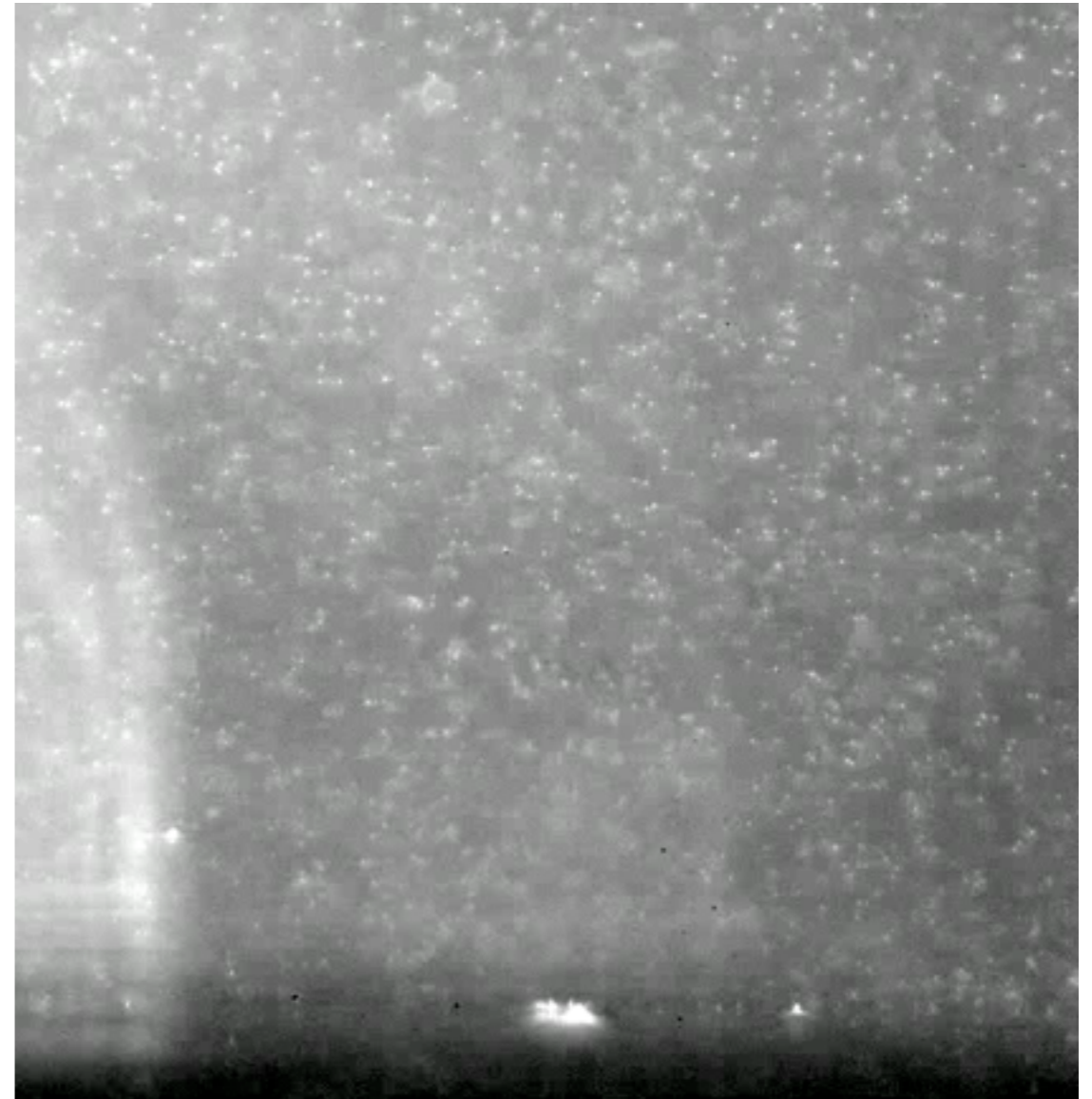


Rivas, Prosperetti, Zijlstra, Lohse, Gardeniers, *Angw. Chem. Int. Ed.* 49, 9699 (2010);  
Rivas, Stricker, Zijlstra, Gardeniers, Lohse, Prosperetti, *Ultrason. Sonochem.* 20, 510 (2013).

# Acoustic streaming around **weakly** driven meniscus



top view



side view

# Stronger driving: Bubble pinch-off out of pit

$f = 80.8 \text{ kHz}$

500,000 fps (0.16 acoustic cycles/frame)

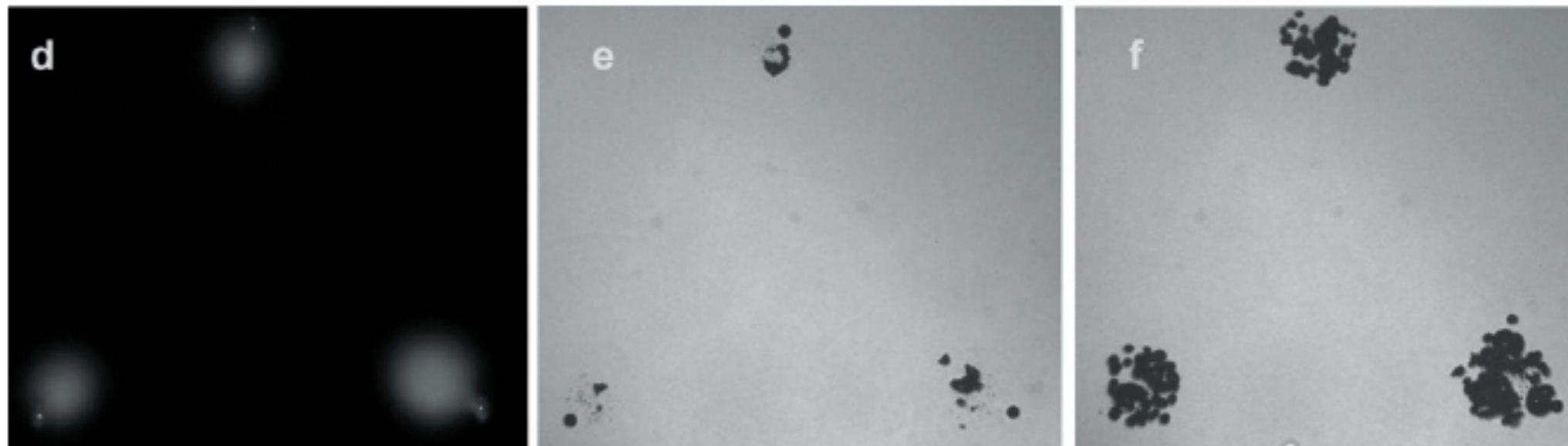


Average pinch-off rate: 13.4 kHz (1 per 6 acoustic cycles)

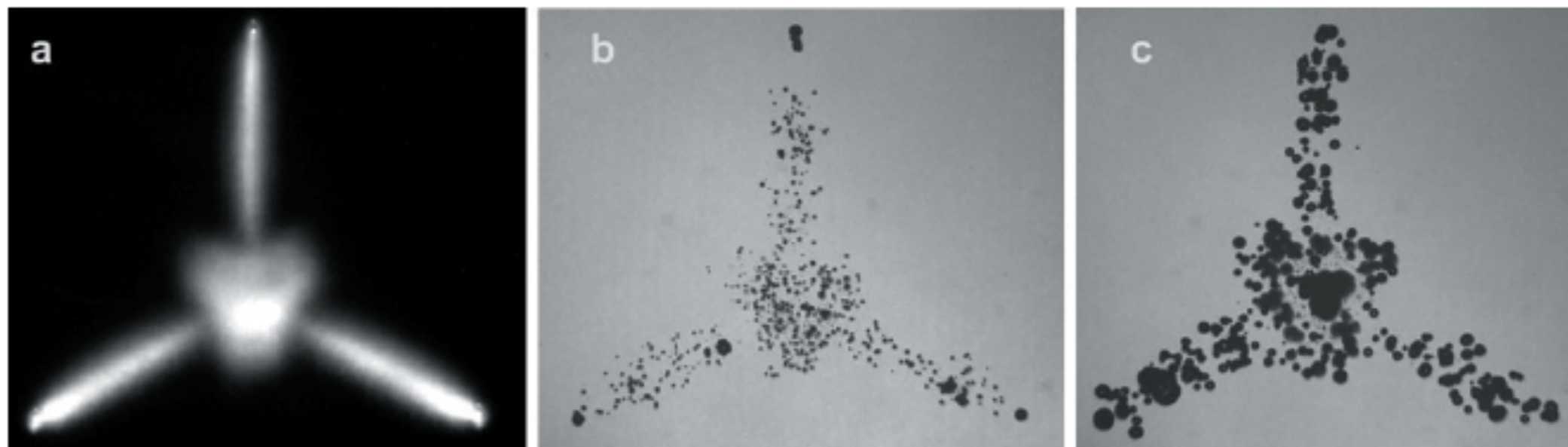


# Three-pit geometry

low power



high power

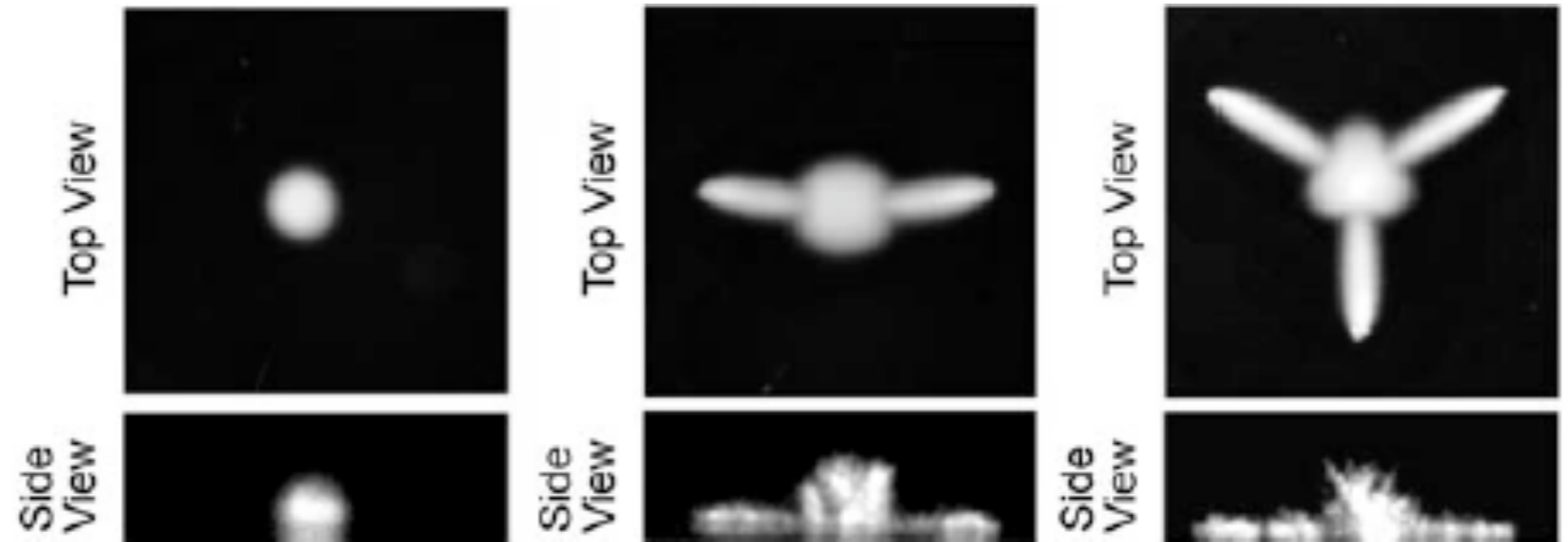


# Switching between low and high intensity

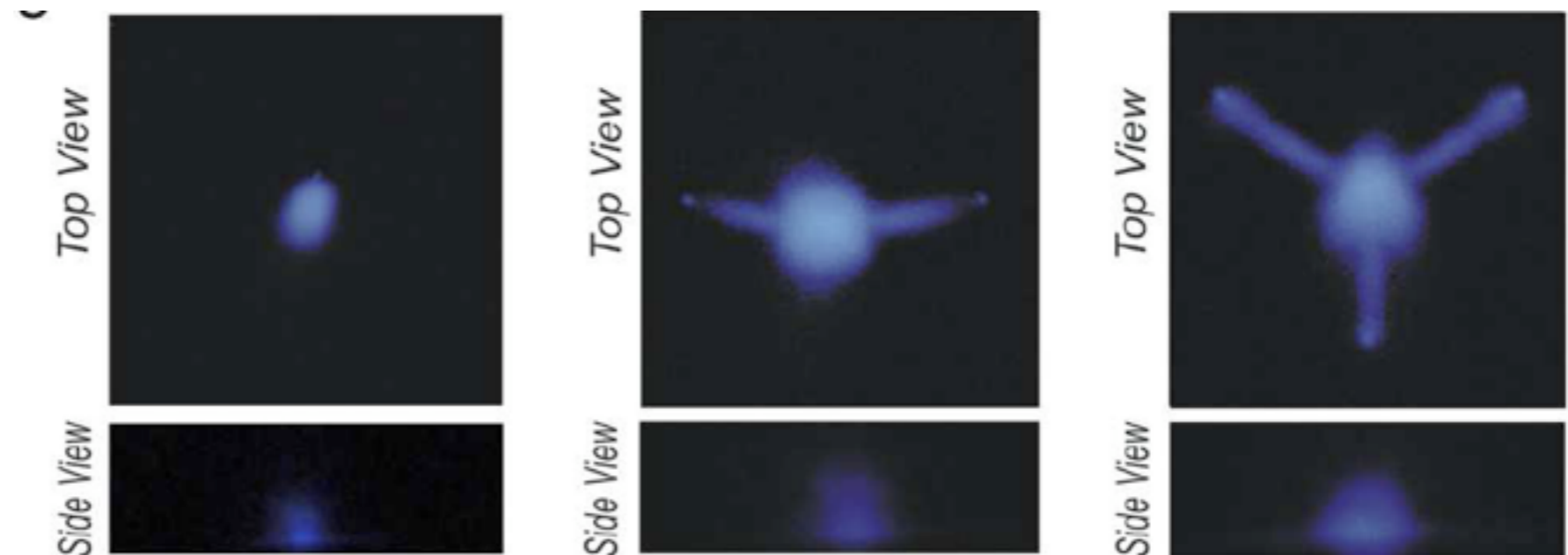


# Correlate light reflection & chemical activity of microbubbles emitted from micropits

Light reflection



Chemical activity





# Strongly driven multiple micropits

Micro-pit dimensions

$R=9.6 \mu\text{m}$

$H=20 \mu\text{m}$

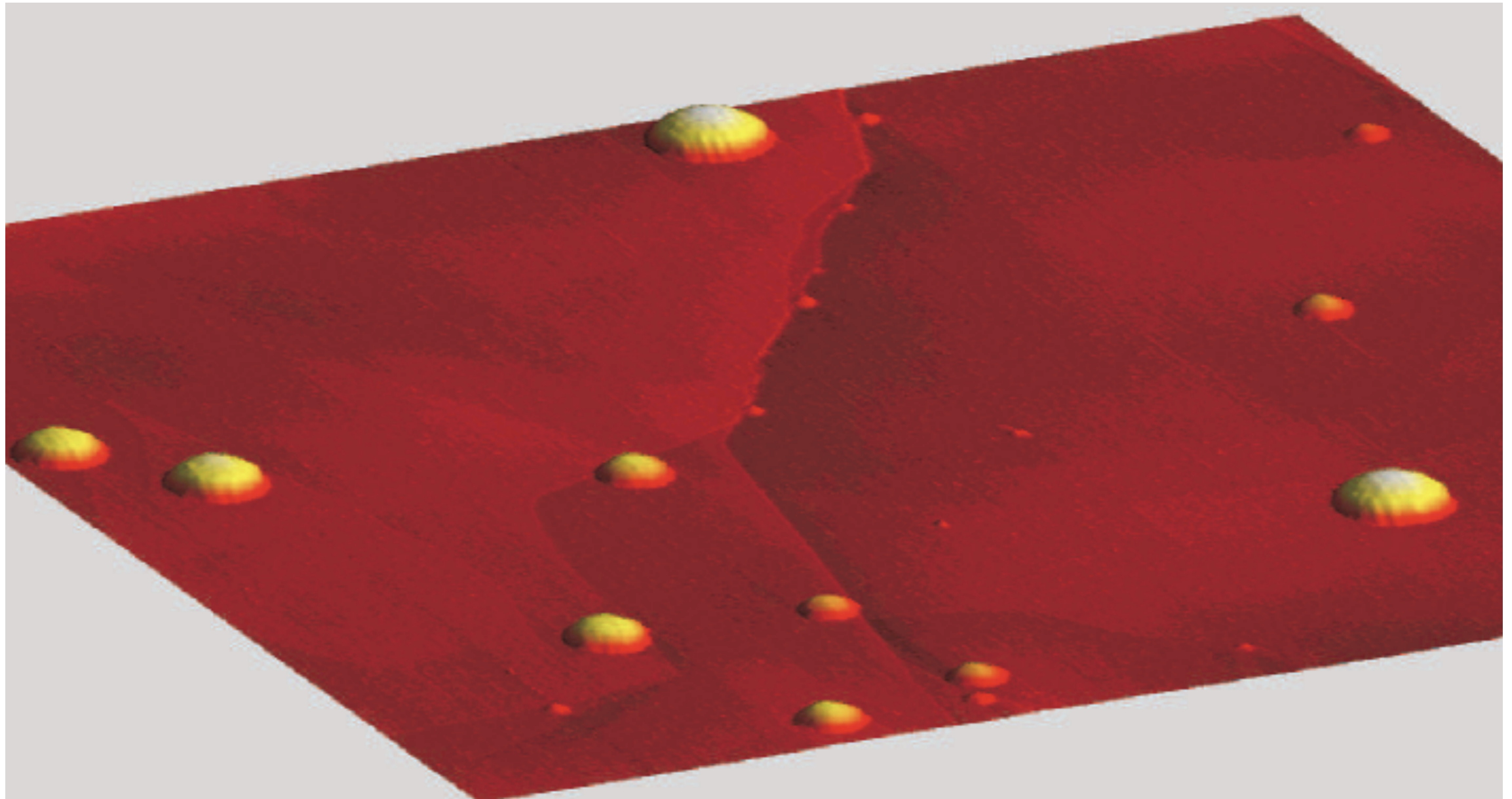
$f = 221 \text{ kHz}$

$f_r = 210 \text{ kHz}$



**Surface nanobubbles  
and controlled  
nucleation of droplets**

# Surface nanobubbles



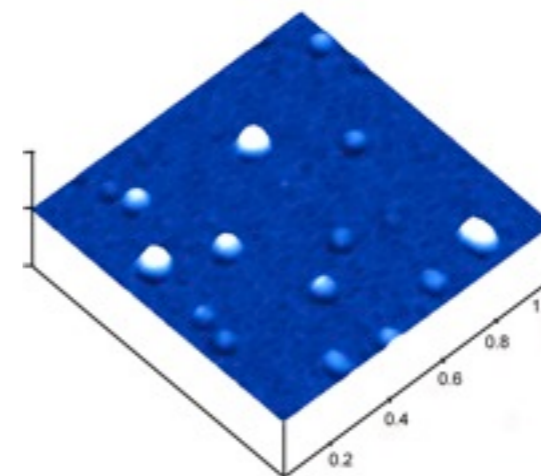
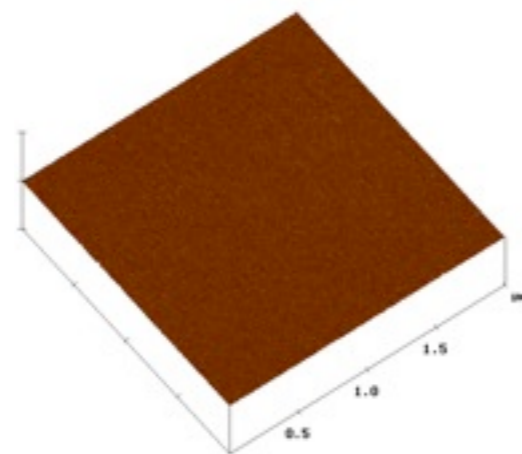
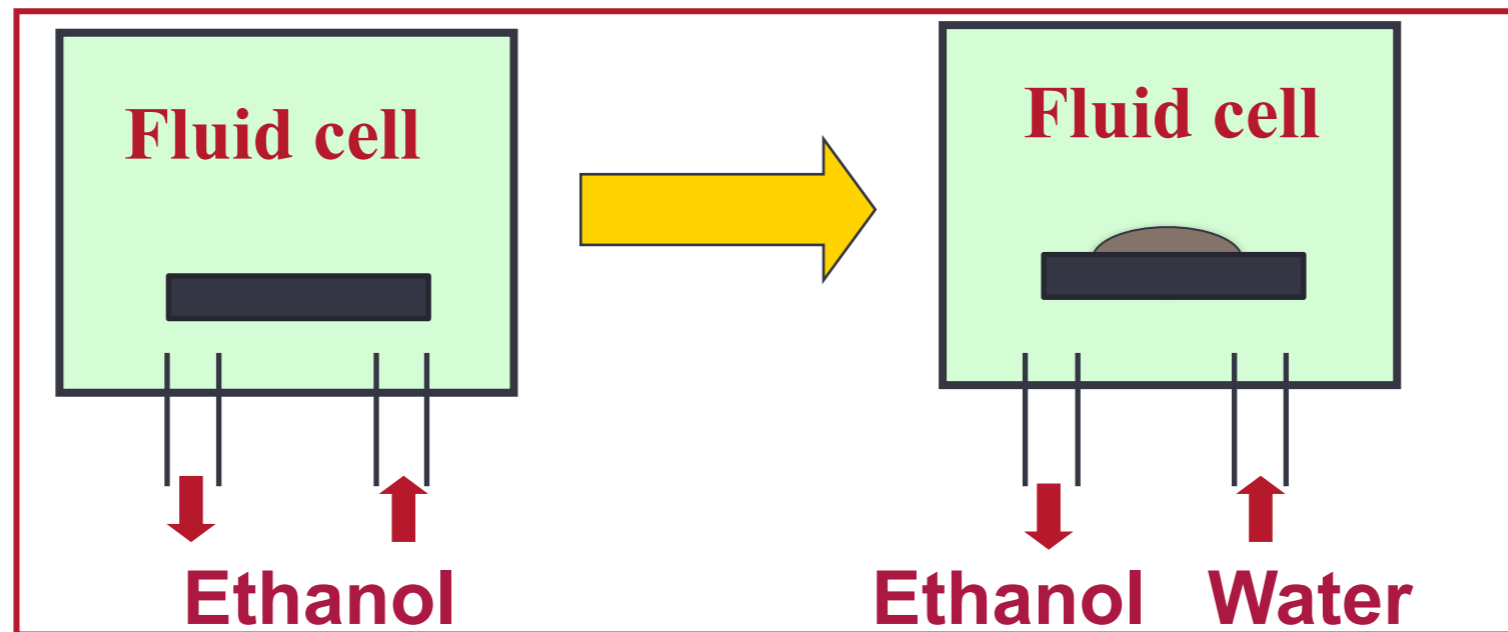
Why don't nanobubbles dissolve?

AFM

# How to easily produce surface nanobubbles?

Stage 1

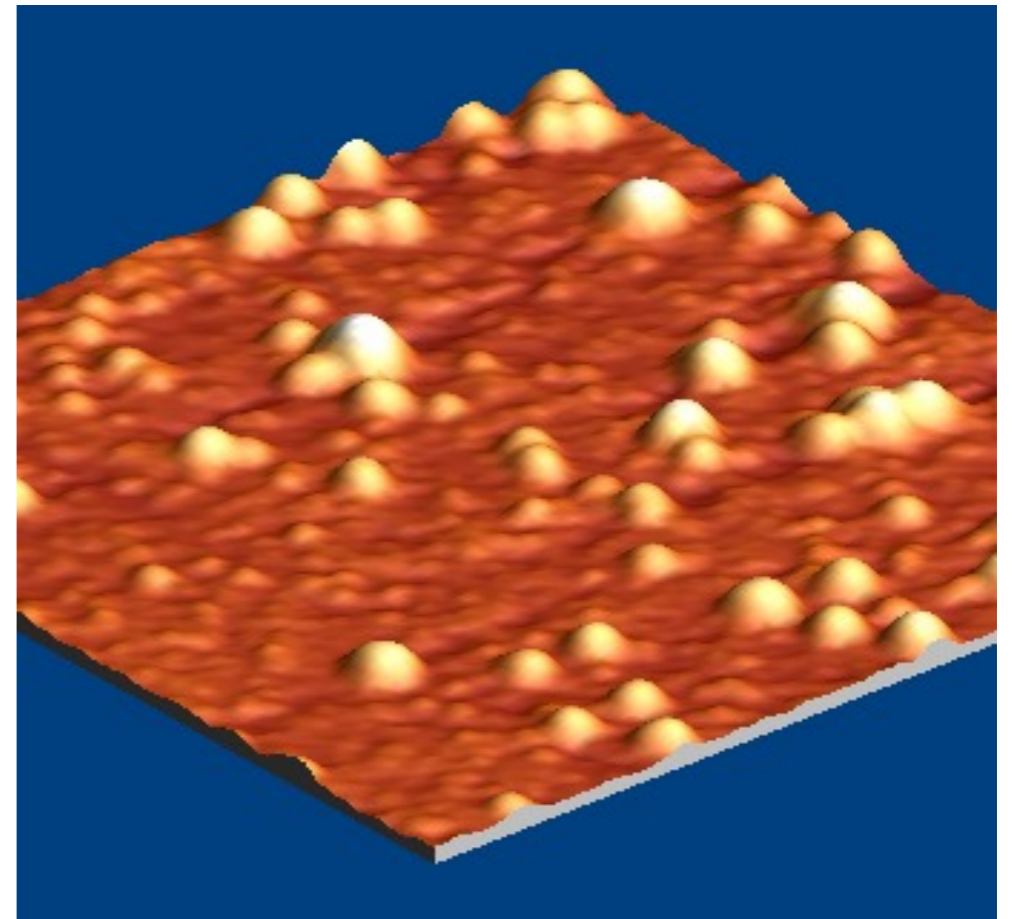
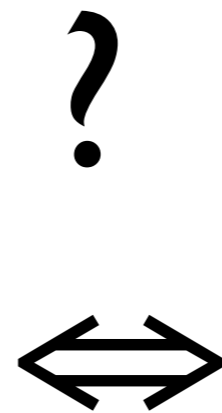
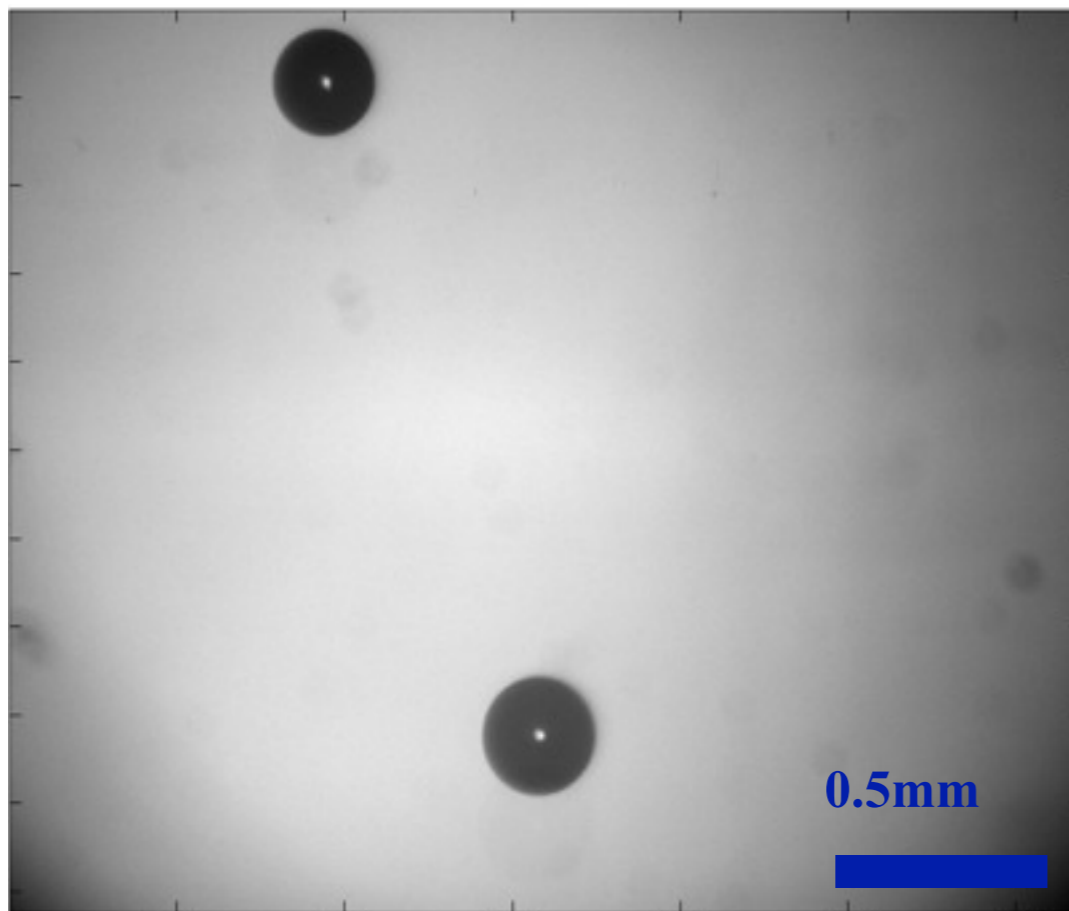
Stage 2



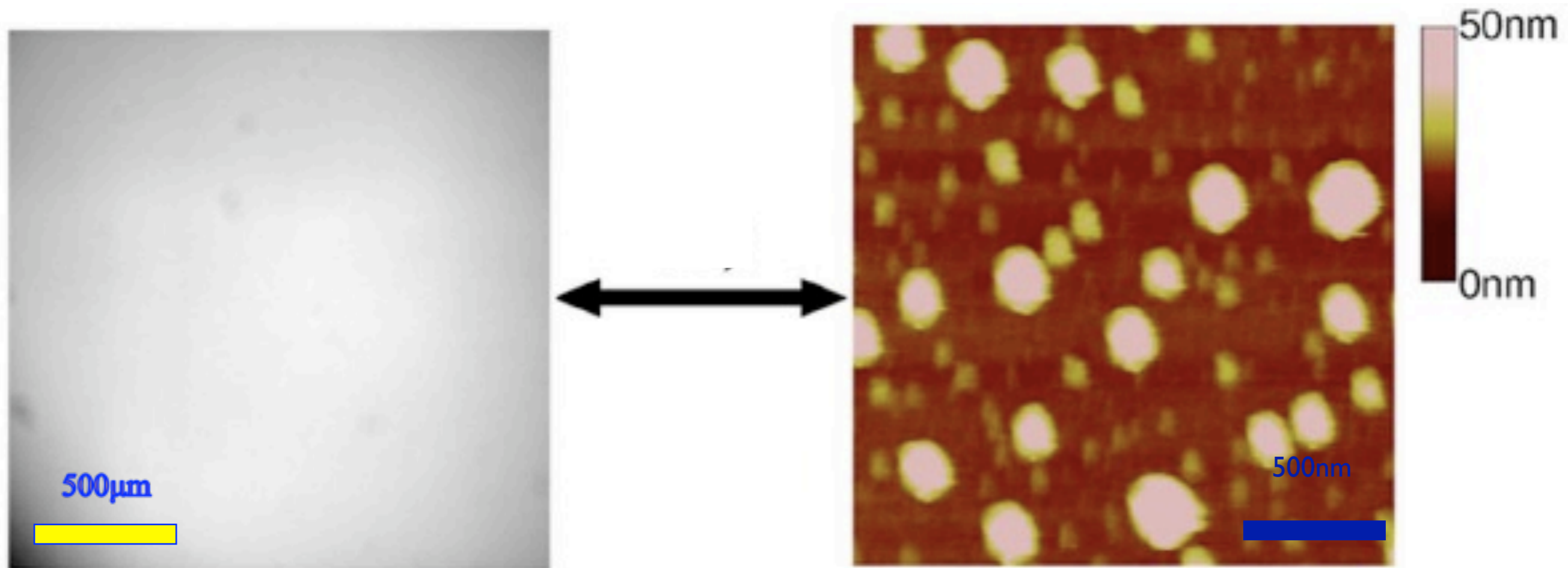
“Ethanol-water exchange”

*S. T. Lou et al., J. Vac. Sci. Tech.-B 18, 2573 (2001)*

# Do they cavitate?



# There is no correlation between nanobubbles and nucleated bubbles: Superstability of surface nanobubbles



(Lack of) cavitation  
activity

Nanobubbles in AFM

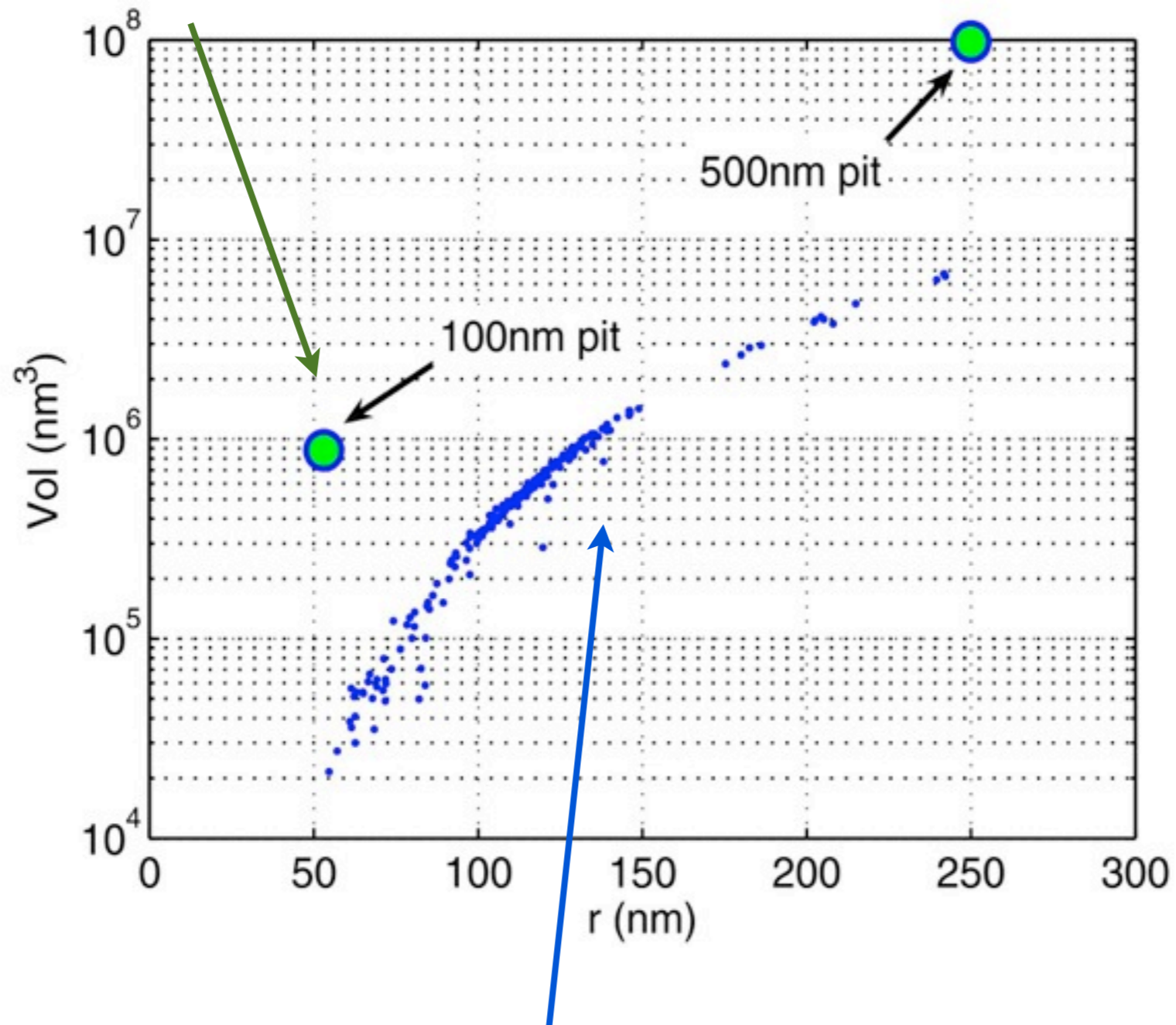
**There is no such correlation!**

**Nanobubbles even “survive” pressure  
reductions down to –60 bar!**

**“Superstability”!**



# Nanopits do nucleate bubbles



**Nanobubbles (of same size/volume)  
do NOT nucleate bubbles**

# Coworkers & funding

Bram Borkent

Wilco Bouwhuis

Nicolas Bremond

Henri Lhuissier

Hanneke Gelderblom

Stefan Gekle

Alvaro Marin

Devaraj van der Meer

Ivo Peters

Andrea Prosperetti

David Rivas

Jacco Snoeijer

Erikjan Staat

Chao Sun

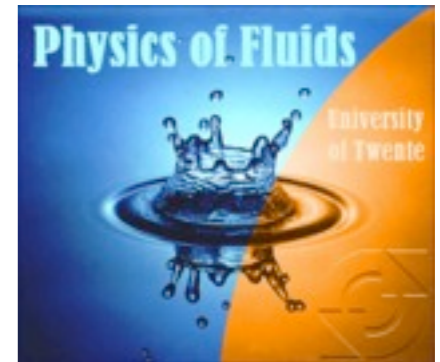
Peichun Amy Tsai

Tuan Tran

Roeland van der Veen

Koen Winkels

Xuehua Zhang



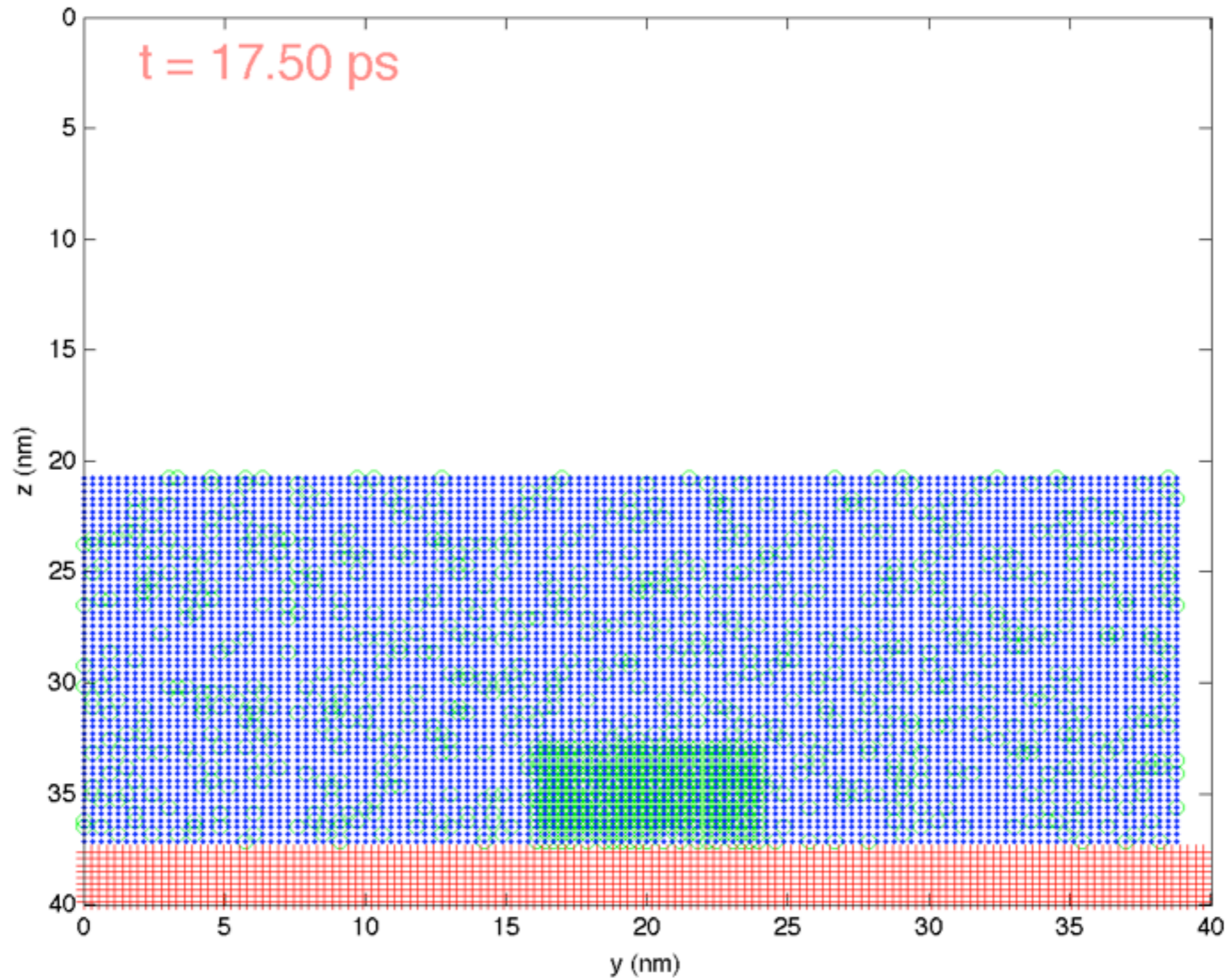








# Nanobubbles in MD simulations



# **Nanobubbles pin receding surface contact line and cause deposits on surface**

