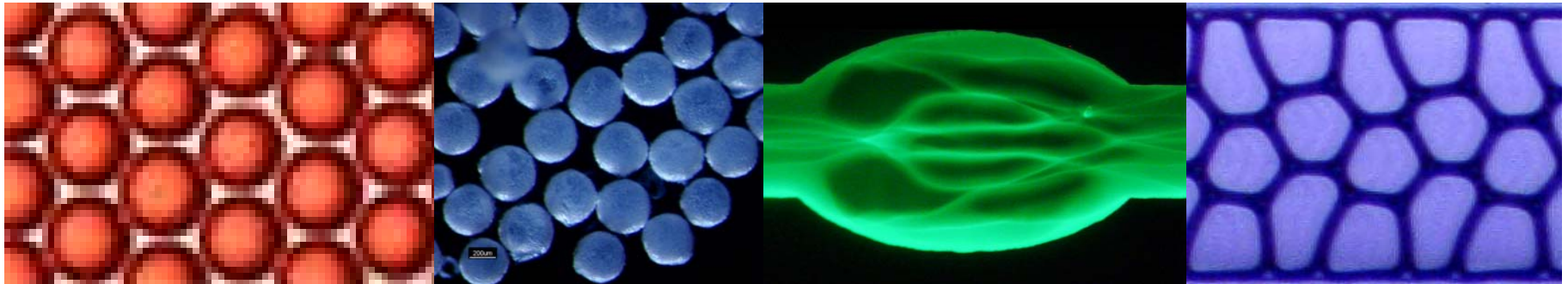


*bubbles and droplets in microfluidics:
formation, non-linear phenomena
and applications.*

Piotr Garstecki, Institute of Physical Chemistry, PAS

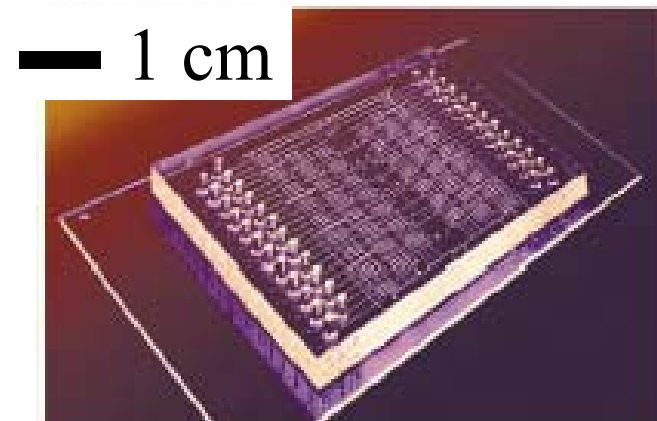


Institute of Fundamental Technological Research, 11/01/2006



- **microfluidics**
 - simple fluids
 - droplets and bubbles
- **formation of drops and bubbles**
 - flow-focusing
 - T-junction
- **stable oscillations with long periods**
- **time-reversible non-linear dynamics**
- **applications**
 - micromixing, portable assays
 - micro-particles and micro-capsules
 - diffraction gratings

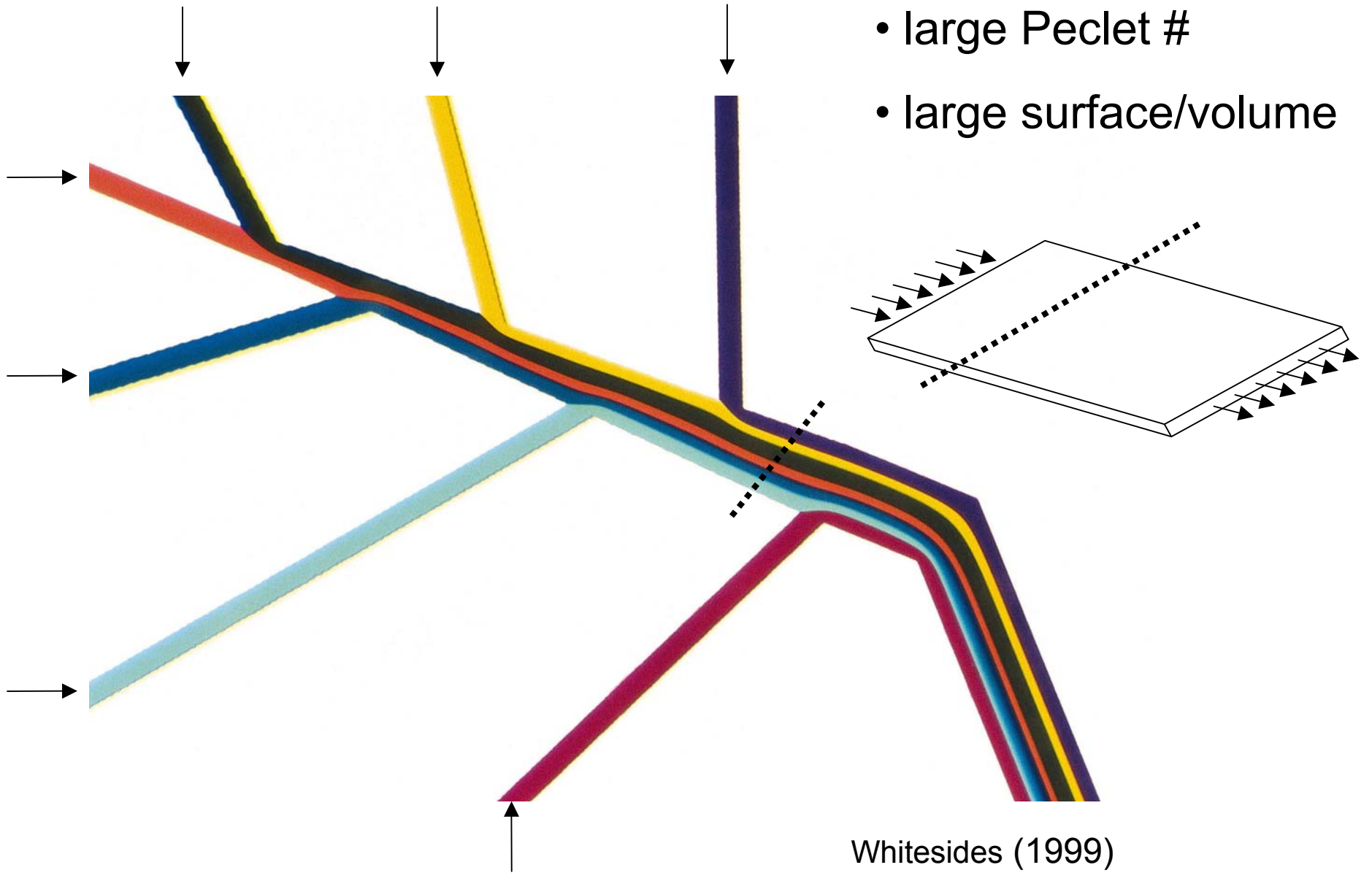
microfluidics



- small dimensions (10 – 100 μm)
- small rates of flow ($\sim 1 \mu\text{L/s}$)

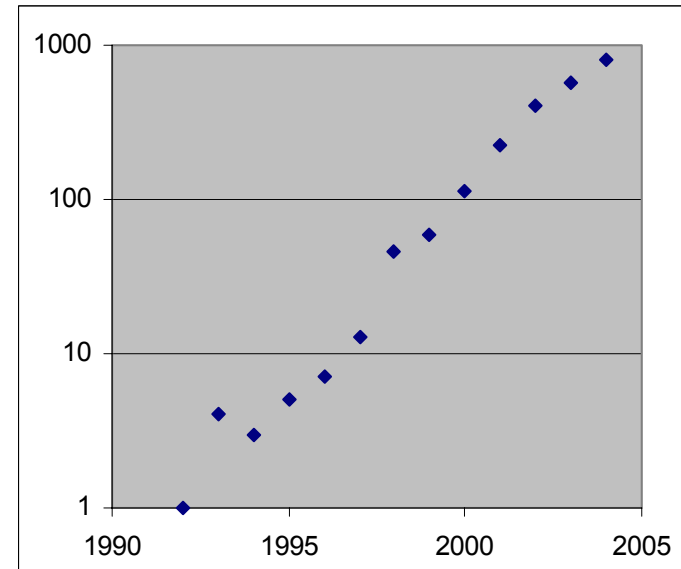
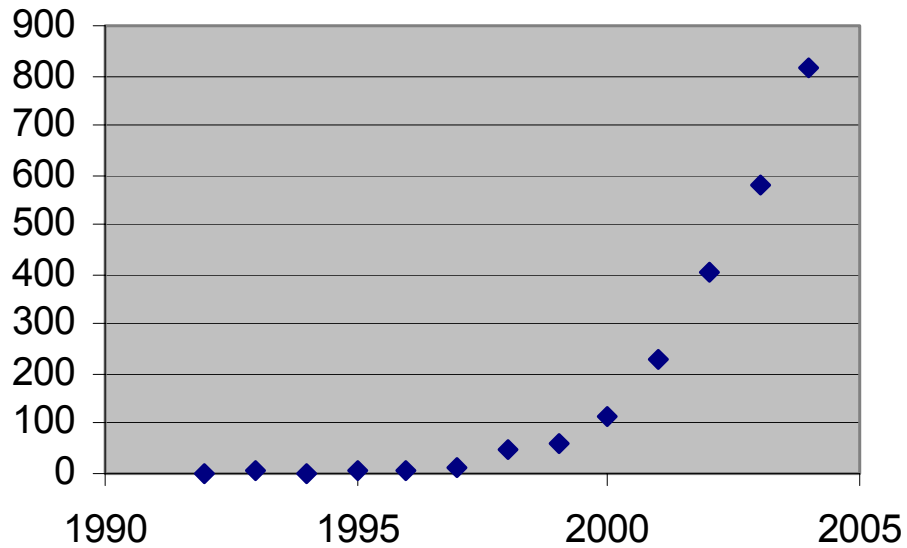
laminar flow

- small Reynolds #
- large Peclet #
- large surface/volume



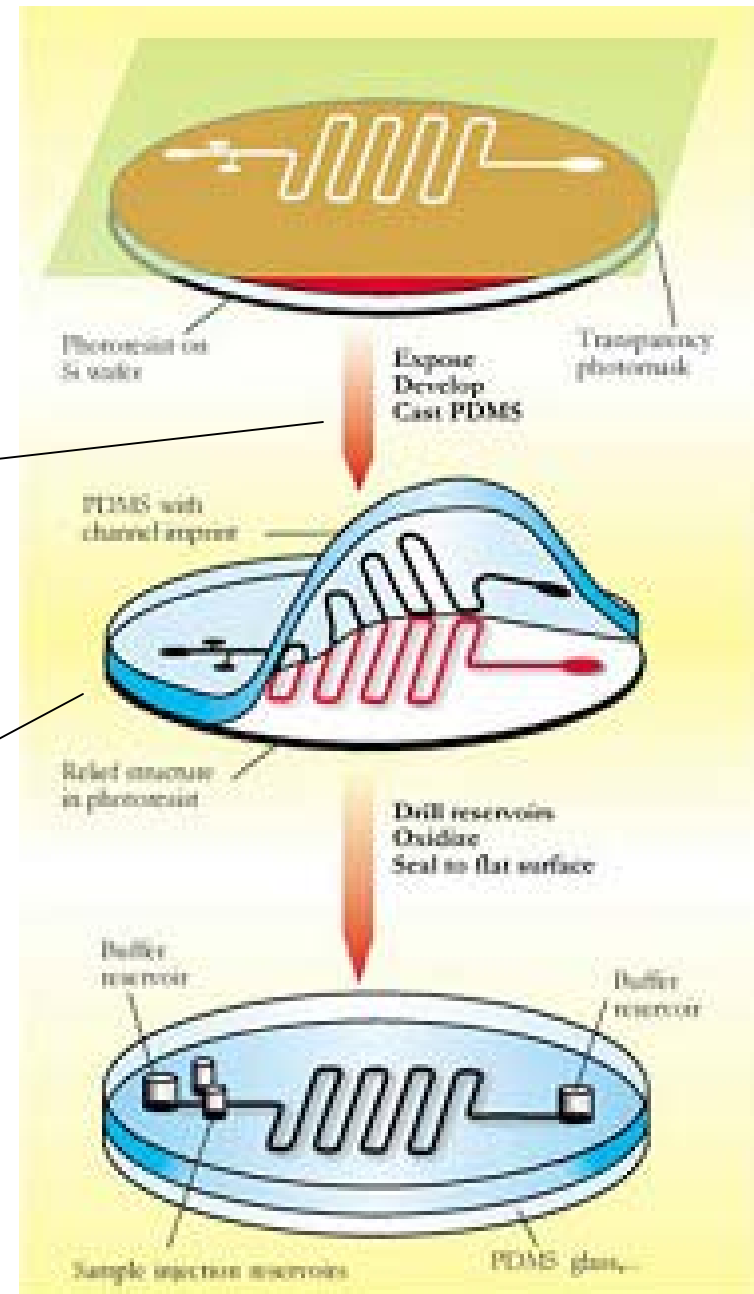
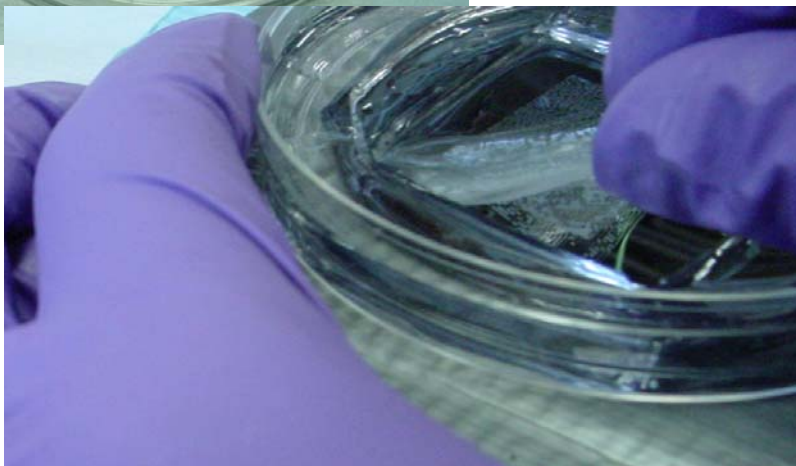
Whitesides (1999)

number of papers containing the term "microfluidics"



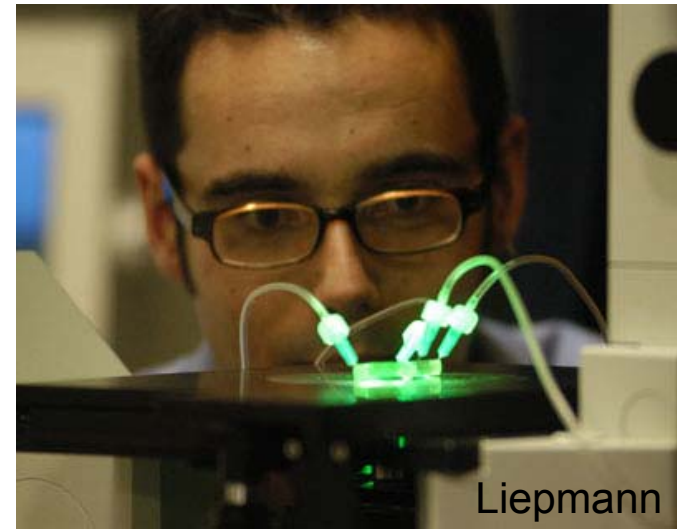
motivated by • **interest**
enabled by • **technology**

enabling technology –
– soft lithography
and rapid prototyping



planar geometries

rapid prototyping



design ~ 1 hour

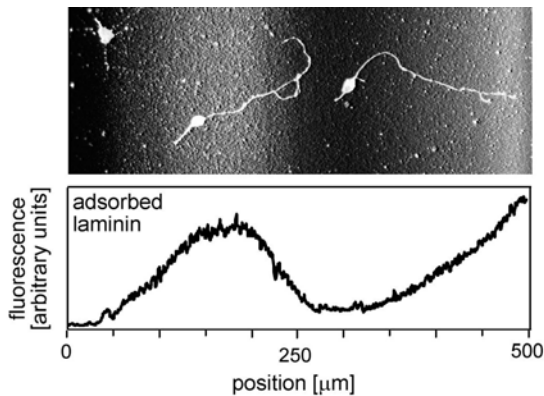
print out ~ 1 day

fab master ~ 3 hours ~ 2 days

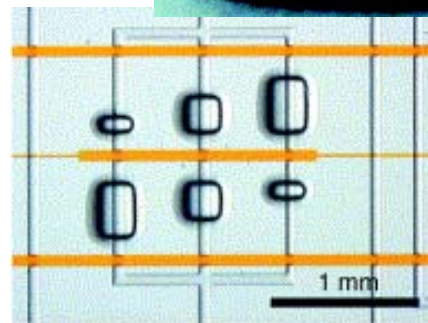
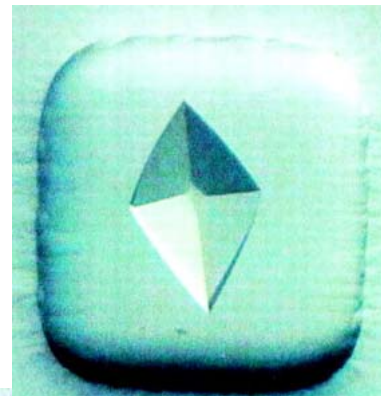
make copies of the device ~ 2 hours each

interest and applications

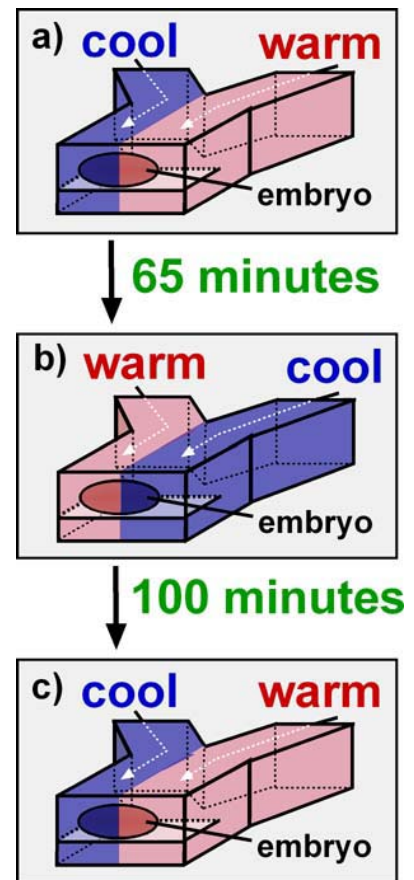
- chemistry (kinetics, organic / inorganic synthesis)
- drug design (hts)
- biotechnology (genomics, proteomics ...)
- material science
- physics – new flow phenomena
- biology (cell response)
- optics



Whitesides, Harvard

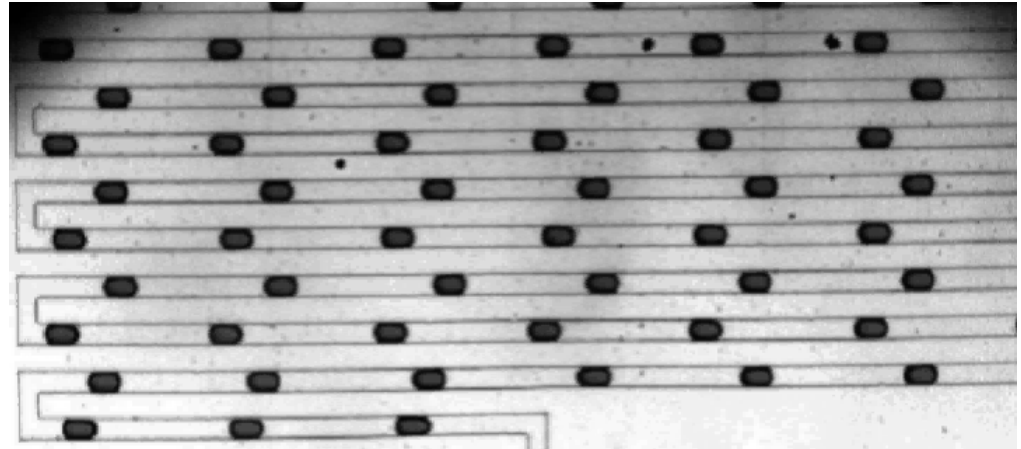


Quake, Caltech



Ismagilov, UChicago

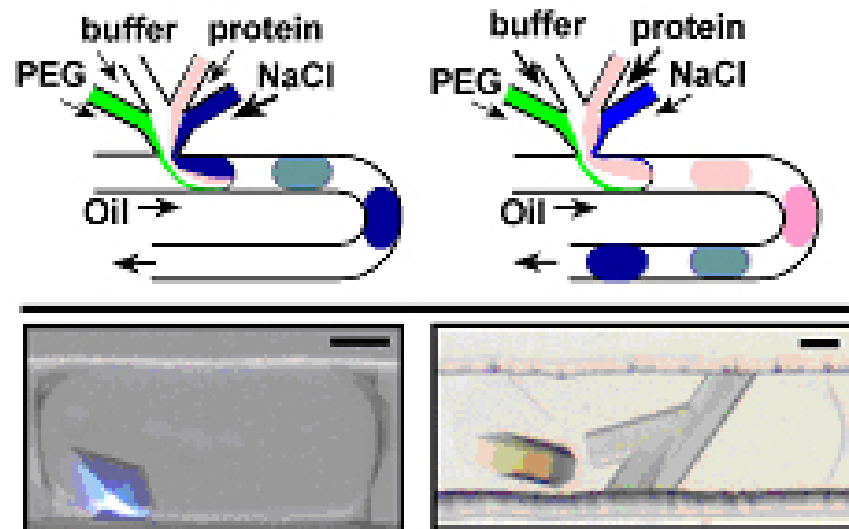
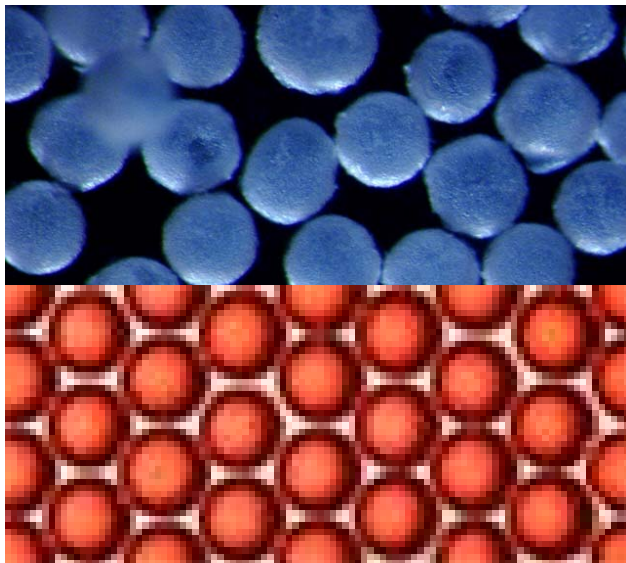
microfluidics with drops



applications



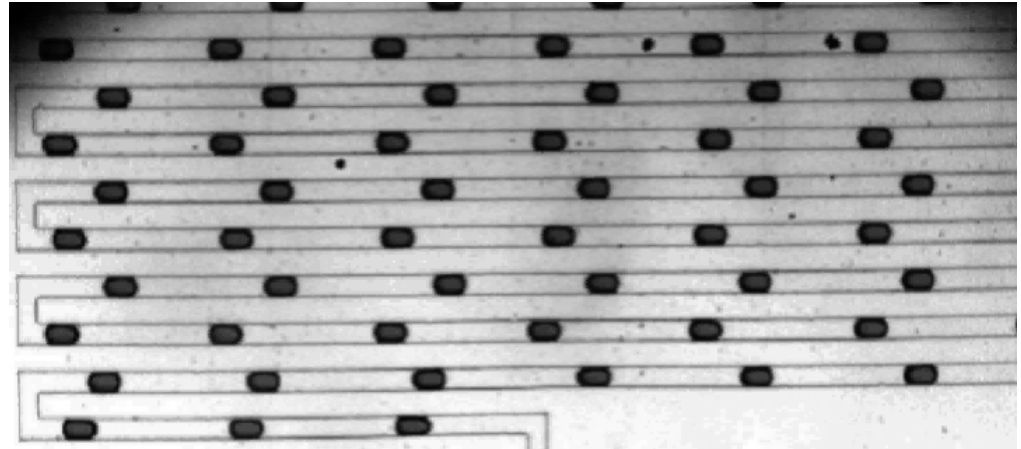
- controlled emulsification
- droplet as a beaker
 - aqueous chemistry
 - biochemistry
 - organic chemistry
- processing / screening / kinetics
- material synthesis



Thaumatococcus

Catalase

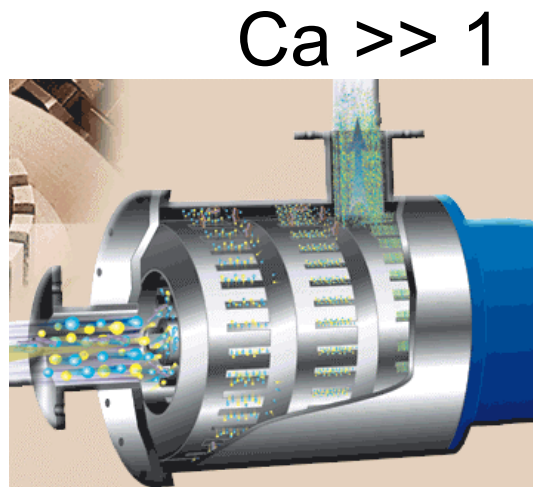
microfluidics with drops



- how do you make drops in a controlled way?
- how do you guide them through networks?

- **microfluidics**
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- **formation of drops and bubbles**
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emulsification



Fluko

$We \gg 1$



$Bo > 1$



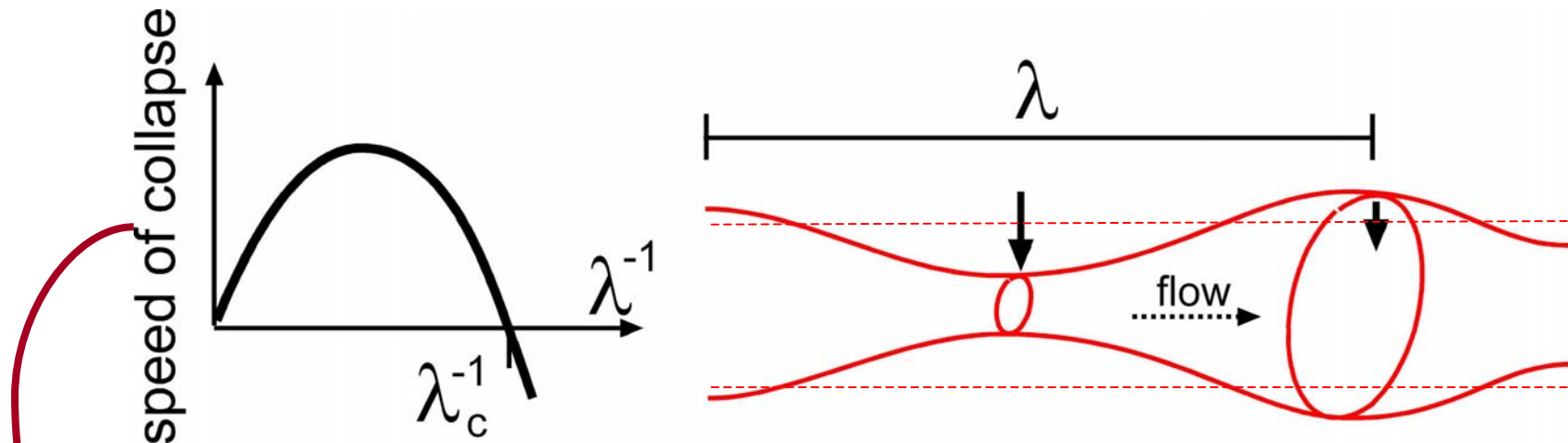
instability

capillary number (Ca) = **viscous / interfacial**

Weber number (We) = **inertial / interfacial**

Bond number (Bo) = **gravitational / interfacial**

Rayleigh-Plateau instability



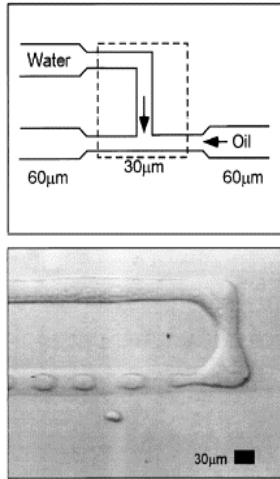
$u \sim \gamma / \mu$ viscous dynamics
 $u \sim (\gamma / \rho l)^{1/2}$ inertial dynamics

→ typical size $\sim 1/\lambda$

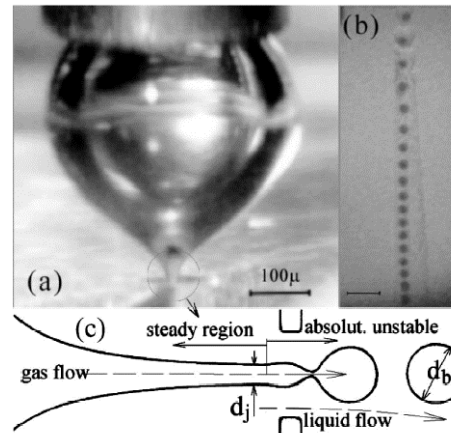
→ typically broad size distribution



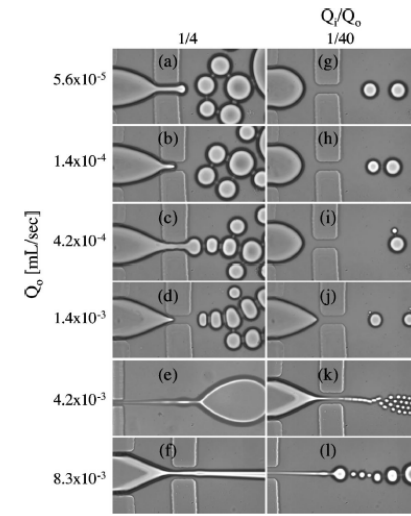
micro emulsification



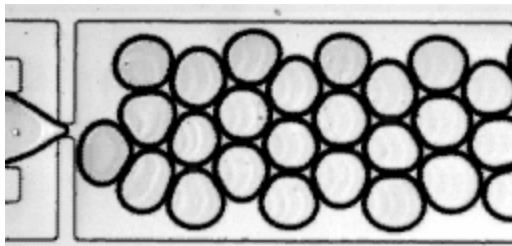
Thorsen (2001)



Ganan-Calvo (2001)



Anna (2003)

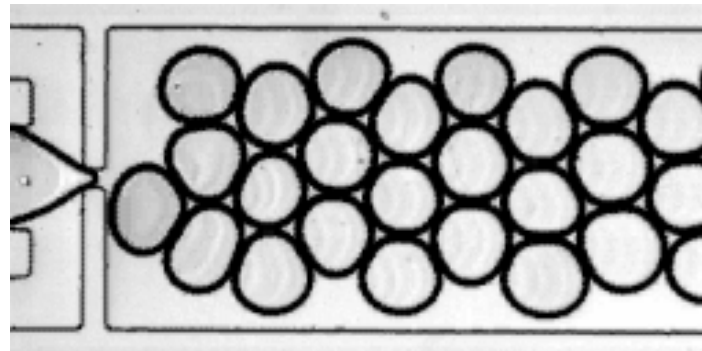
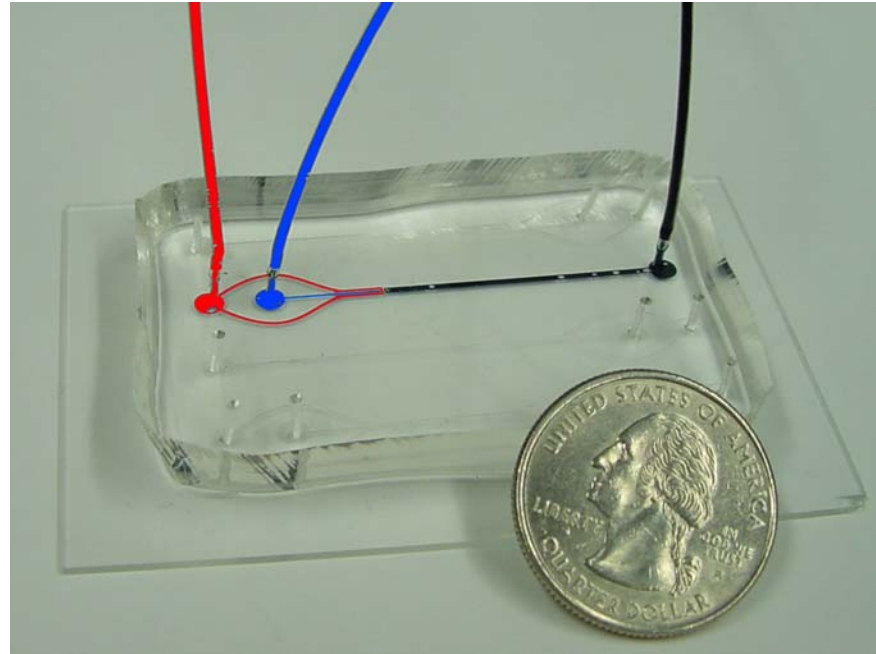


Garstecki (2004)

- liquid/liquid & gas/liquid
- possible to obtain narrow size distributions

flow focusing

water ↓ air ↓ ↑ out

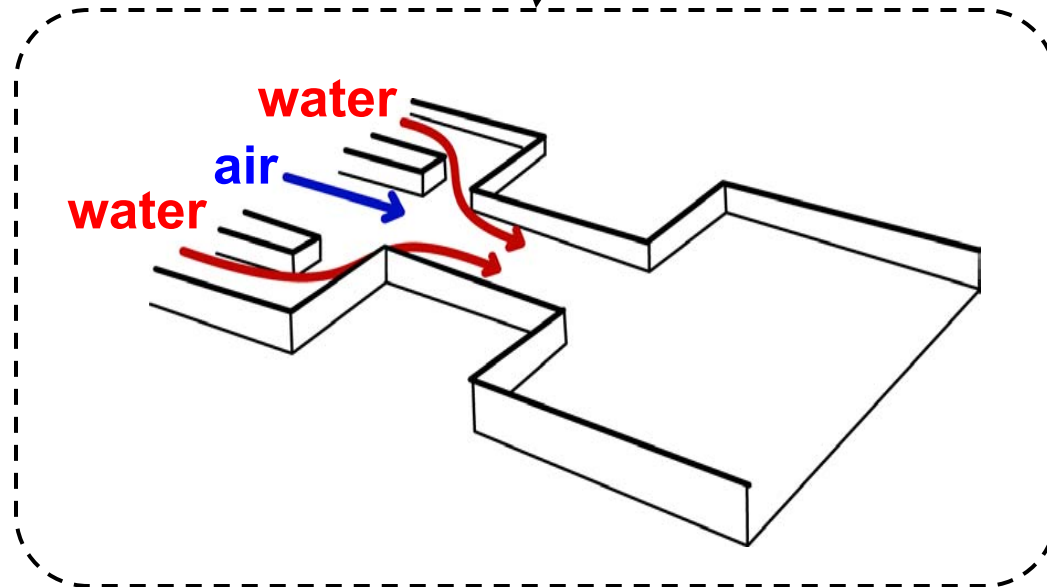
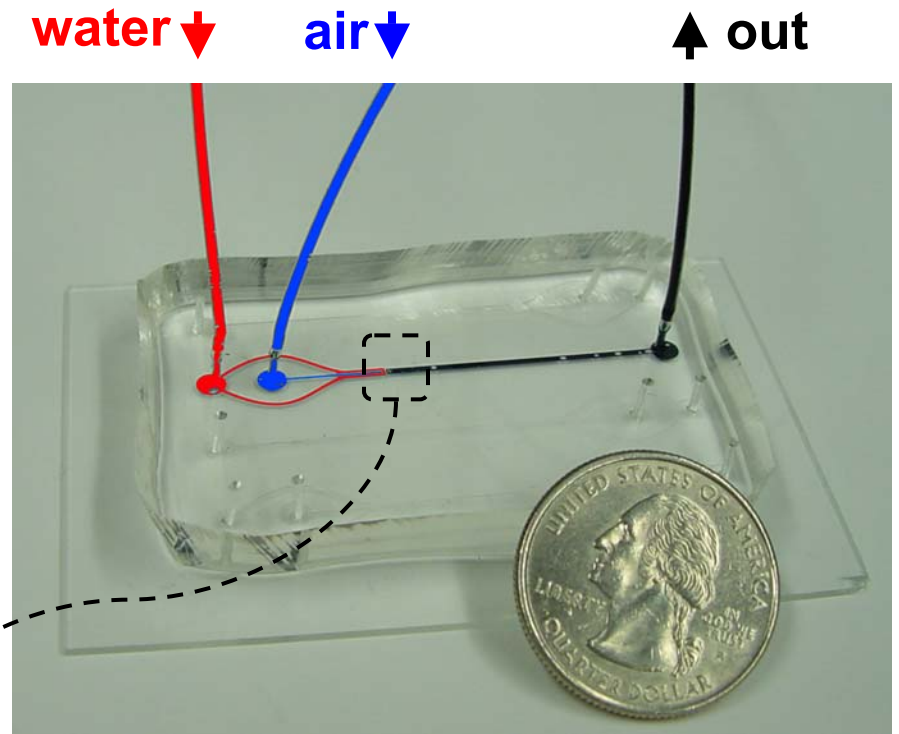


flow focusing

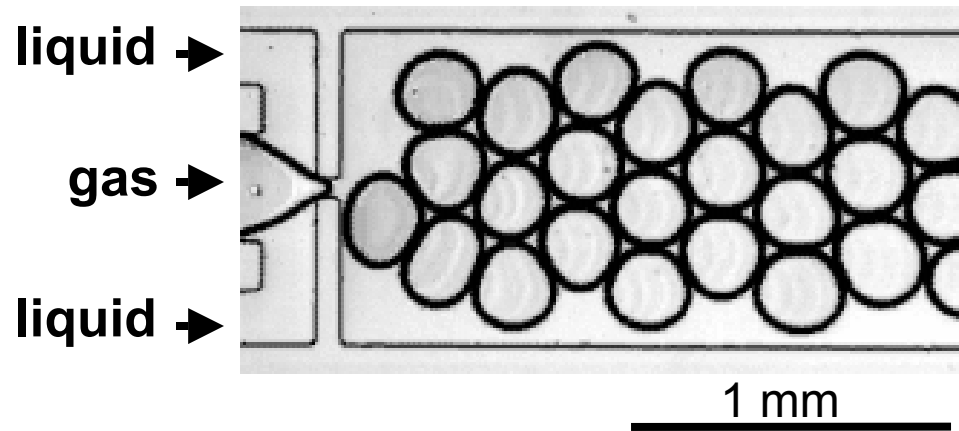
confined (planar) geometry

inlets:

- rate of flow of the liquid (Q)
- pressure app. to gas stream (p)



results



bubbles:

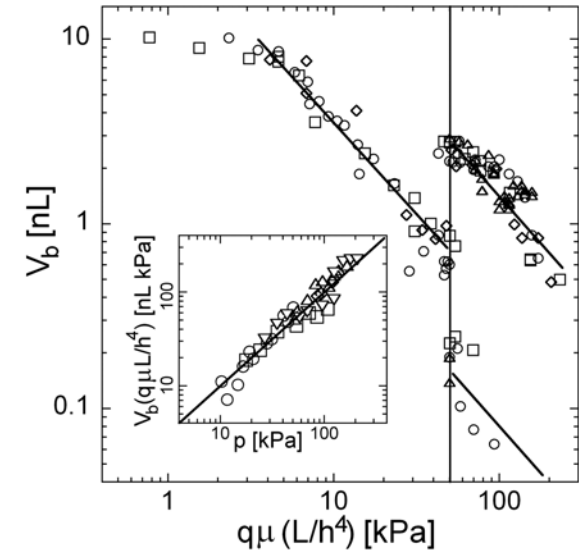
- size: 10 – 1000 μm
- standard deviation < 5 %
- volume fraction: 0 – 100 %

scaling

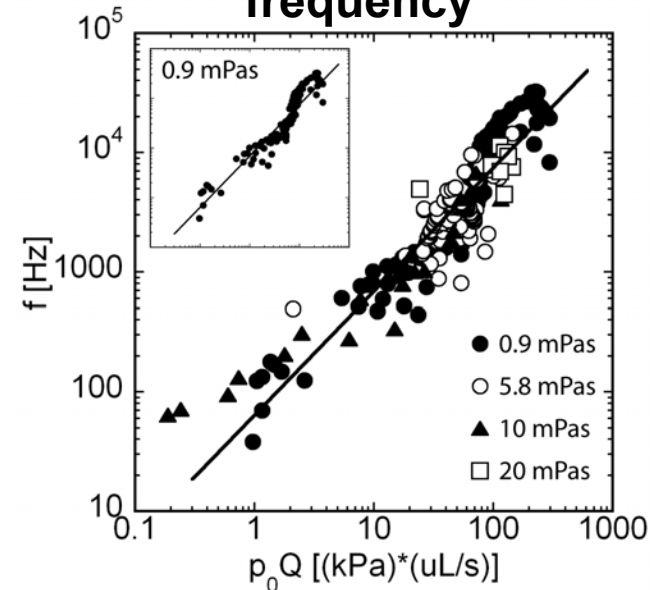
$$V_b \propto p/Q\mu$$
$$f \propto pQ$$

simultaneous,
independent control
of the
size and **volume fraction**

volume of the bubble



frequency



questions

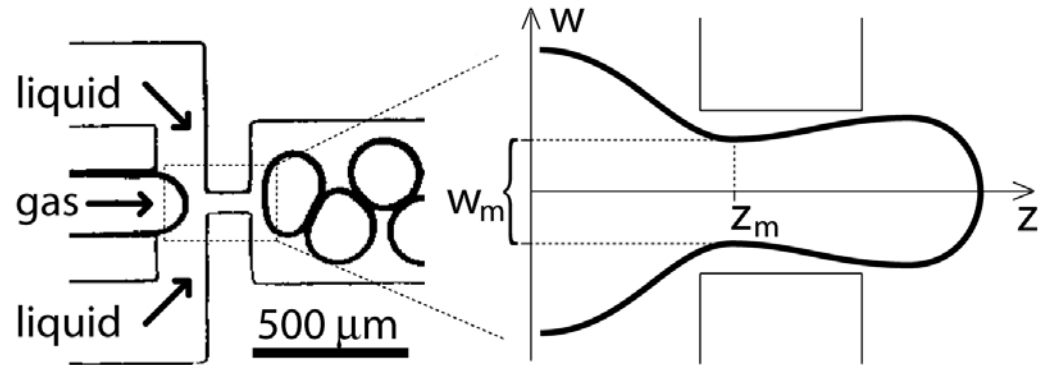
$$V_b \propto \rho/Q\mu$$
$$f \propto \rho Q$$

**no dependence
on surface tension**

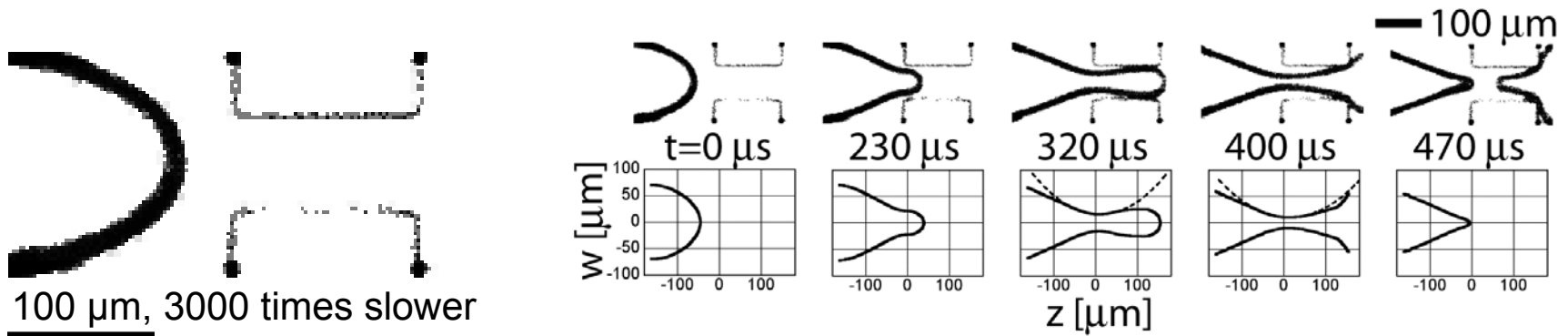
Ca: 10^{-3} to 10^{-1}

- **why** doesn't surface tension come into the equations?
- **what** is the mechanism of break-up?

break-up: evolution of the interface

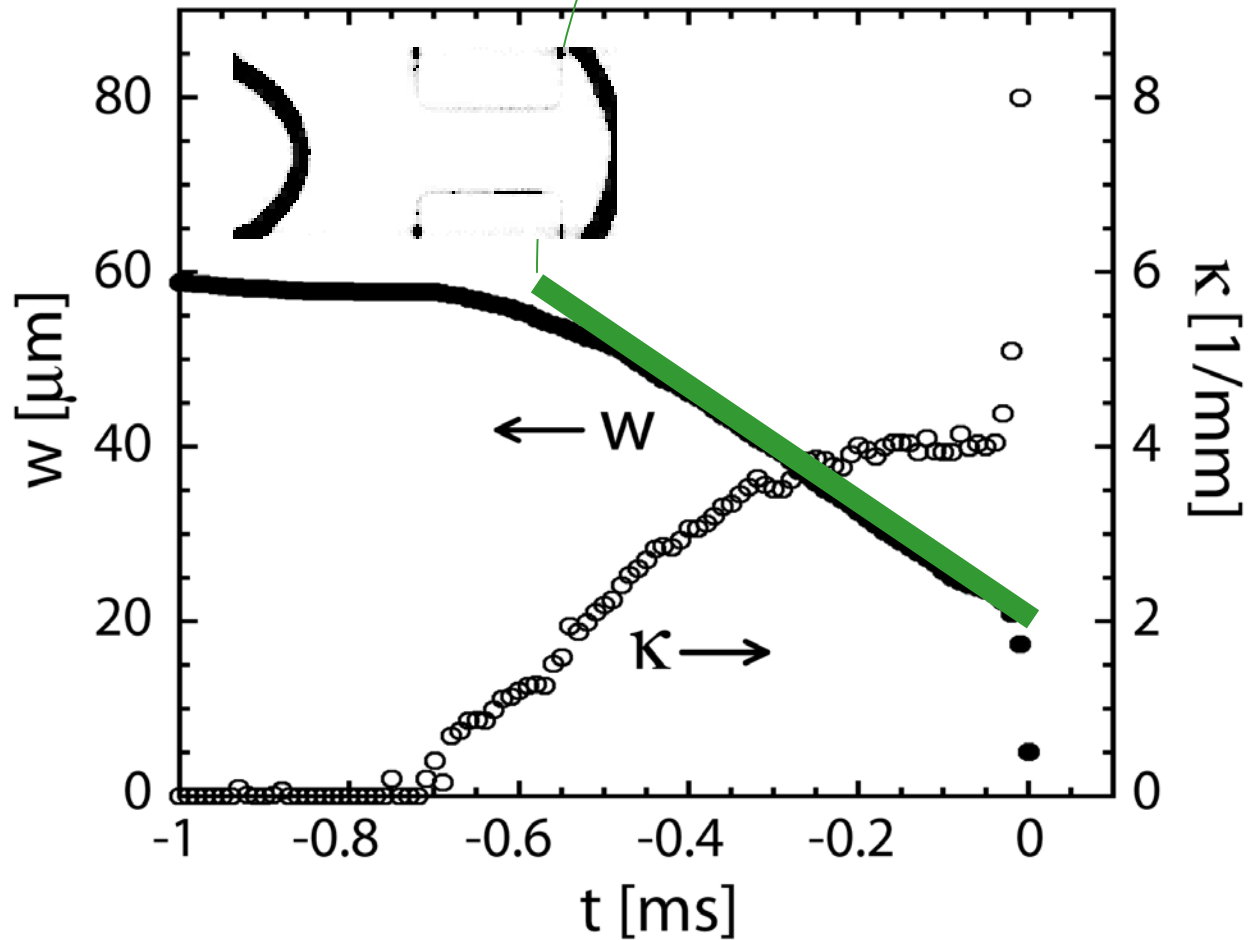


recording and image analysis

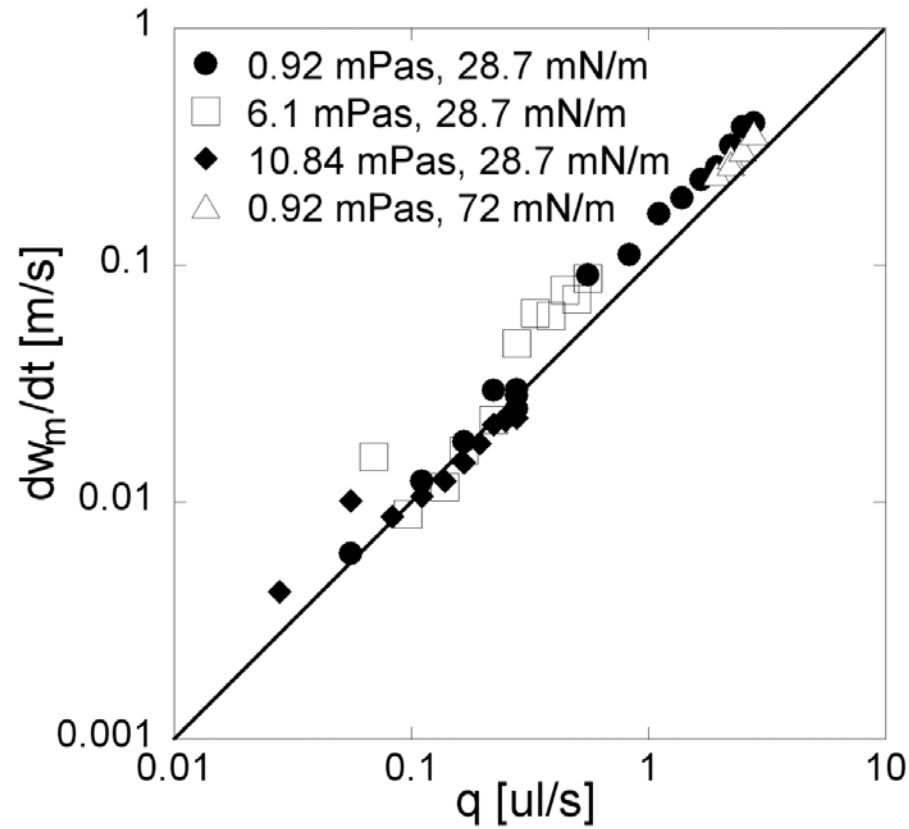


single break-up

“speed of collapse”
 $dw/dt = f(Q, \mu, \rho, \gamma)$



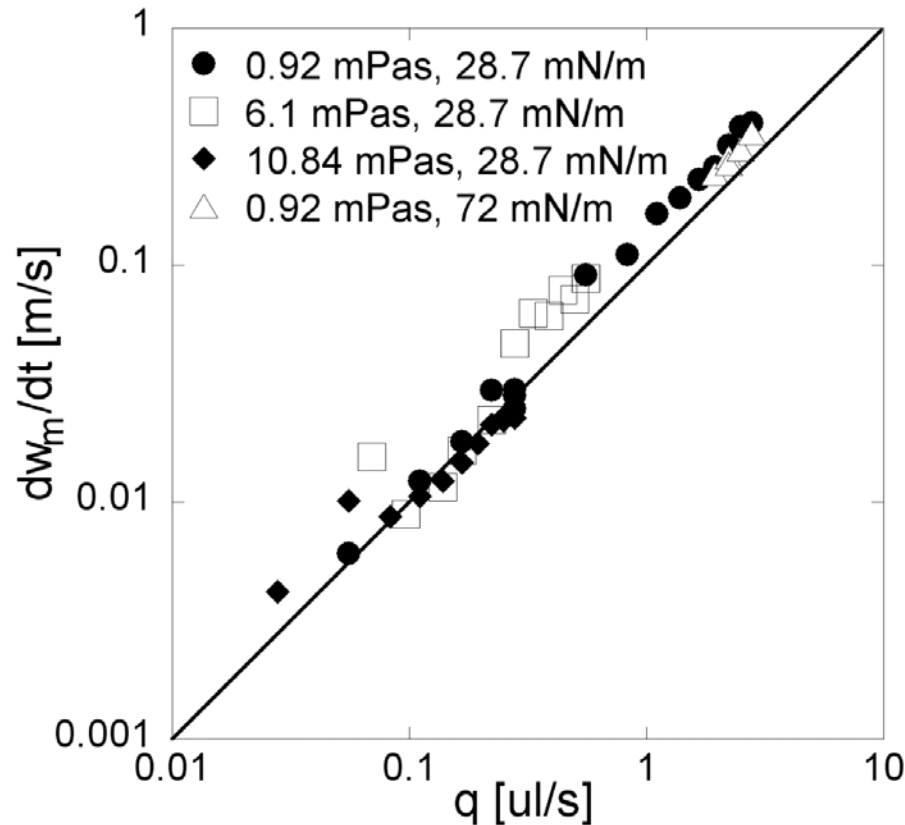
speed of collapse



$dw/dt =$

$f(Q, \mu, \rho, \gamma) =$

speed of collapse



$$dw/dt =$$

$$f(Q, \mu, \rho, \gamma) = \alpha Q$$

‘interfacial’ speeds:

viscous regime

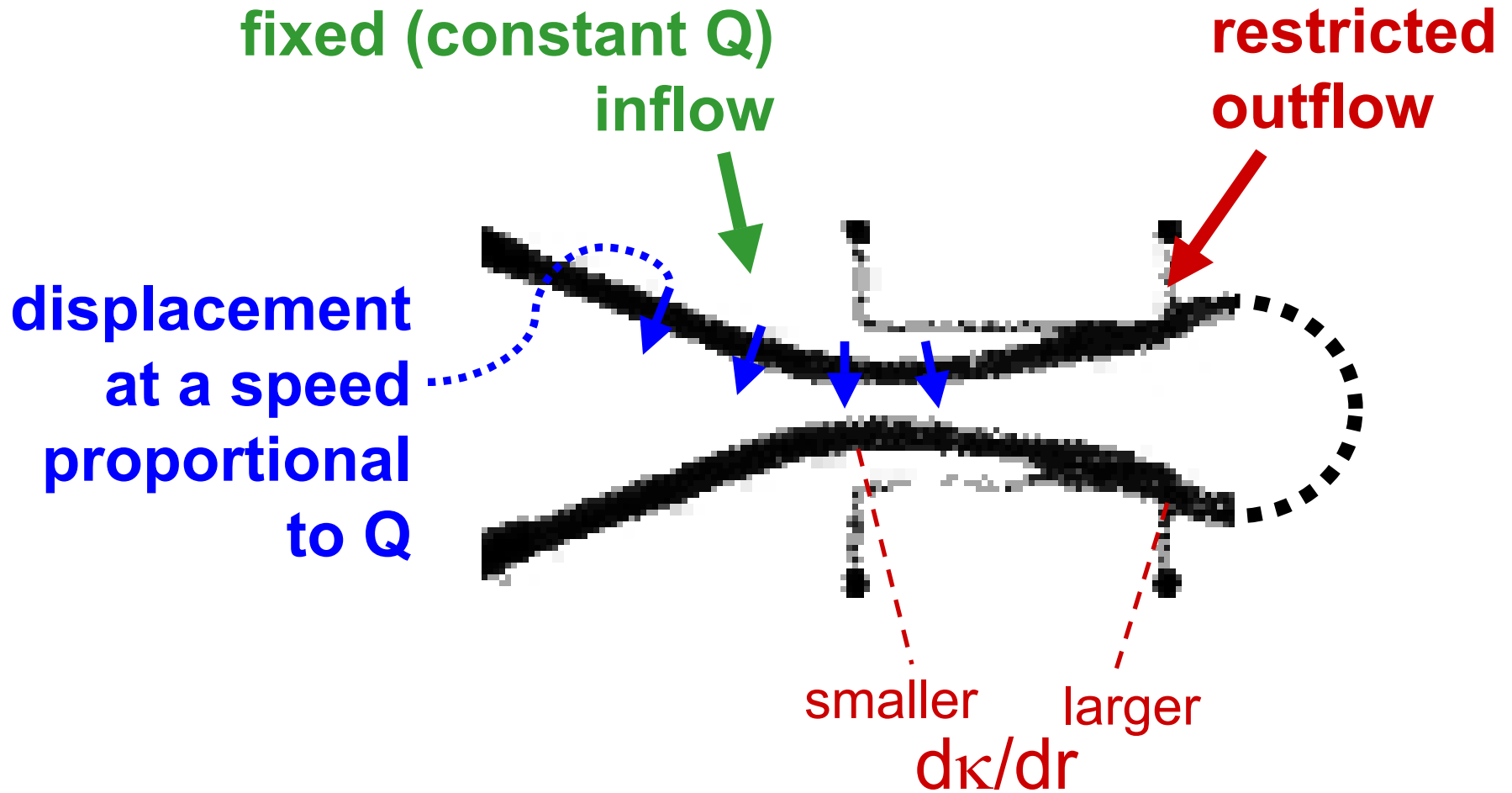
$$u \sim \gamma/\mu \sim \mathbf{10-100 \text{ m/s}}$$

inertial regime

$$u \sim (\gamma/\rho l)^{1/2} \sim \mathbf{1-10 \text{ m/s}}$$

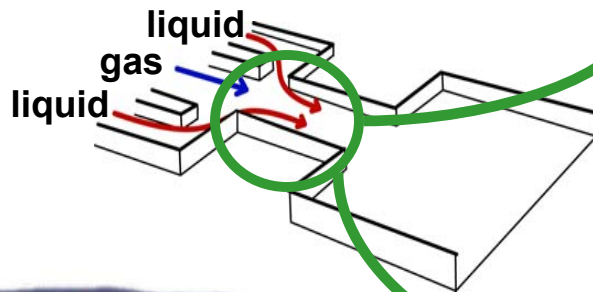
- the **speed depends only on the rate of flow** of the continuous fluid
- the **evolution is much slower** than the speed of a capillary wave
collapse is *not* driven by surface tension

'squeezing'

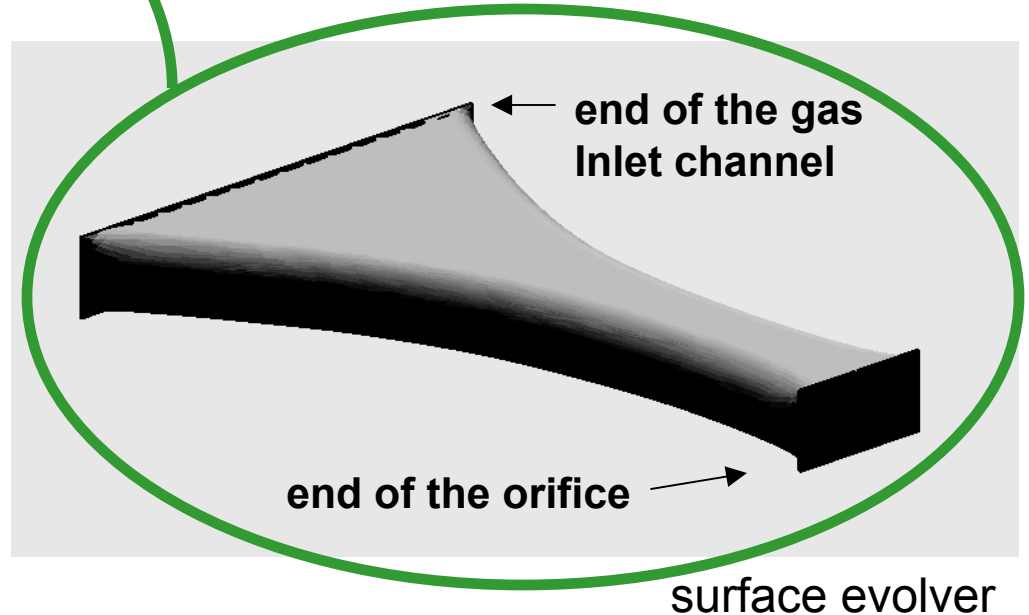
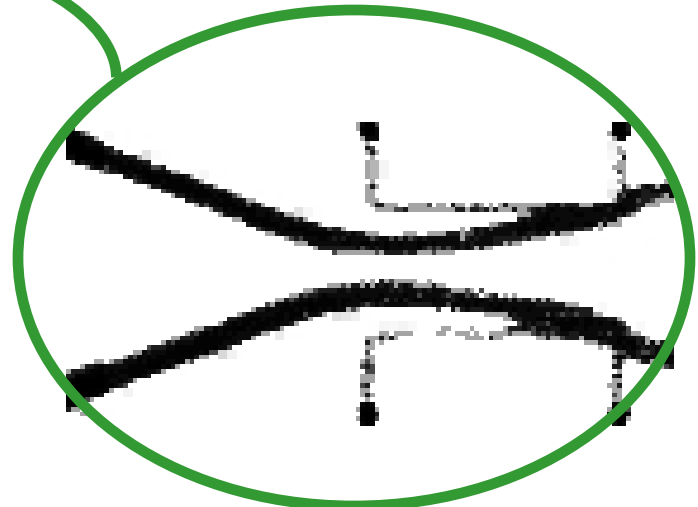


but, is the thread stable?

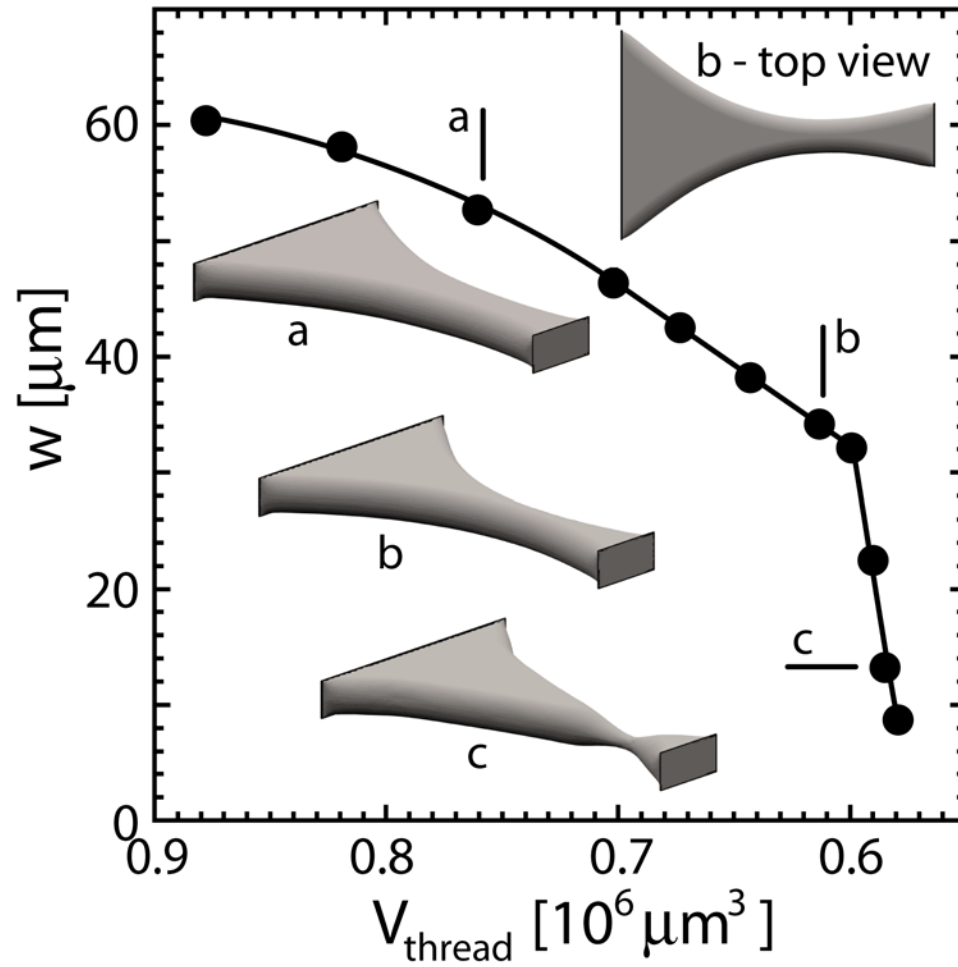
confinement



equilibrium shape for a given volume enclosed by the gas-liquid interface



equilibrium

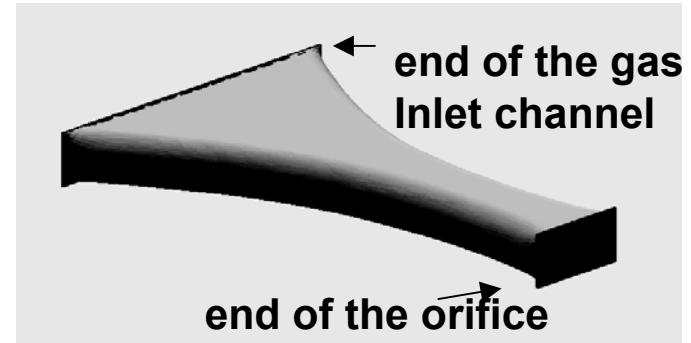


no dynamics here

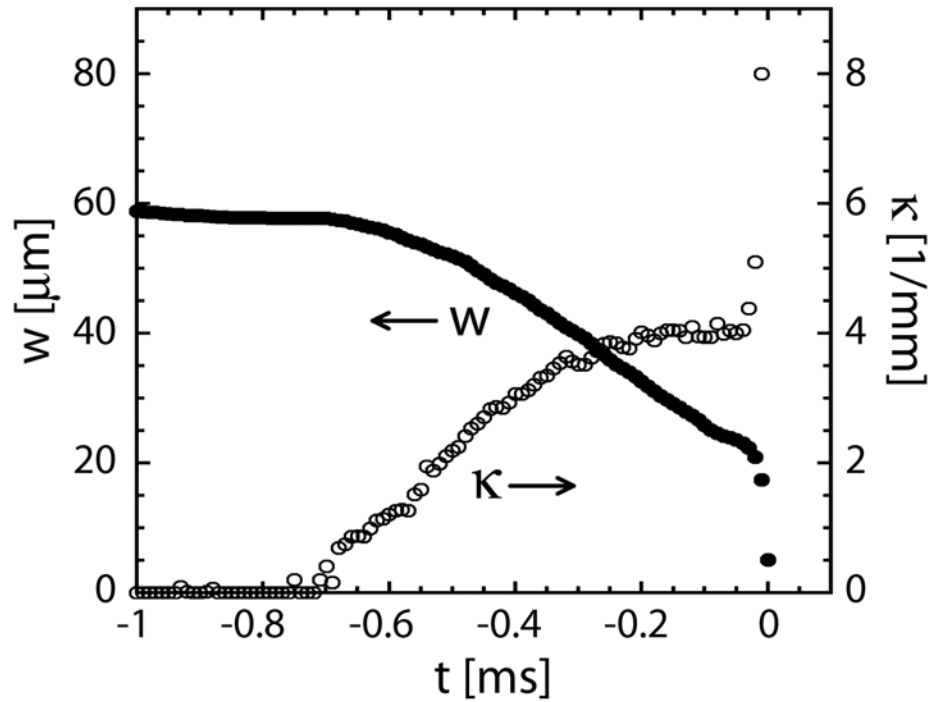
experiment



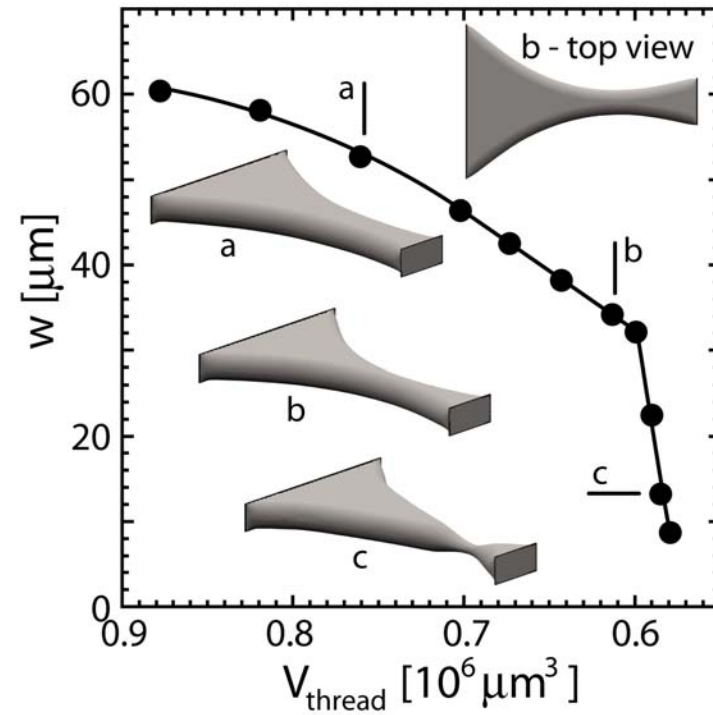
simulation



surface evolver

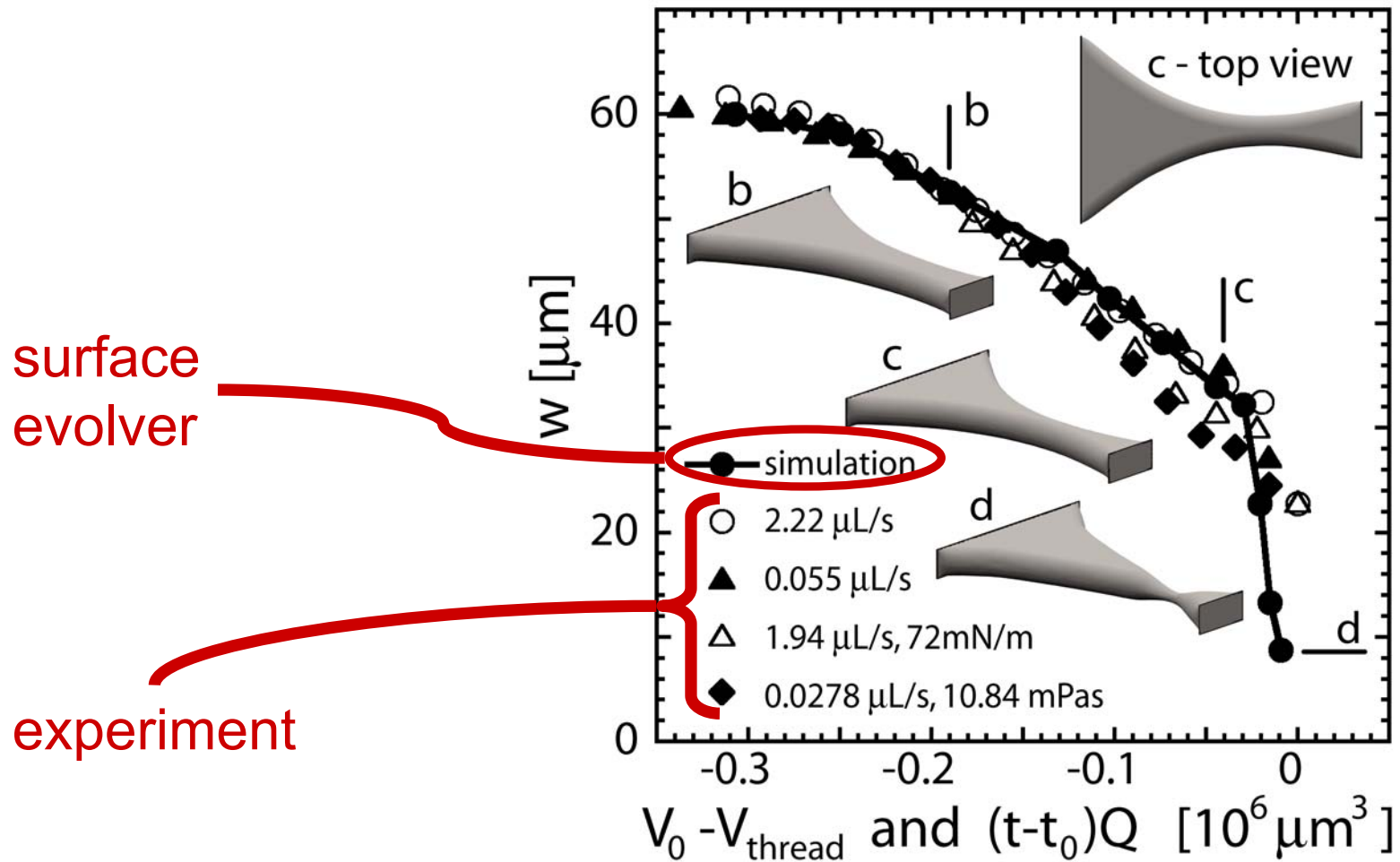


dynamics



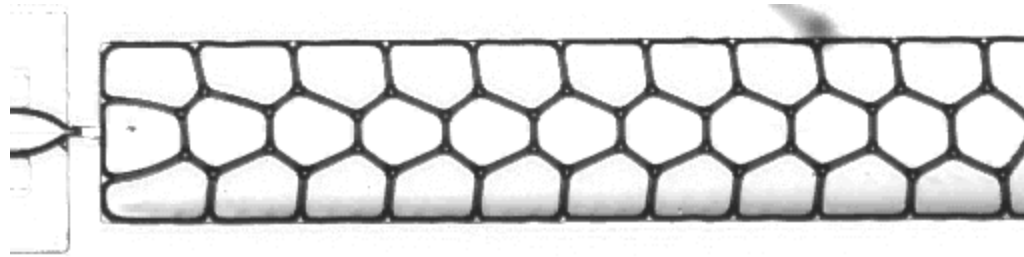
equilibrium

quasi-stationary break-up



break-up follows through a series of equilibrium states

bubble growth rate



resistance to flow in the outlet channel: $R \propto \mu$

rate of inflating the bubble: $Q_{\text{gas}} \propto p/\mu$

volume of the bubble

$$V_b \propto t_{\text{open}} Q_{\text{gas}}$$

$$V_b \propto (1/Q) (p/\mu) = \boxed{p/Q\mu}$$

as observed

rate of flow controlled break-up

- **quasi-stationary break-up**

- break-up governed by the evolution of pressure
- strong effects of confinement
- slow compared to relaxation rates
- (new) mechanism specific to microgeometries and low Ca



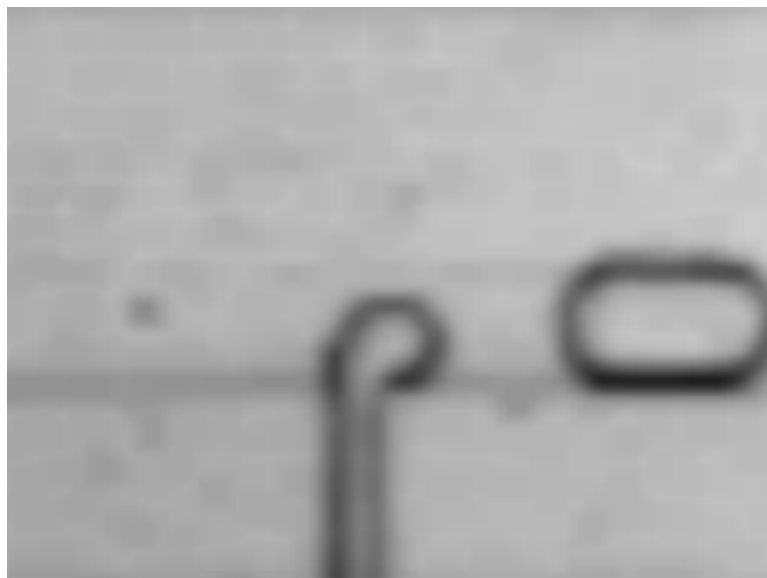
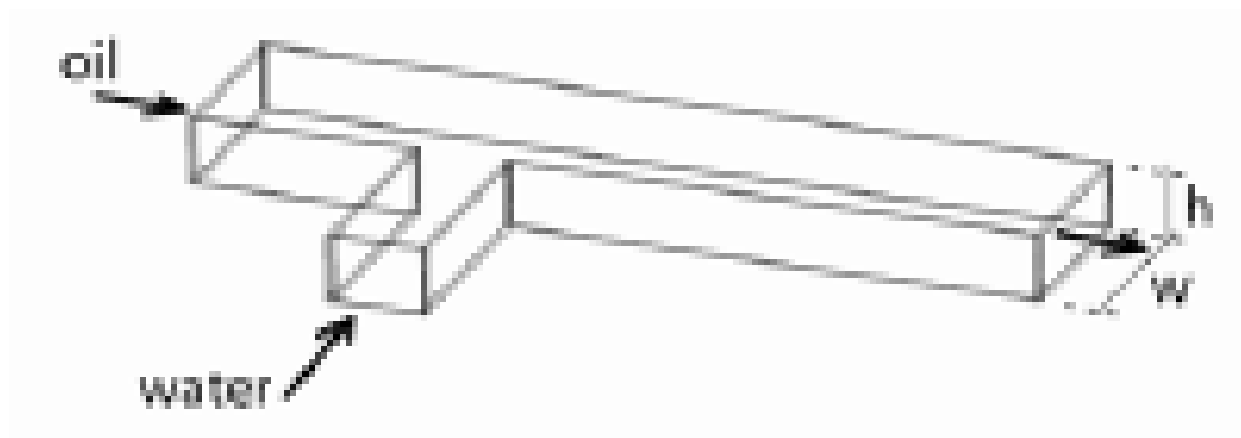
- **bubbles**

- both the size and the volume fraction can be controlled

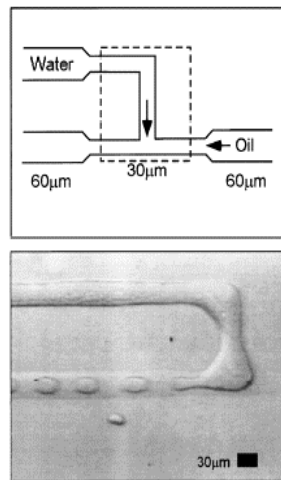
- **applications**

- formation of emulsions (*food, cosmetics, ultrasound contrast, artificial blood*)
- formation of monodisperse droplets
lab-chip, micro-particles, micro-capsules, lattices

T-junction



mechanism of break-up



Thorsen (2001)

shear vs interfacial forces

linear size $\sim Ca^{-1}$

not checked rigorously

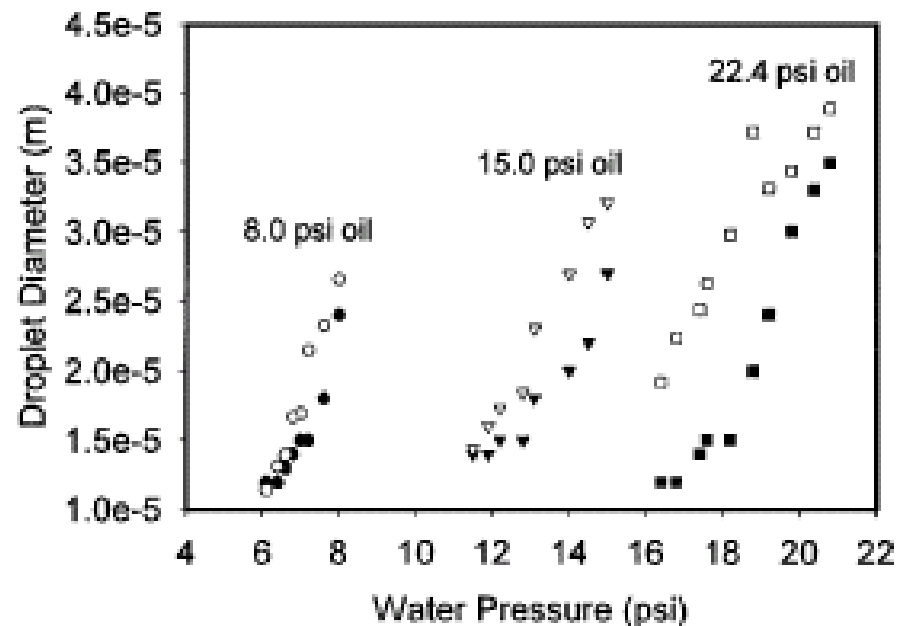
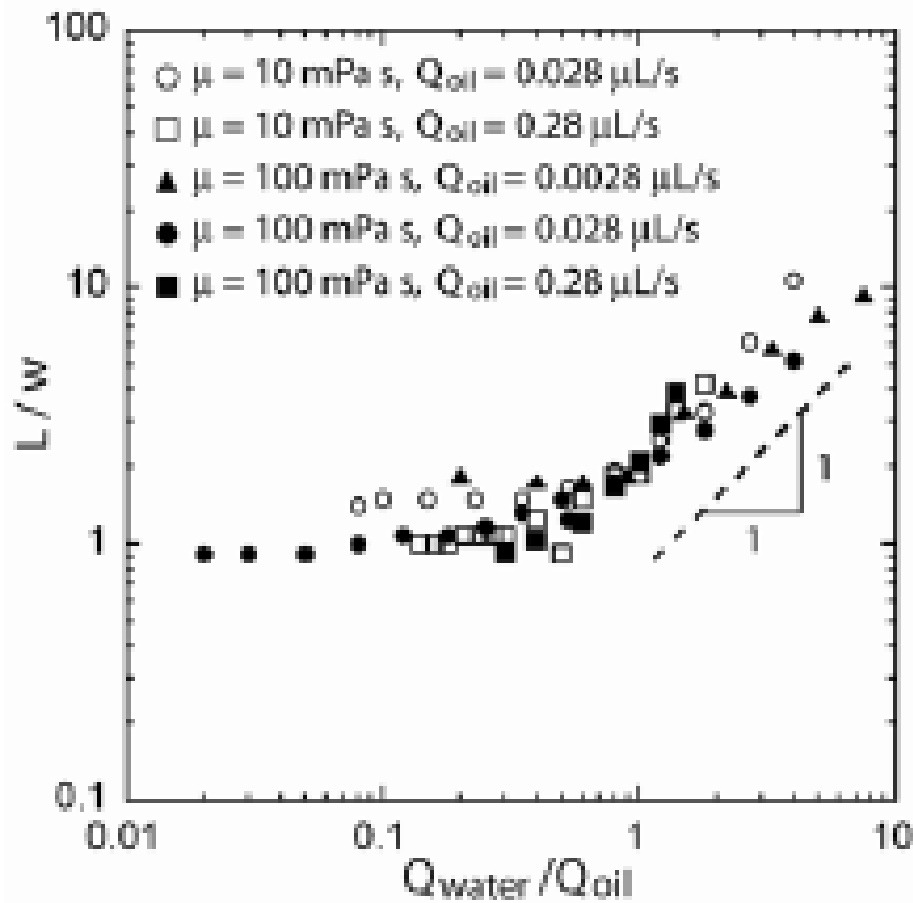


FIG. 4. Predicted vs actual drop size at different water and oil/surfactant pressures. The predicted sizes were calculated using Eq. (1). Open symbols, predicted size; solid symbols, experimental.

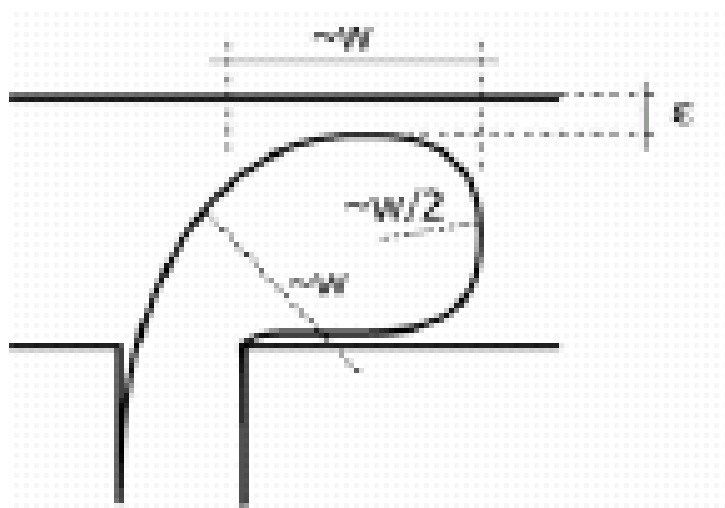
Thorsen (2001)

mechanism of break-up



100-fold change in shear stress
no change in size

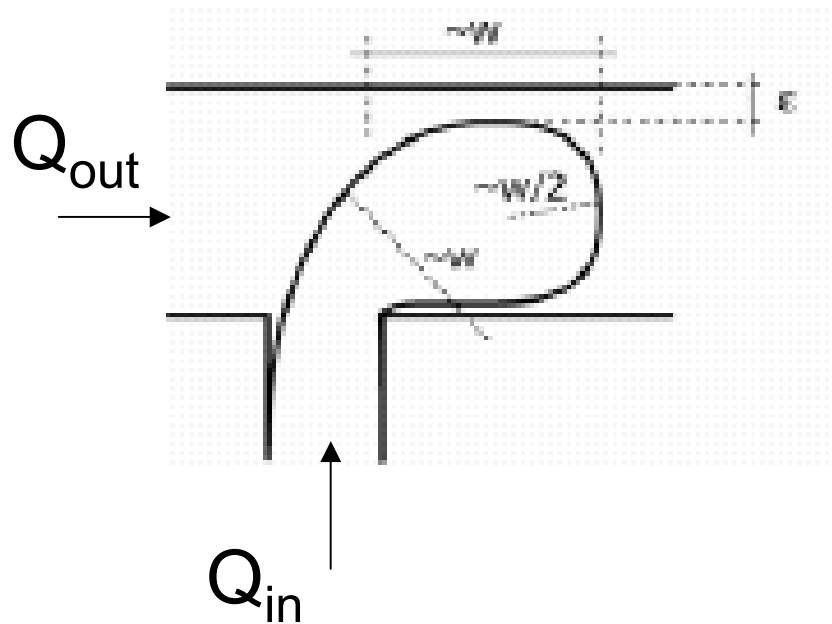
forces



- interfacial stresses (stabilizing)
- shear stresses (destabilizing)
overestimate scaling as ϵ^{-2}
- pressure drop (destabilizing)
scaling as ϵ^{-n} with $n > 2$

‘squeezing’ –

– rate of flow controlled break-up



- blocking the channel

$$t_{\text{block}} \propto 1/Q_{\text{in}}$$

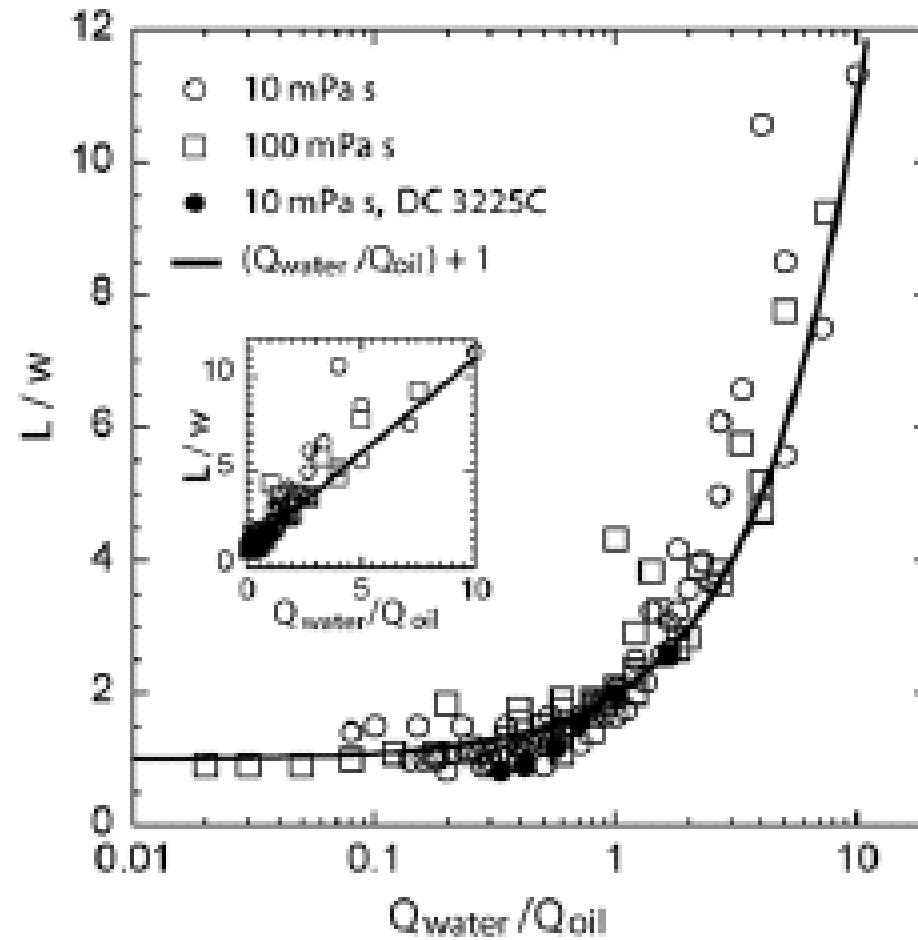
- squeezing

$$t_{\text{squeeze}} \propto 1/Q_{\text{out}}$$

$$\text{size} \sim (t_{\text{block}} + t_{\text{squeeze}}) Q_{\text{in}}$$

$$\text{size} \sim 1 + Q_{\text{in}}/Q_{\text{out}}$$

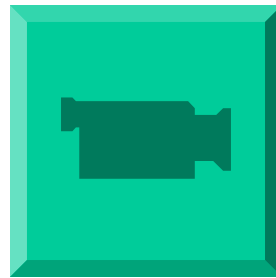
scaling



$$L/W \sim 1 + Q_{\text{in}}/Q_{\text{out}}$$

T-junction: simulations

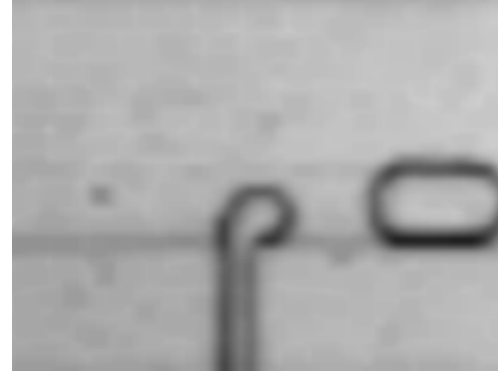
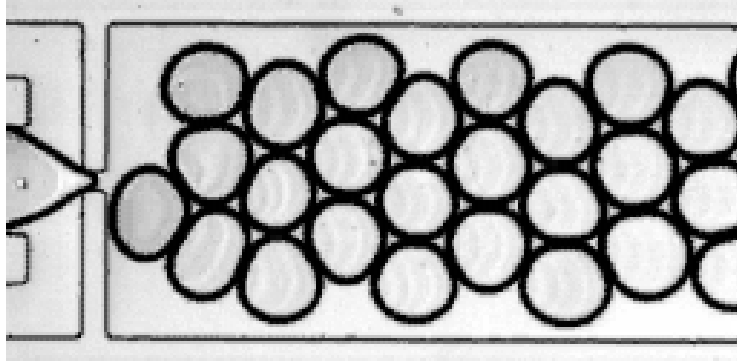
simulations confirm both the details of the dynamics of break-up and the scaling



runs over a wide range of values of Ca show three distinct regimes:

squeezing – dripping – jetting

with **squeezing** being a new break-up mode, specific to microgeometries

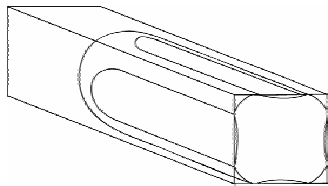
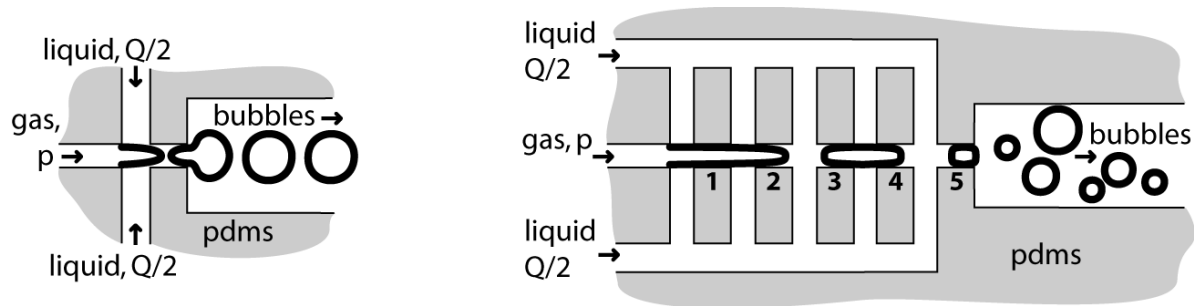


- **quasi-stationary break-up**

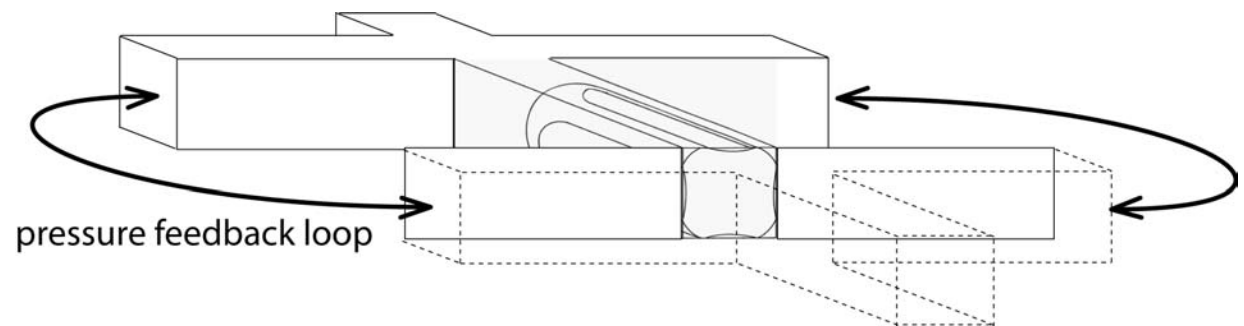
- break-up governed by
 - the evolution of pressure
- strong effects of confinement
- slow compared to relaxation rates
- (new) mechanism
 - specific to microgeometries and low Ca

- **microfluidics**
 - simple fluids
 - droplets and bubbles
- **formation of drops and bubbles**
 - flow-focusing
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coupled flow-focusing oscillators

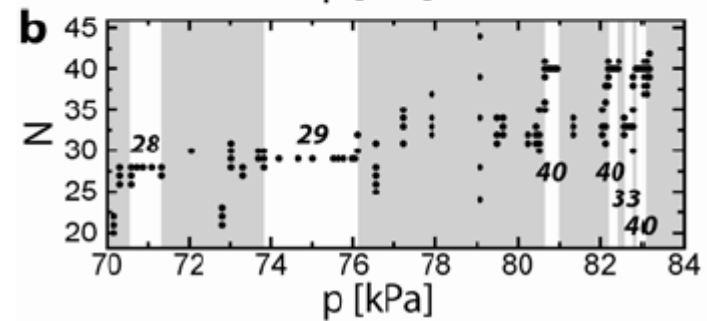
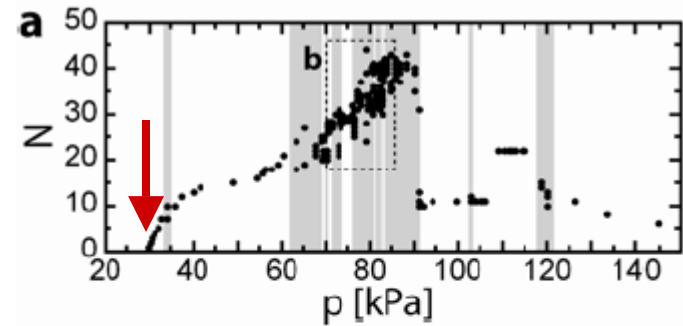
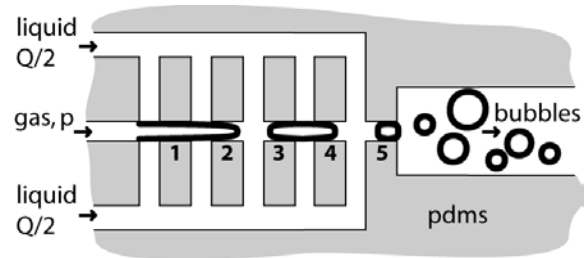


a plug
'blocks' the channel



- 1. the rate of collapse of the thread at each orifice depends on what happens everywhere else***
- 2. the exchange of information (via pressure waves) is much faster than the evolution of the thread***

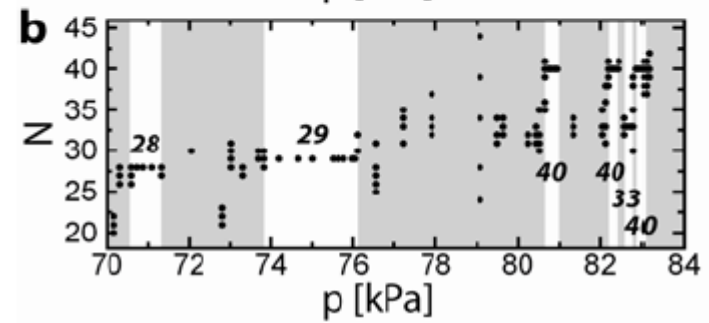
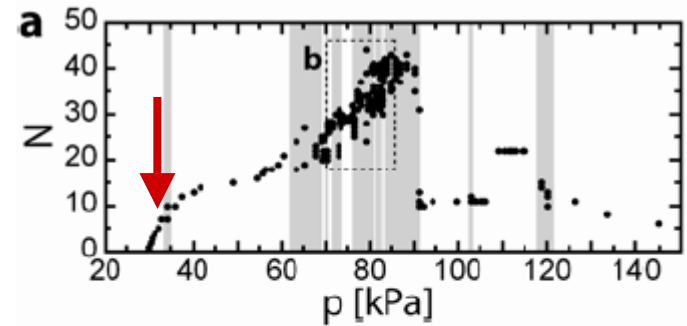
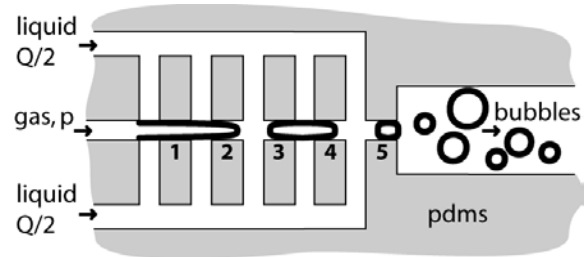
multi-orifice: operation



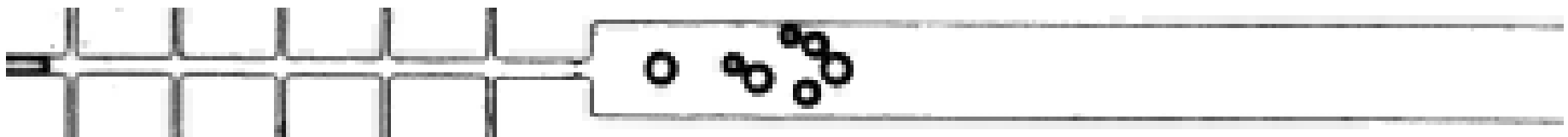
period-1



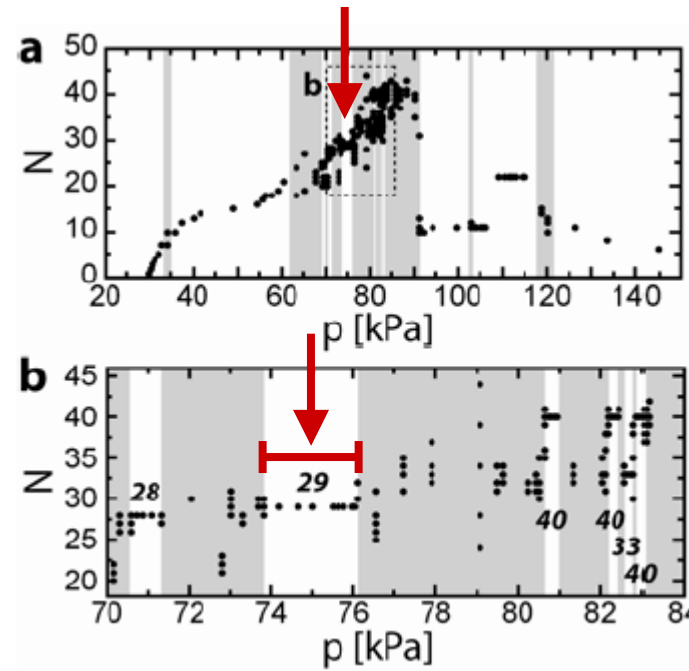
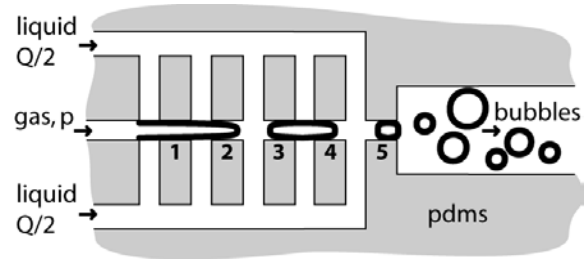
multi-orifice: operation



period-7



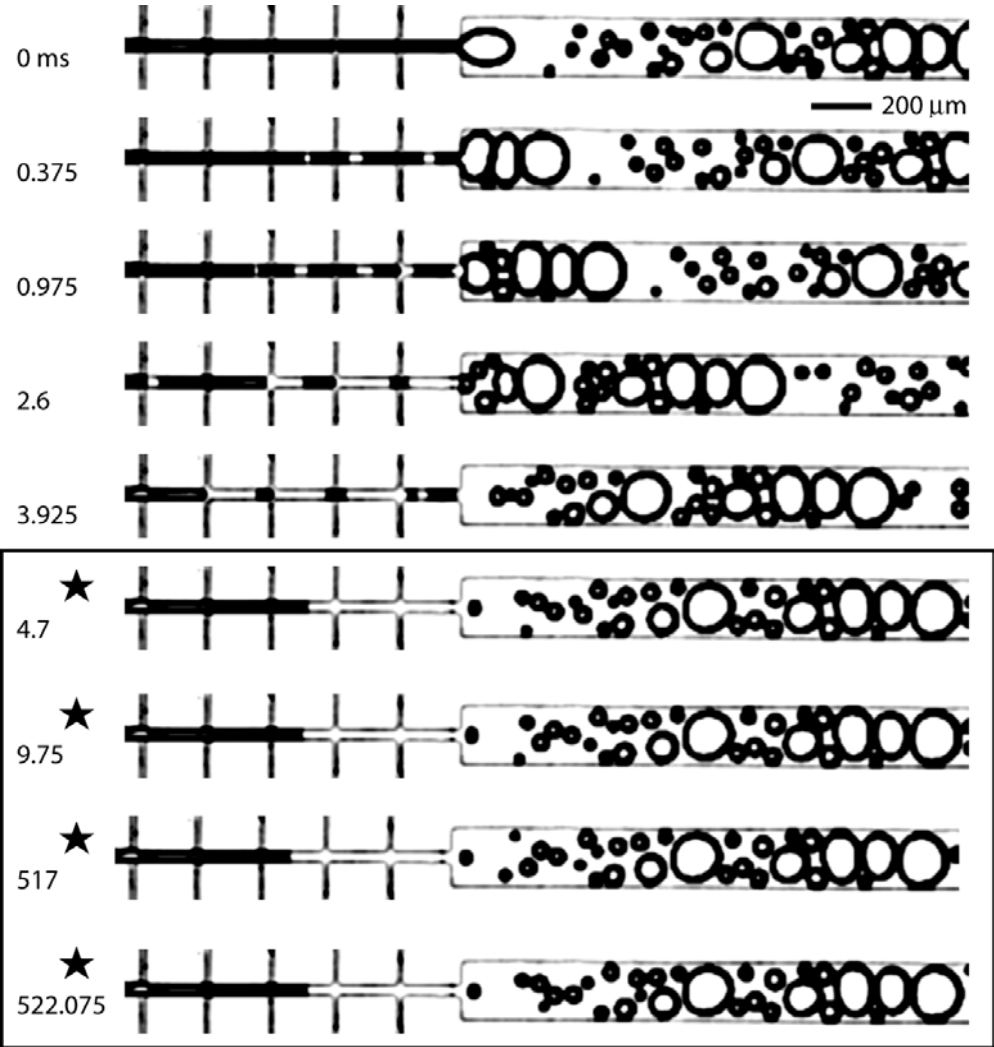
multi-orifice: operation



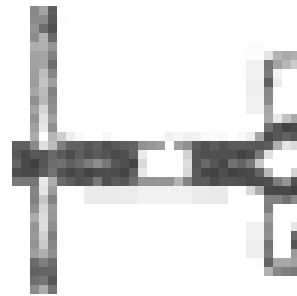
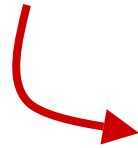
period-29



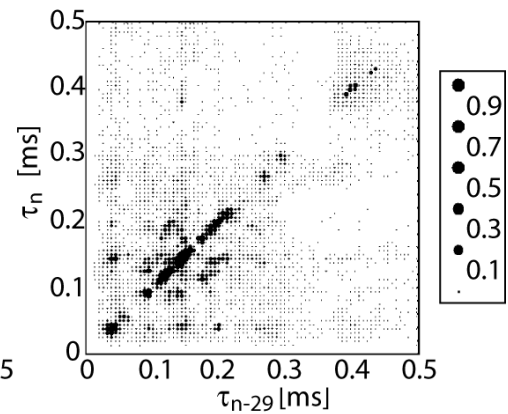
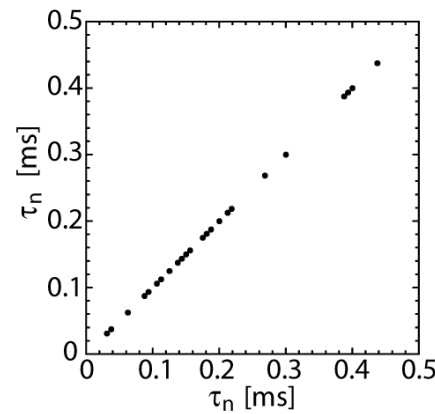
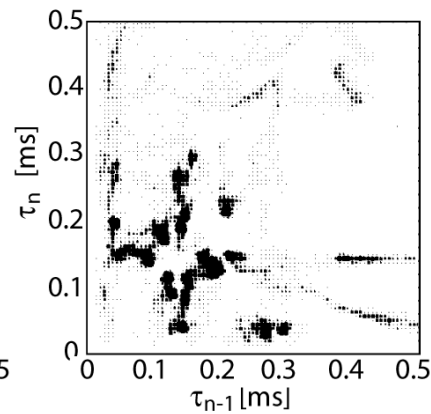
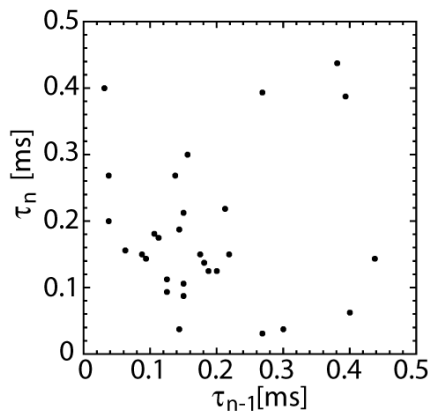
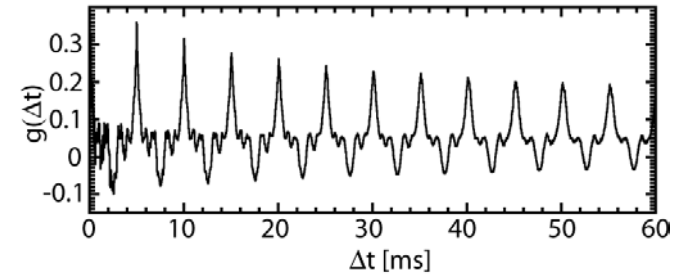
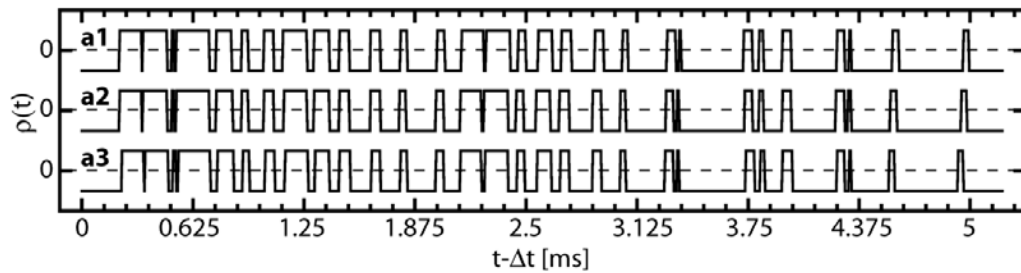
multi-orifice: **period 29**



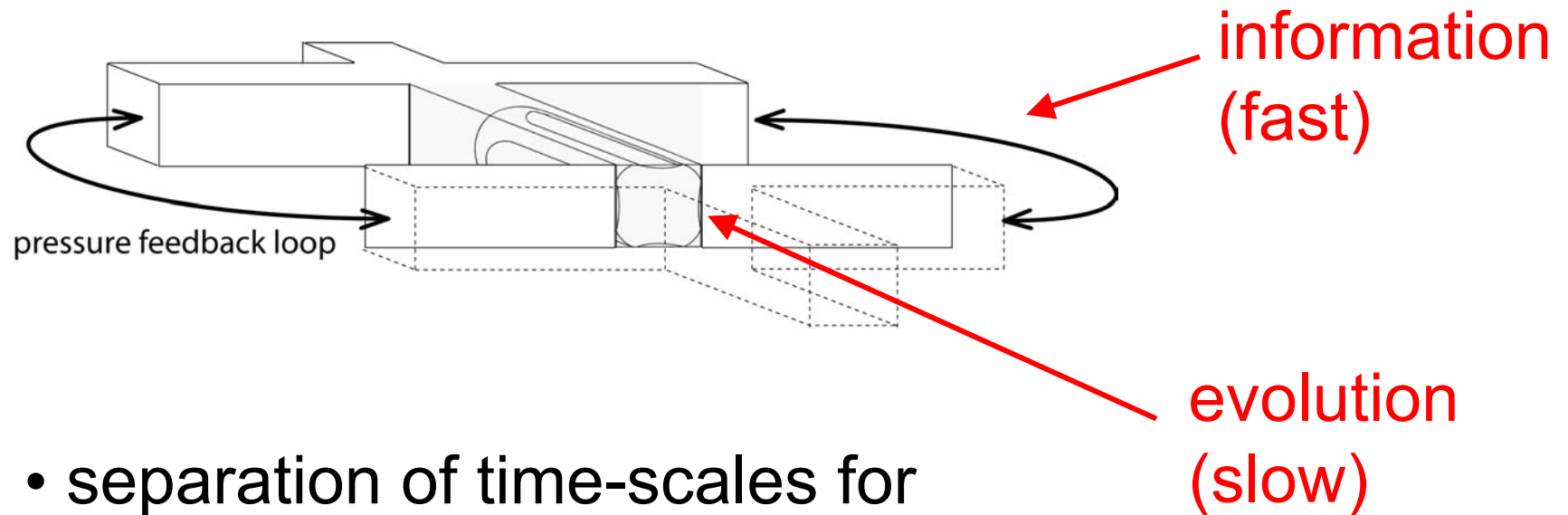
multi-orifice: period 29 – analysis



160 kfps –
– 6.25 μs

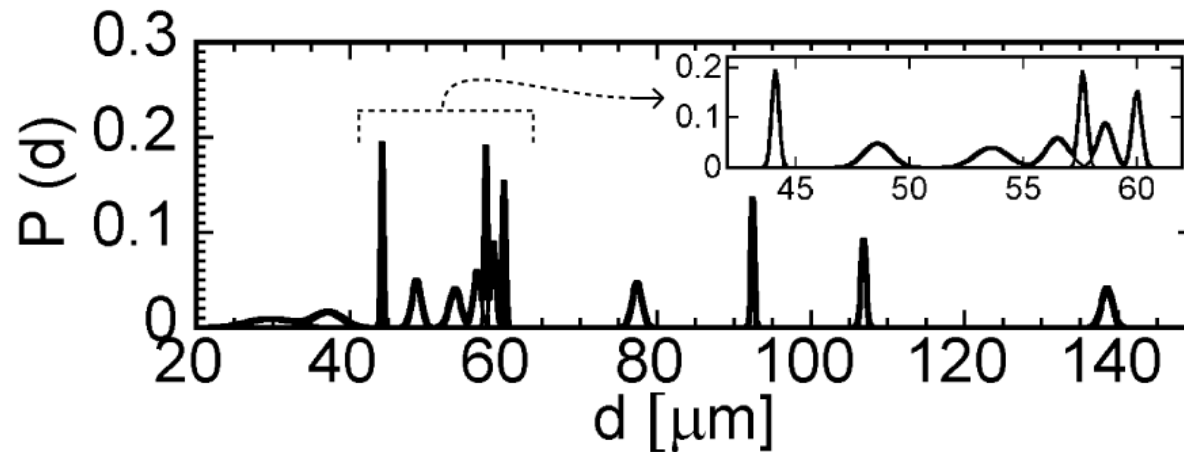
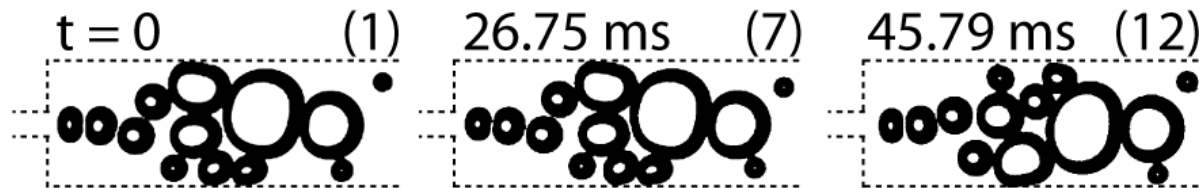
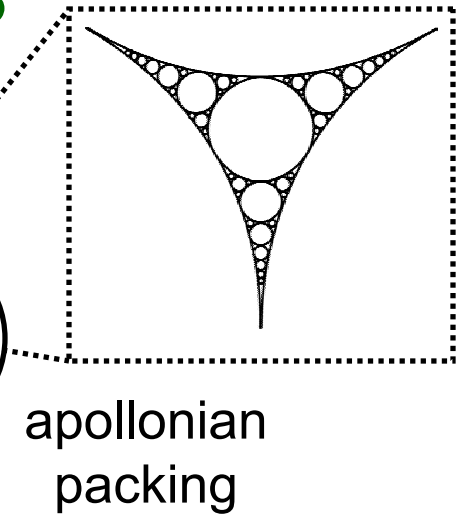
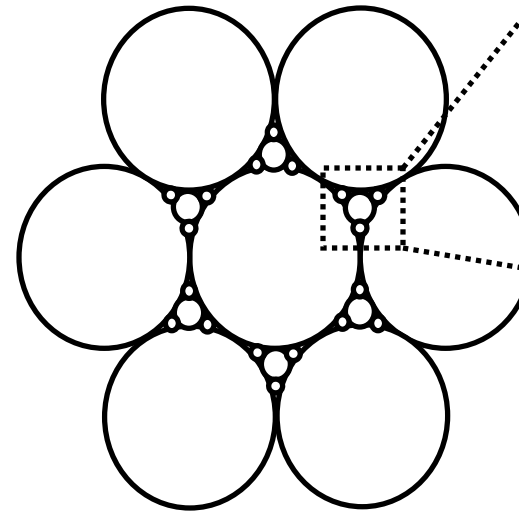
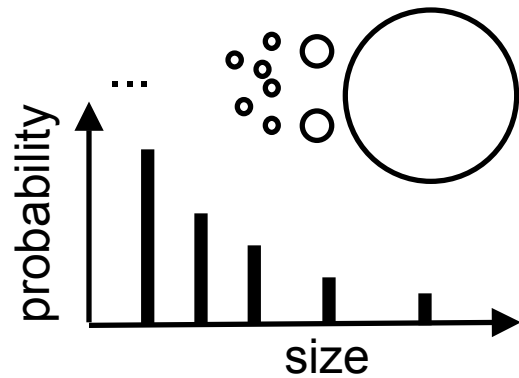


multi-orifice



- separation of time-scales for
 - (slow) **break-up**, and
 - (fast) **exchange of information**
- + dissipative dynamics (low to mod Re)
 - effective isolation of the system
- applications – Apollonian packings?

engineering size distributions

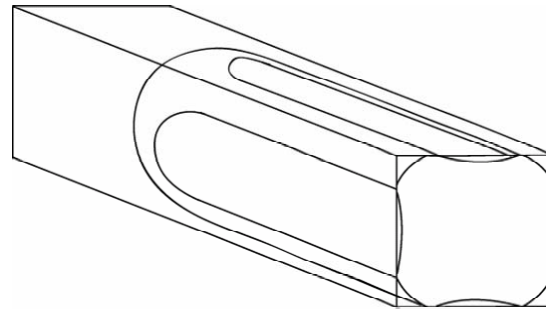


- **microfluidics**
 - simple fluids
 - droplets and bubbles
- **formation of drops and bubbles**
 - flow-focusing
 - T-junction
- **stable oscillations with long periods**
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plug flow

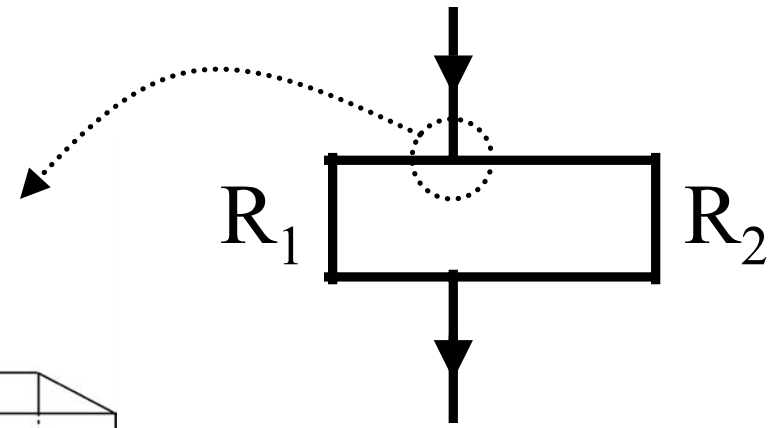
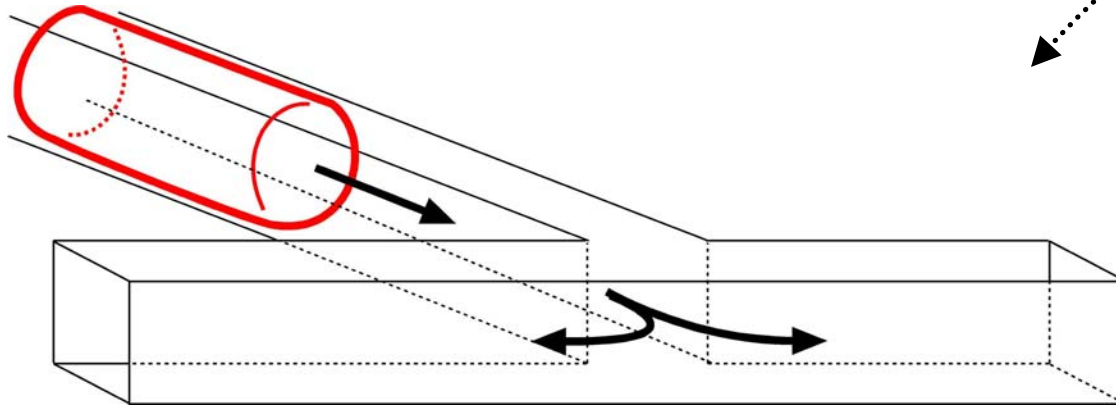


increased
resistance
to flow,
 $\Delta p \propto u^{2/3}$



Taylor
Bretherton
Wong

plug flow – decisions

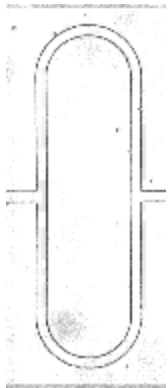


- simple fluid will split as $q_1/q_2=R_2/R_1$
- a bubble has to make a decision
- once it enters a channel it changes (increases) its resistance

a dynamic system with feedback (or memory)

One Loop

Period-1



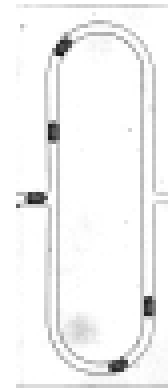
Period-2



Period-3

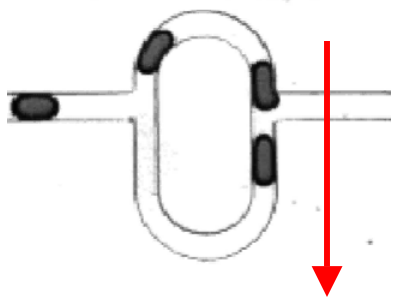


irregular

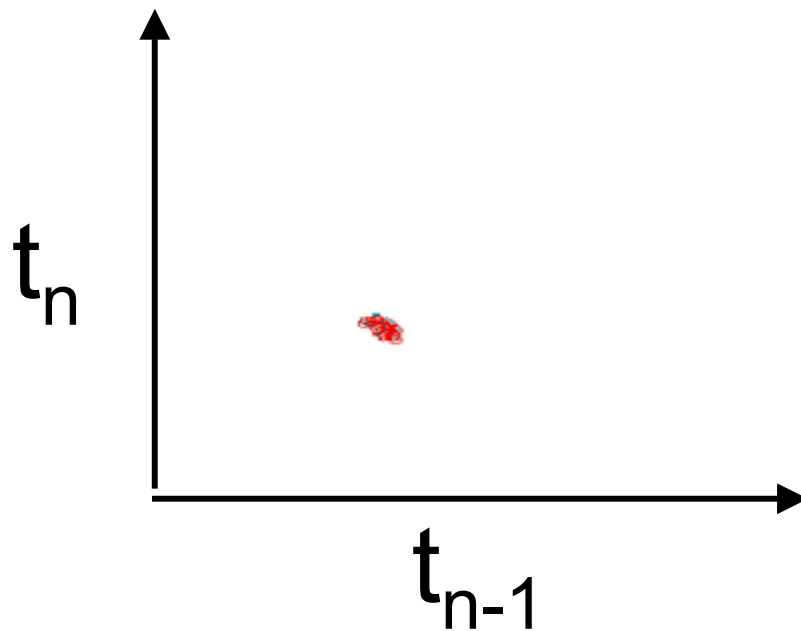
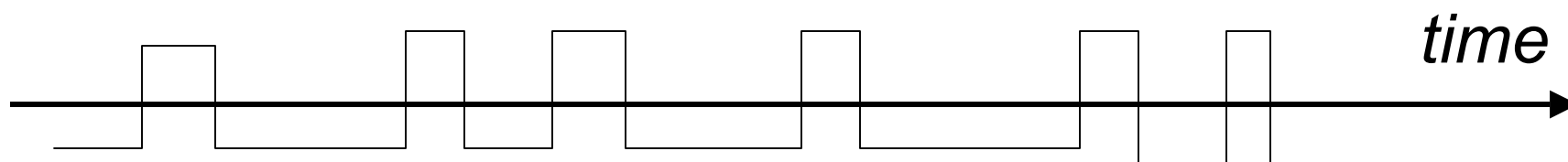


frequency of feeding

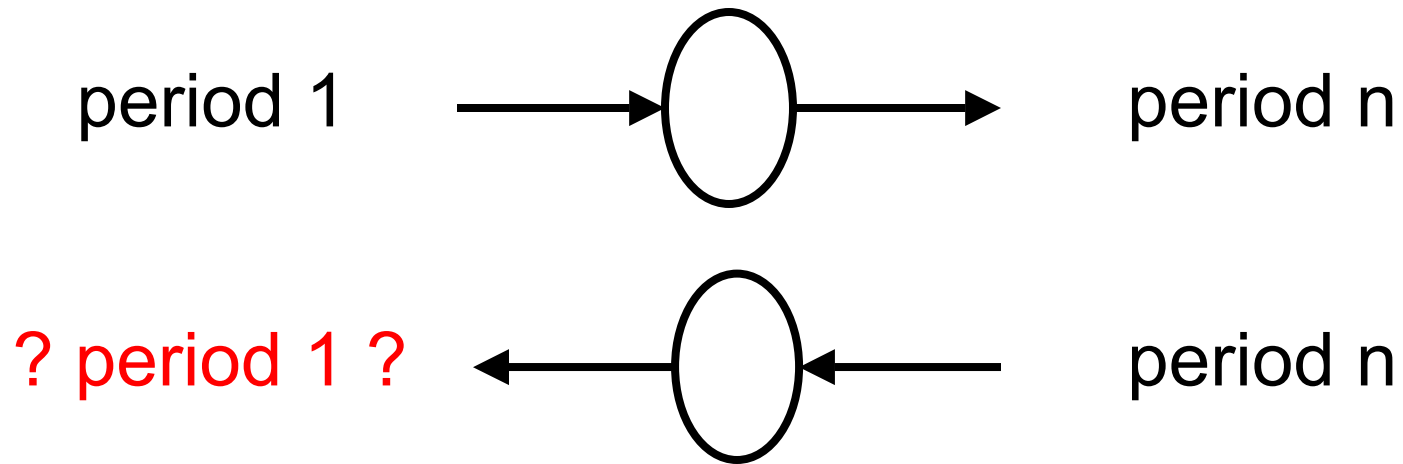
One Loop



t_n



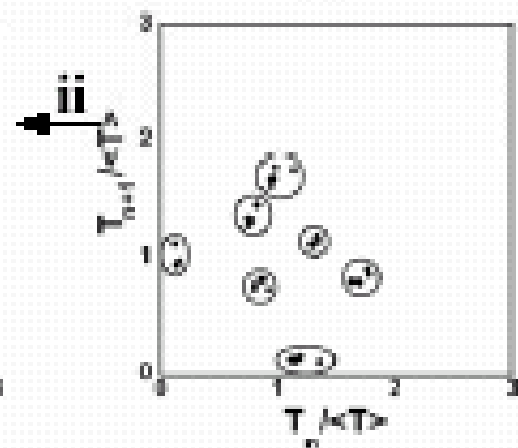
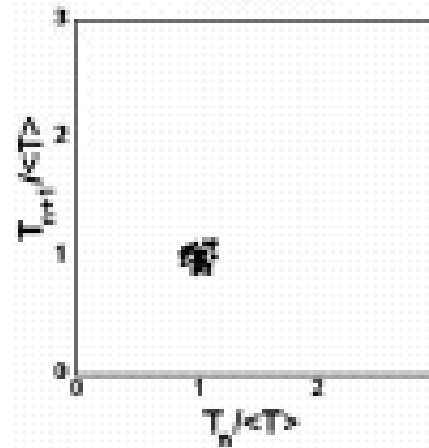
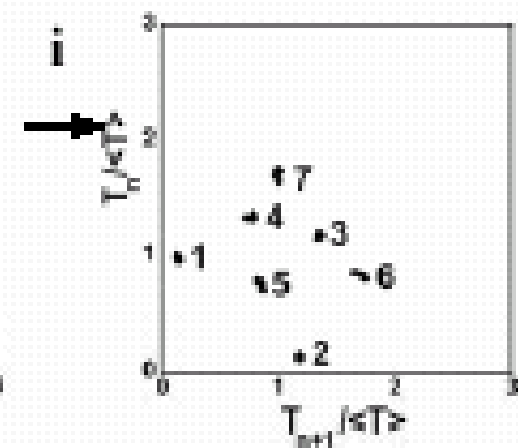
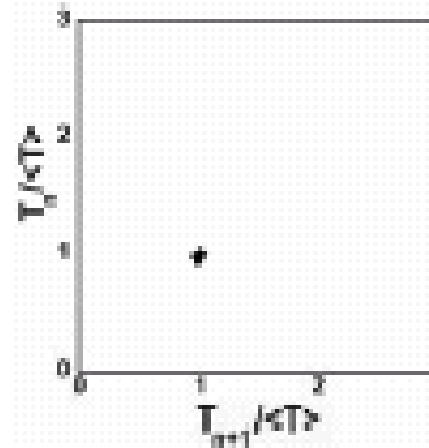
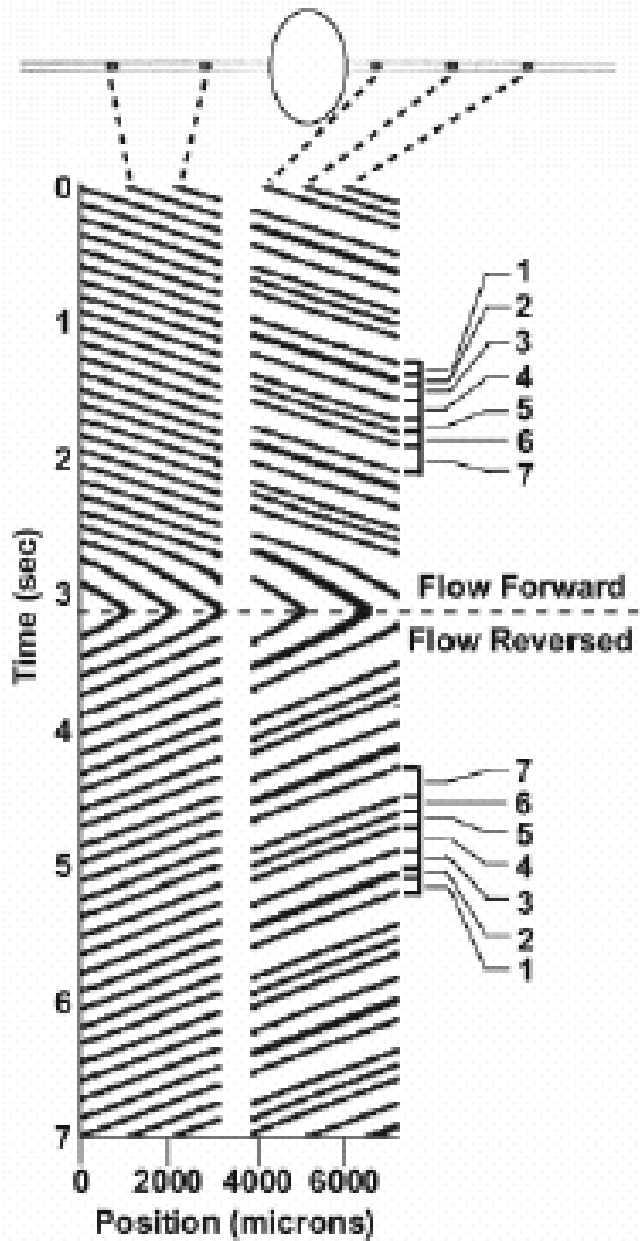
reversibility



$$\eta \Delta \vec{V} - \vec{\nabla} P = 0$$

invariant under: $V \rightarrow -V, P \rightarrow -P$

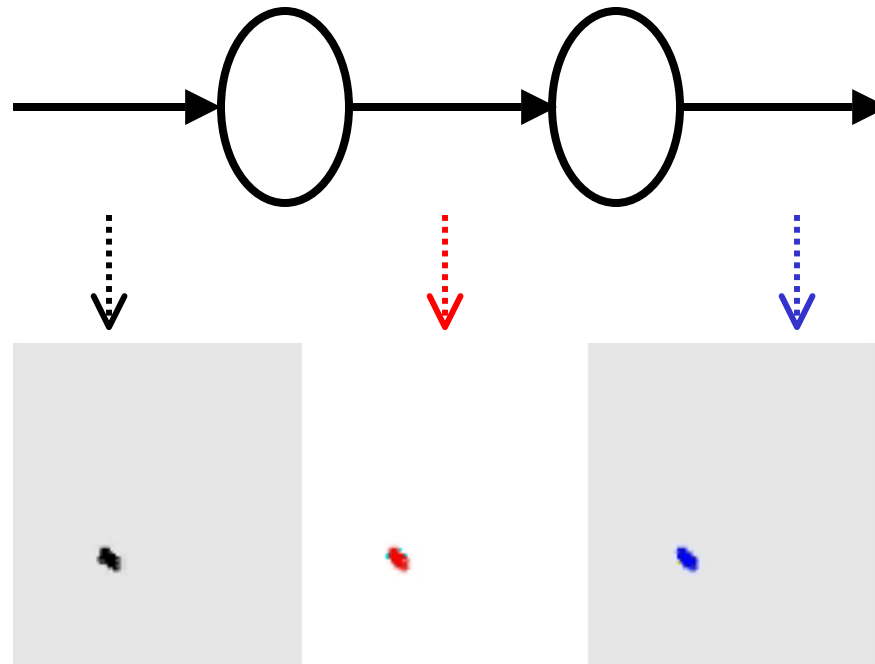
period 1 \longleftrightarrow period 7



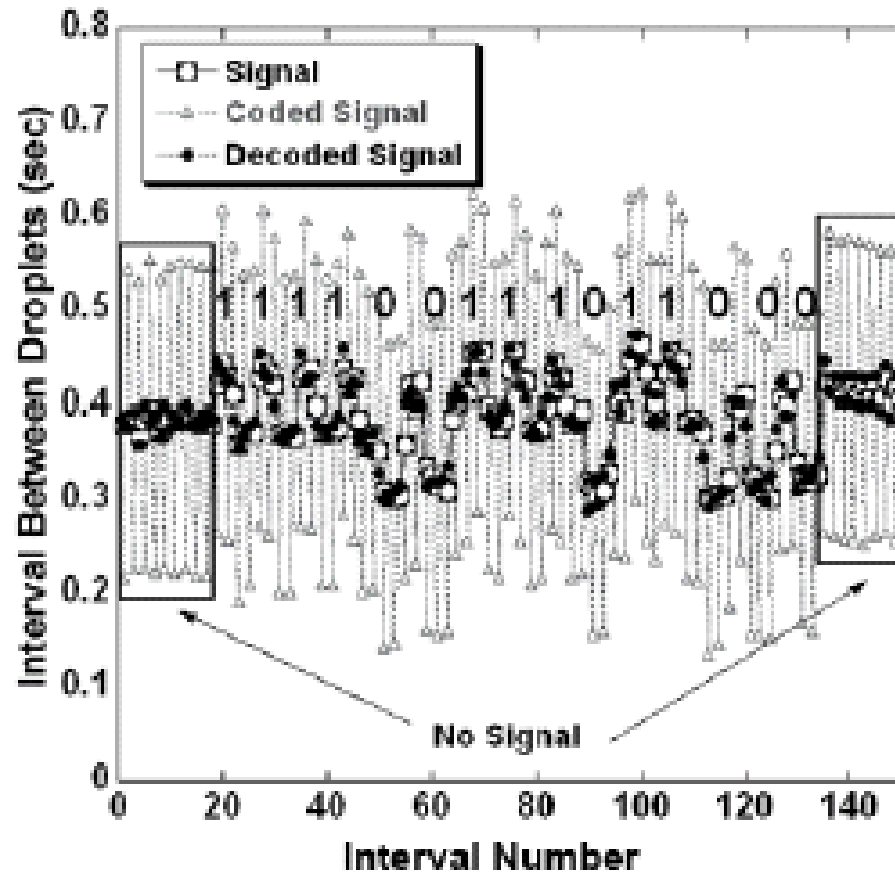
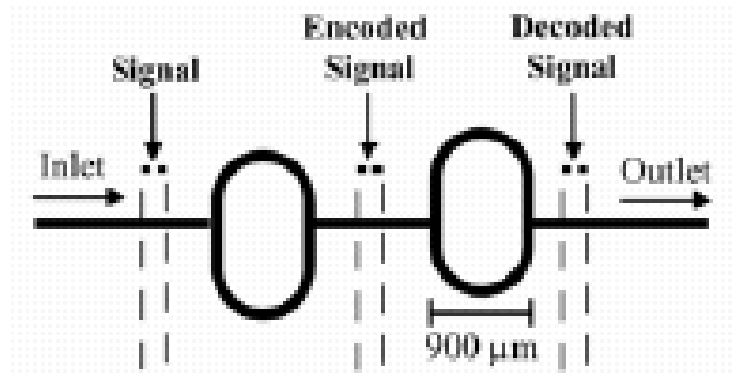
symmetric singals

period 1 \rightarrow period 2 \rightarrow period 1

AAAAA \rightarrow BCBCBC \rightarrow AAAAAA



coding – decoding



reversible flows

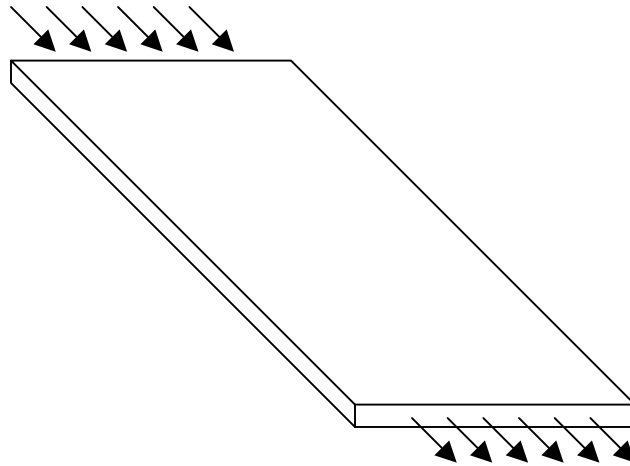
isolated events of amplification
embedded in linear, reversible dynamics of flow

→ probing the fine line between non-linearity
and reversibility

→ potential for automated and robust
processing of signals on chip (potentially
useful in lab-on-chip applications)

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mixing



2D, steady, incompressible flow: $u = \frac{\partial \psi}{\partial y}$, $v = -\frac{\partial \psi}{\partial x}$.

integrable,
no chaotic advection $\frac{dx}{dt} = \frac{\partial \psi}{\partial y}$, $\frac{dy}{dt} = -\frac{\partial \psi}{\partial x}$.

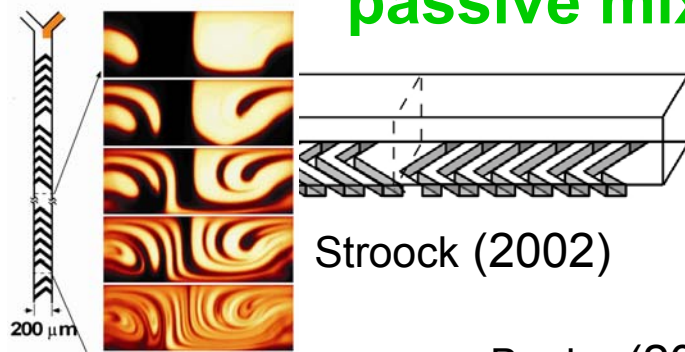
it is difficult to mix fluids in microchannels

mixing – solutions

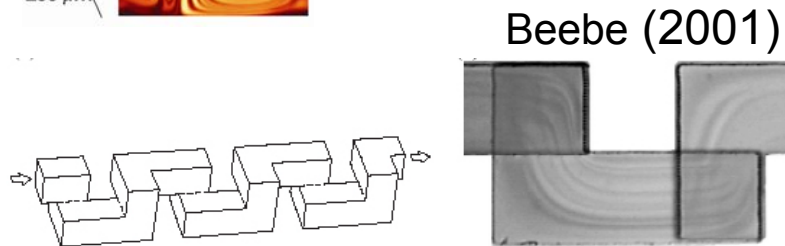
~~two-dimensional, steady flows~~

passive mixers

active mixers

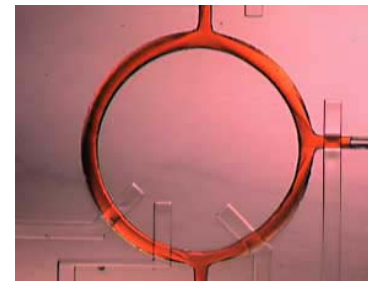


Stroock (2002)

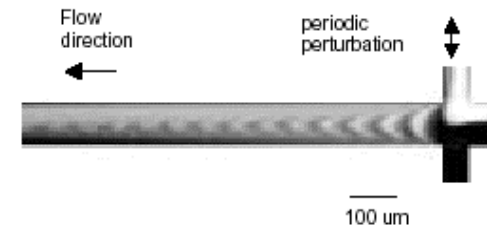


Beebe (2001)

difficult fabrication
always 'on'



S. Quake

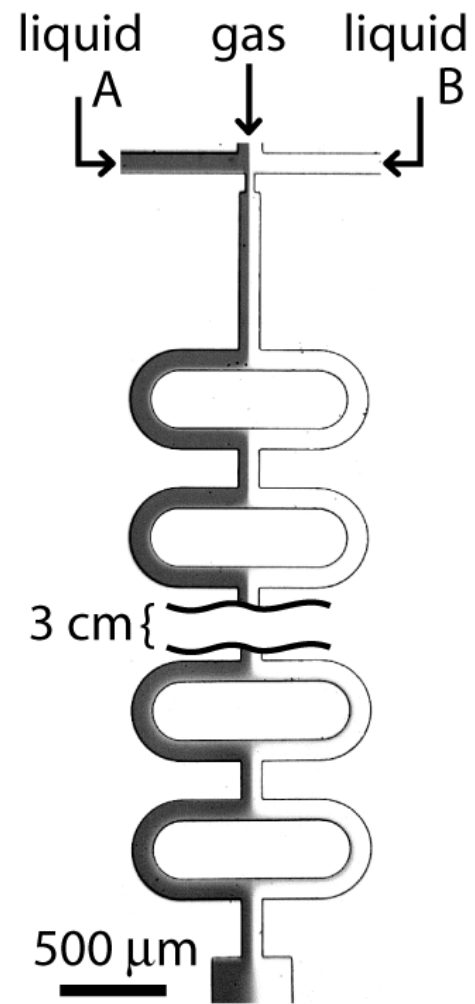


Okkels (2004)

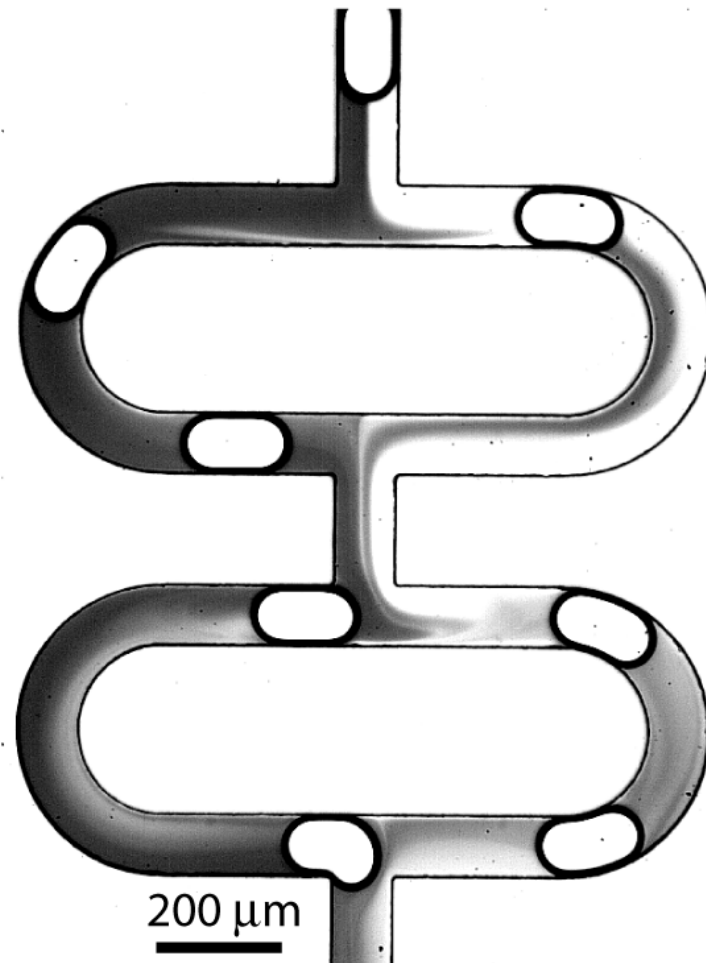
moving parts
external control / agitation

can we achieve exponential folding
in a **planar** device and **steady-state** input?

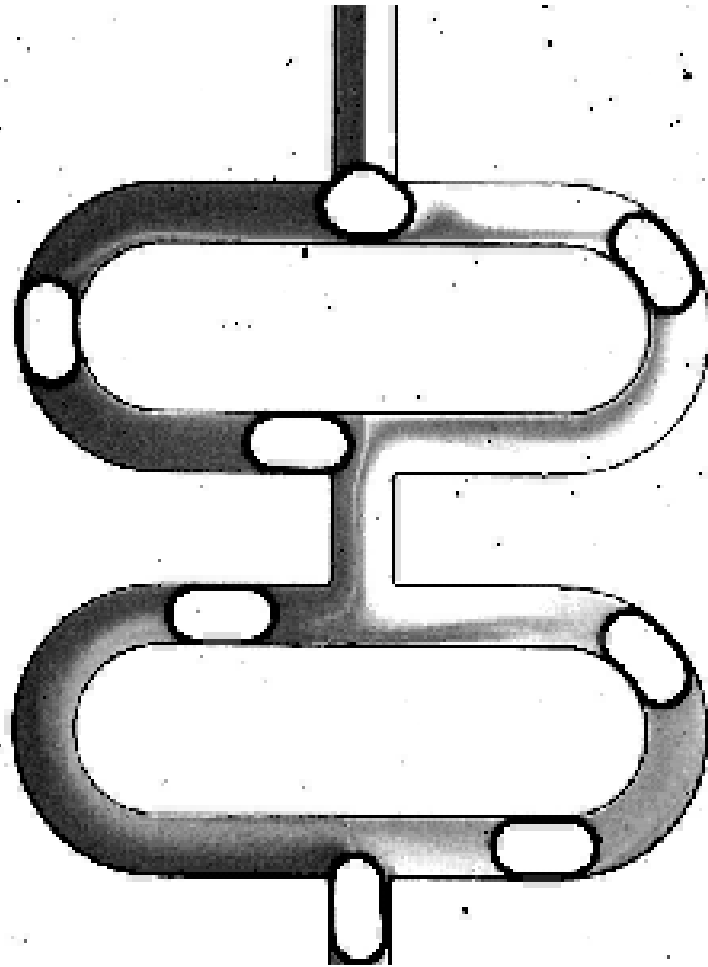
mixer



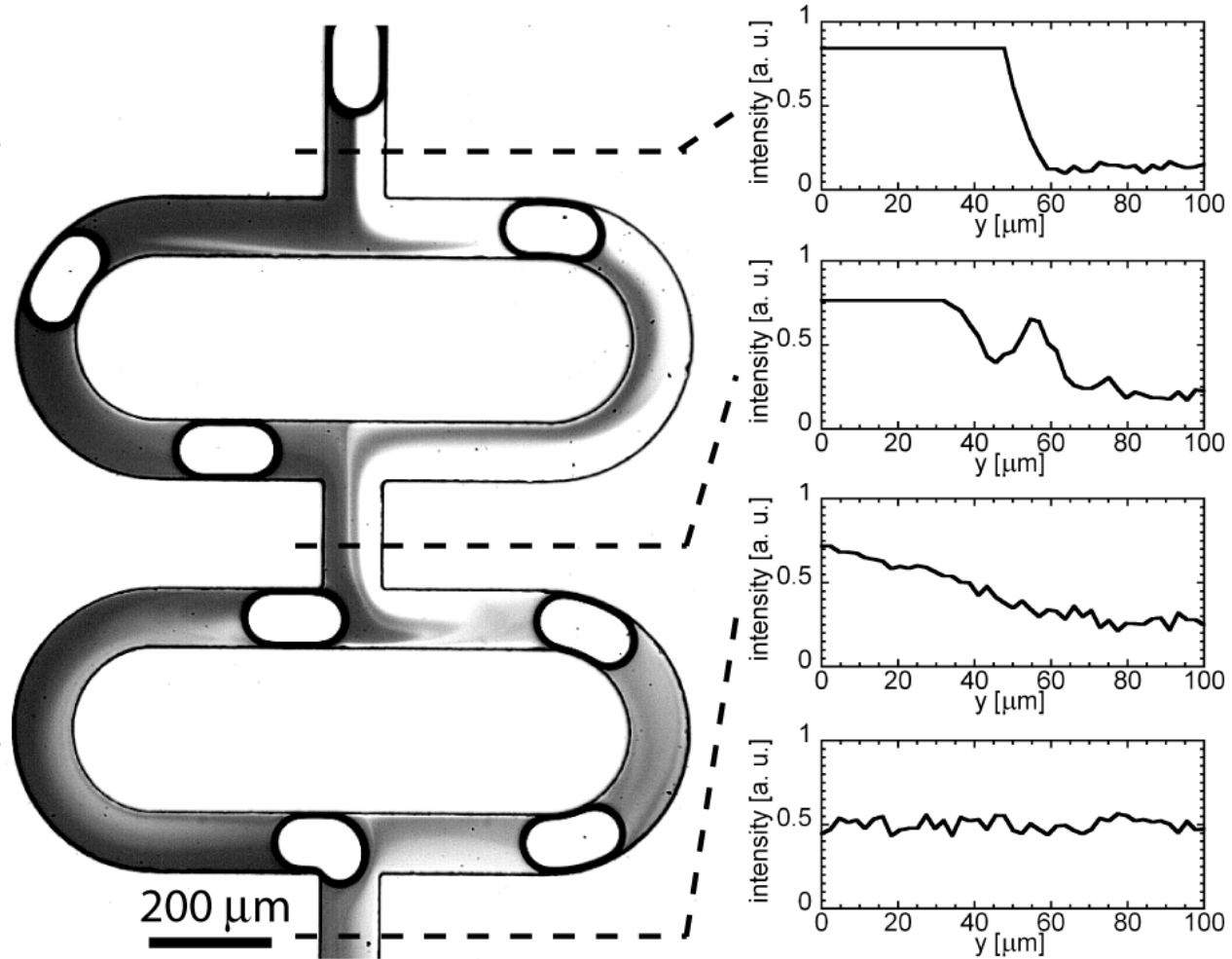
alternating flow



alternating flow



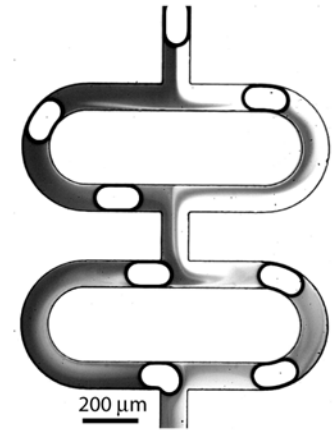
crossing streamlines



folding of the interface

$$d_{\text{inter}} \sim w (2^{-L/a})$$

$$d_{\text{diff}} = (tD)^{1/2}$$



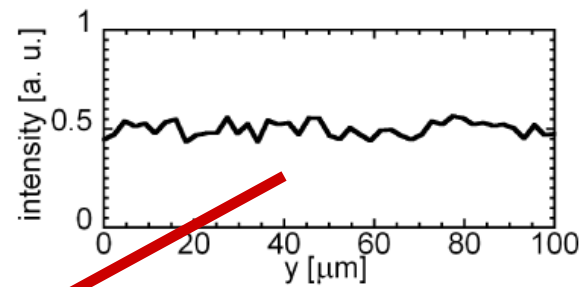
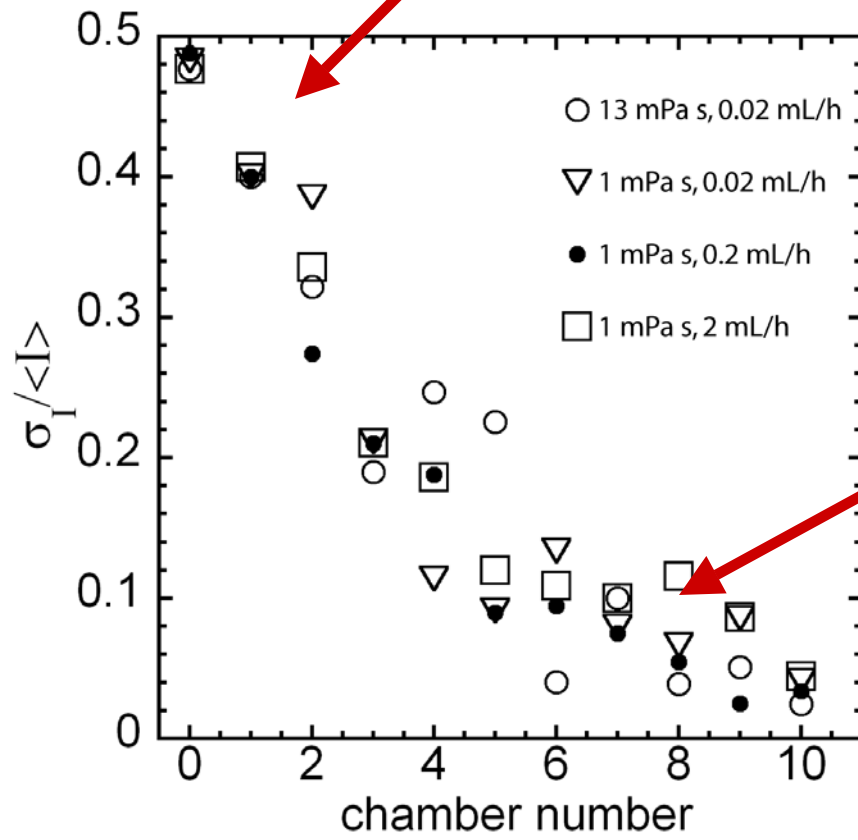
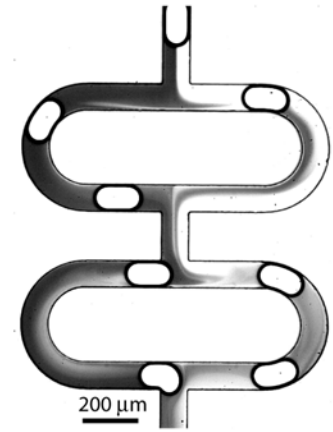
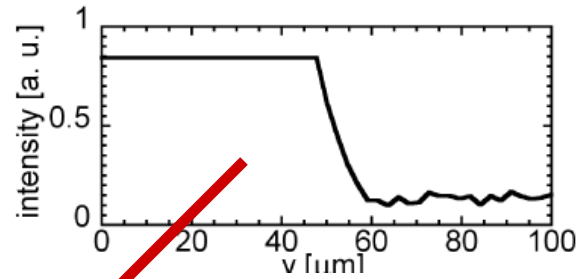
$$(L/a) = (2 \ln 2)^{-1} (\ln Pe - \ln (L/w))$$

$$Pe = Q/Dw = 10^5 \text{ for } Q = 1 \text{ } \mu\text{L/s, } D = 10^{-6} \text{ cm}^2/\text{s and } w = 100 \text{ } \mu\text{m}$$

number of chambers to mix
the two liquid streams:

$$(L/a) \sim 8$$

mixing

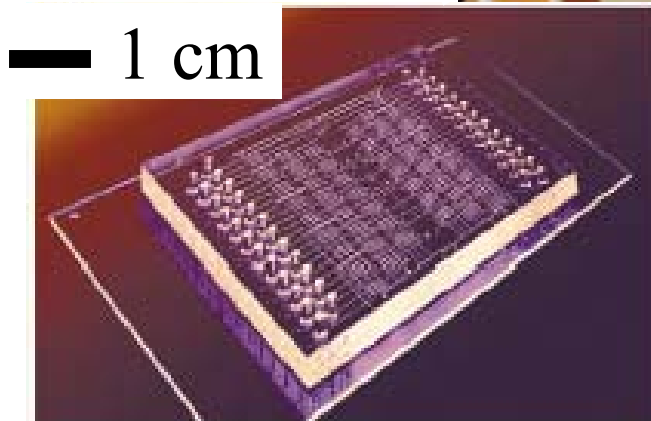


$\text{Re} \in (10^{-2}, 10)$

initial $\text{Pe} \in (10^3, 10^5)$

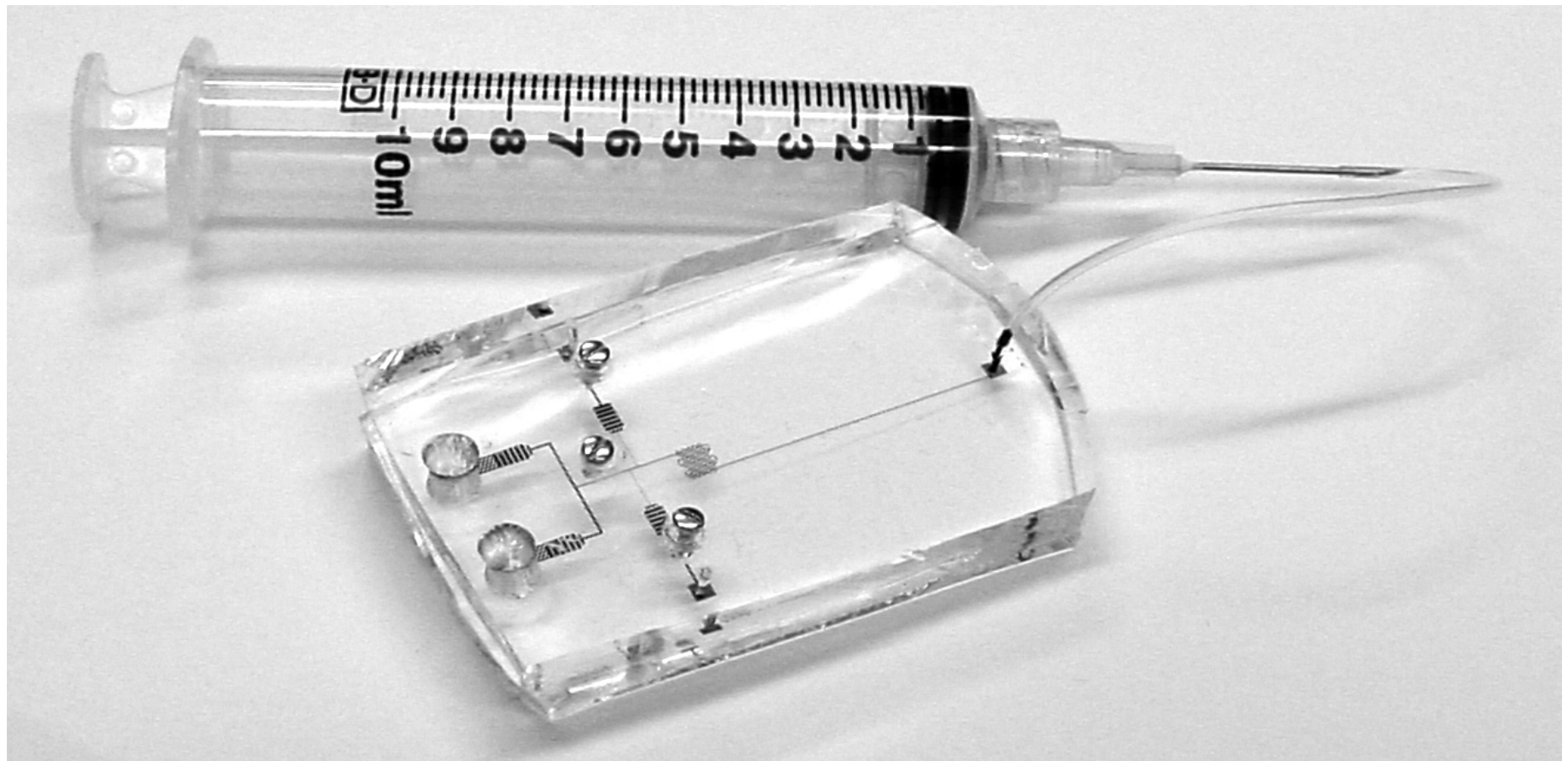
reduced to $\text{Pe} < 100$

portability



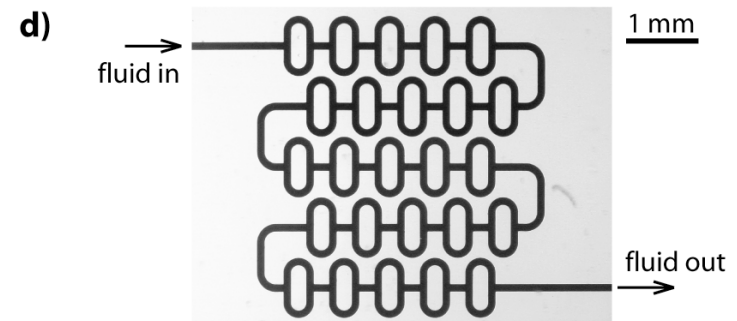
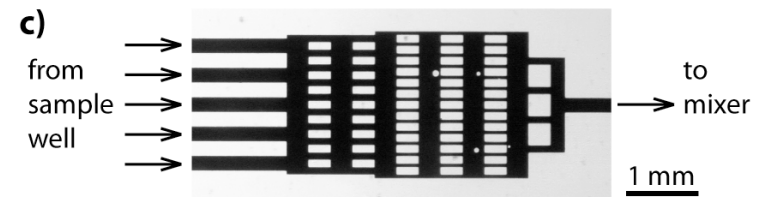
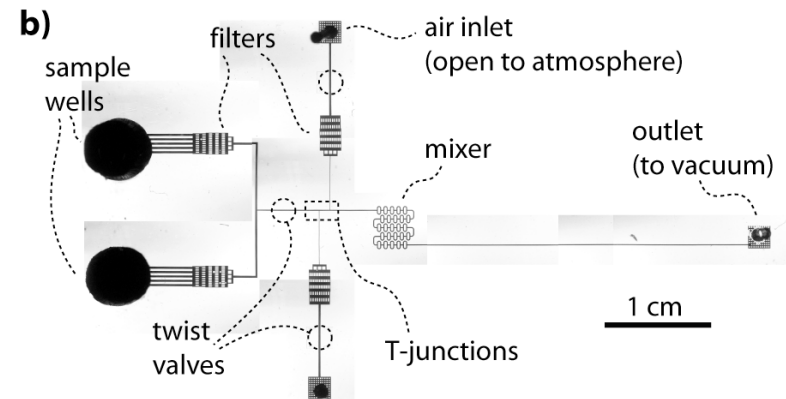
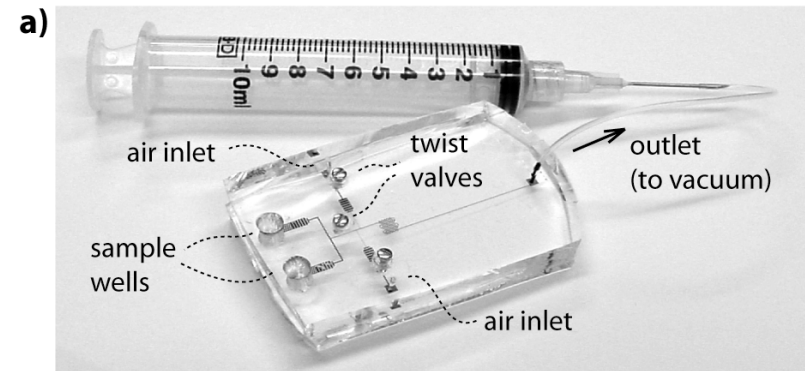
vaxer

A portable platform
solution based micro assays

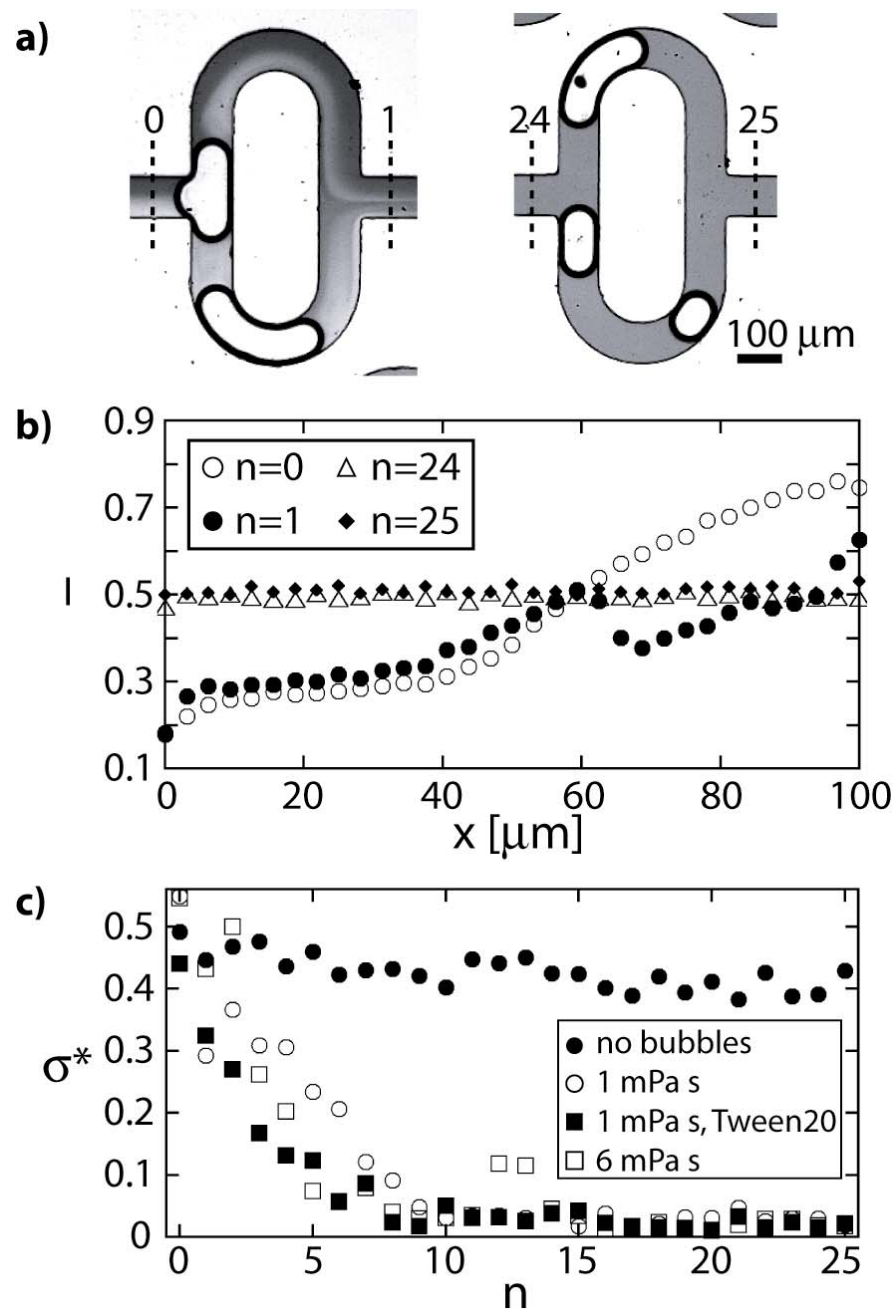
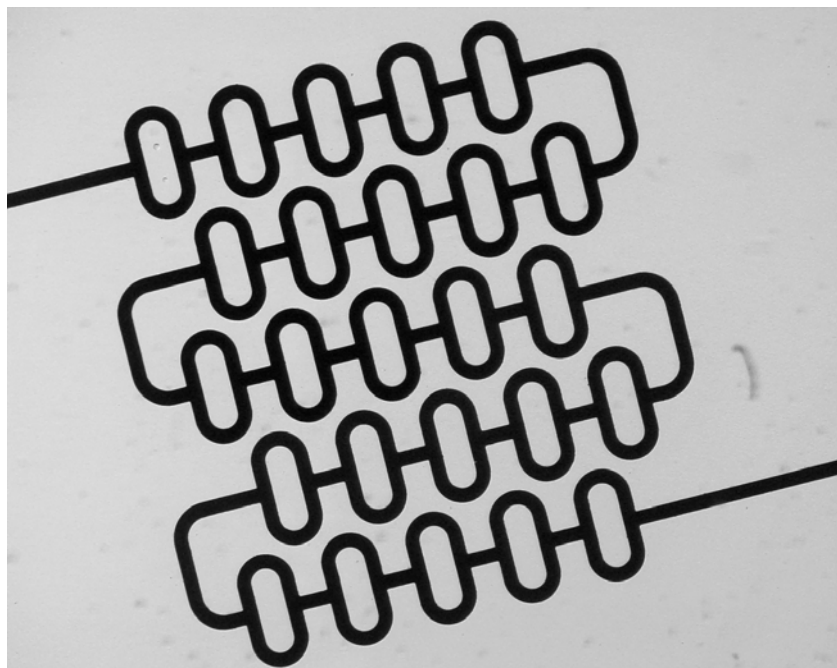


vaxer

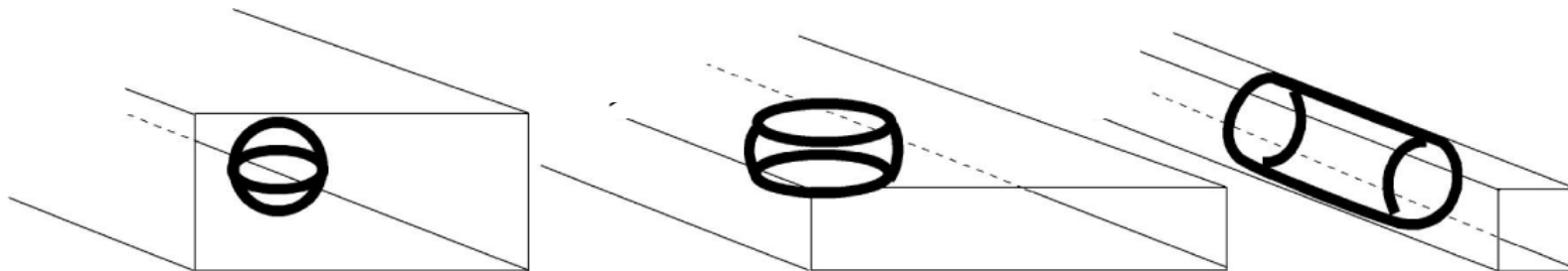
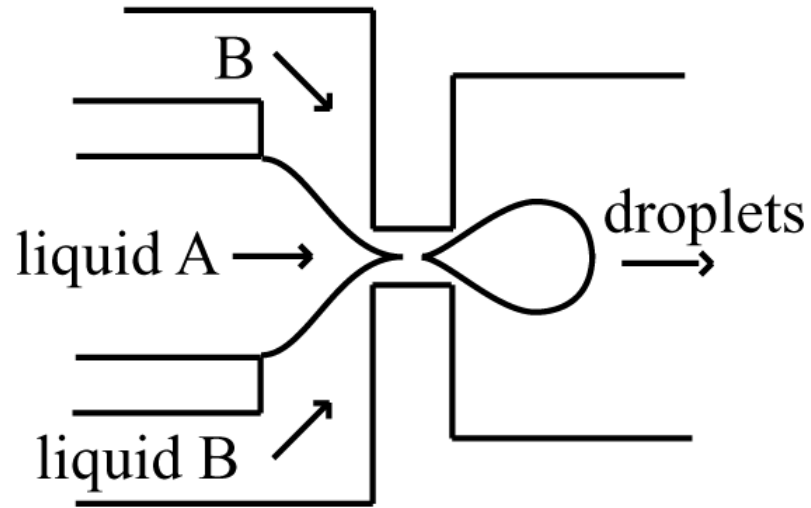
A portable platform
solution based micro assays



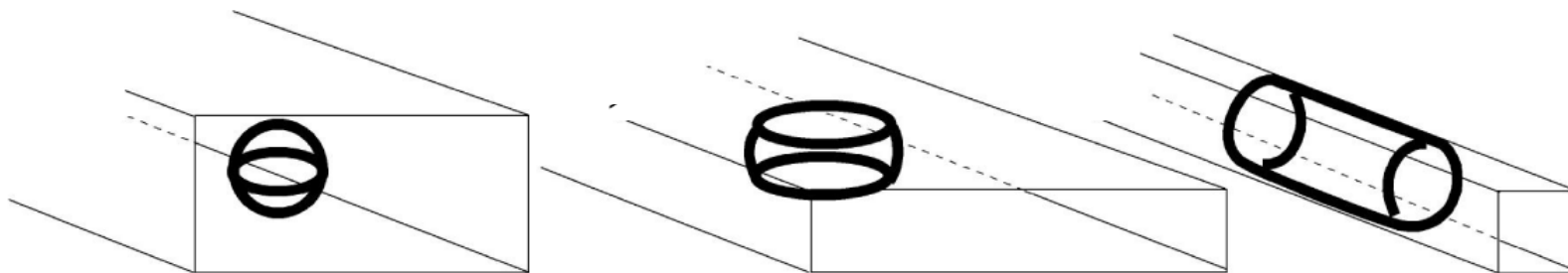
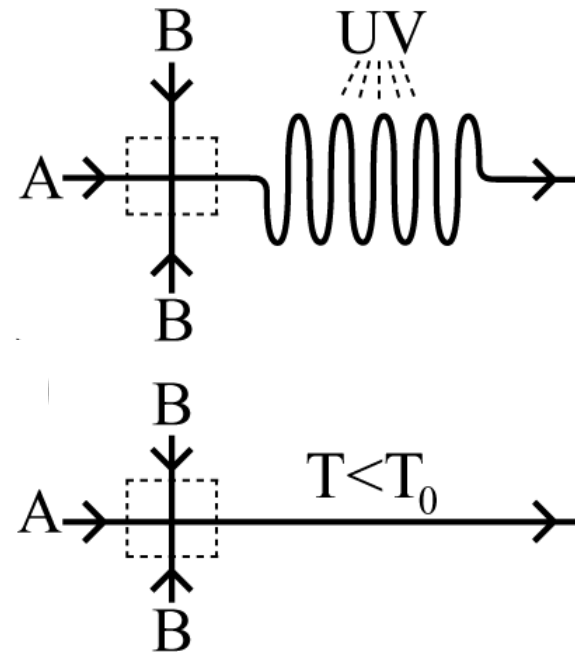
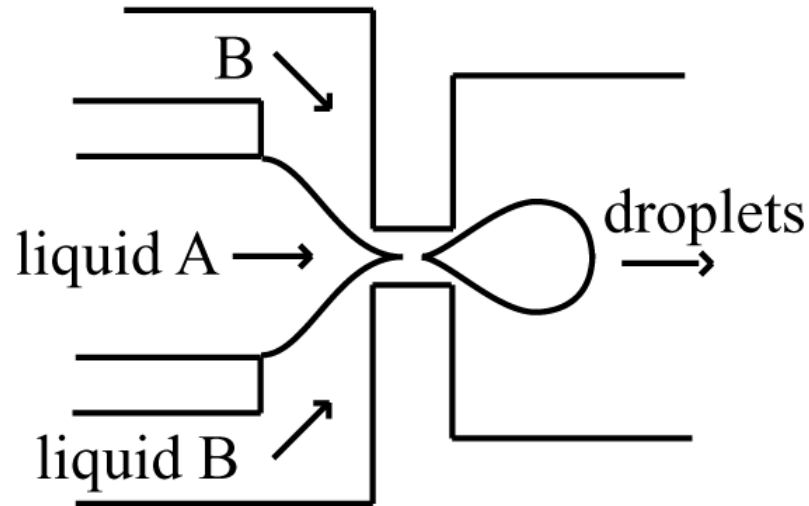
vaxer



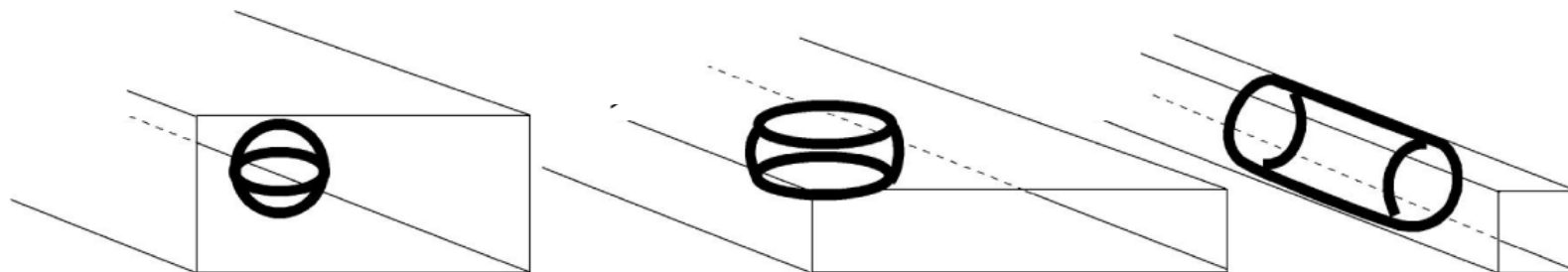
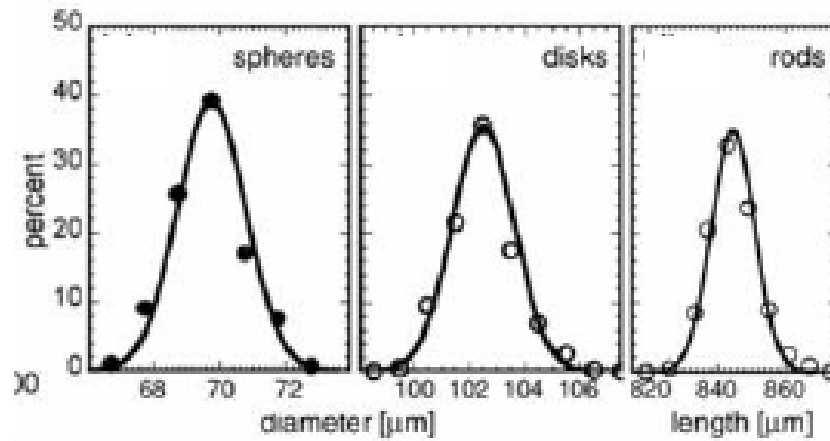
non-spherical particles



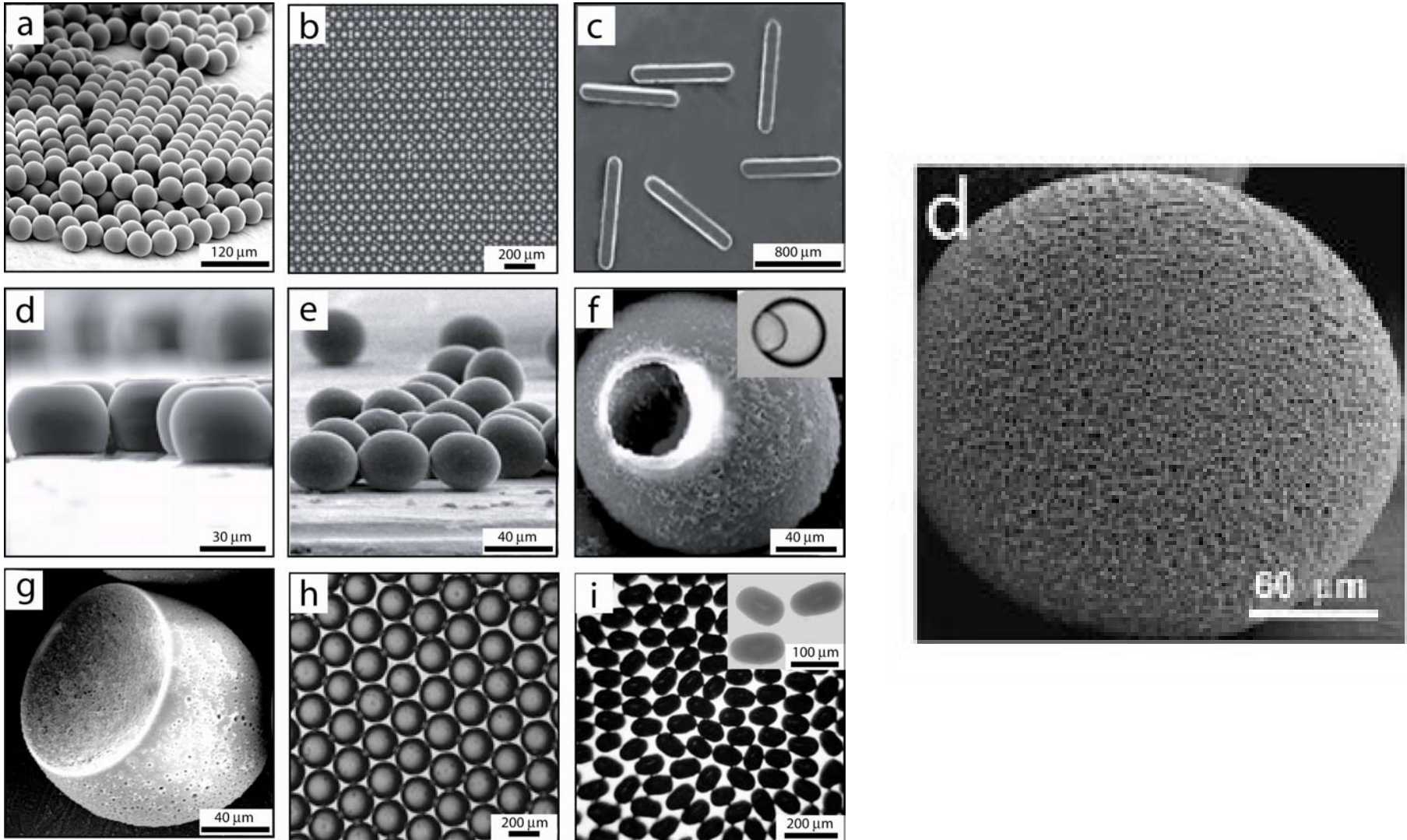
non-spherical particles



non-spherical particles



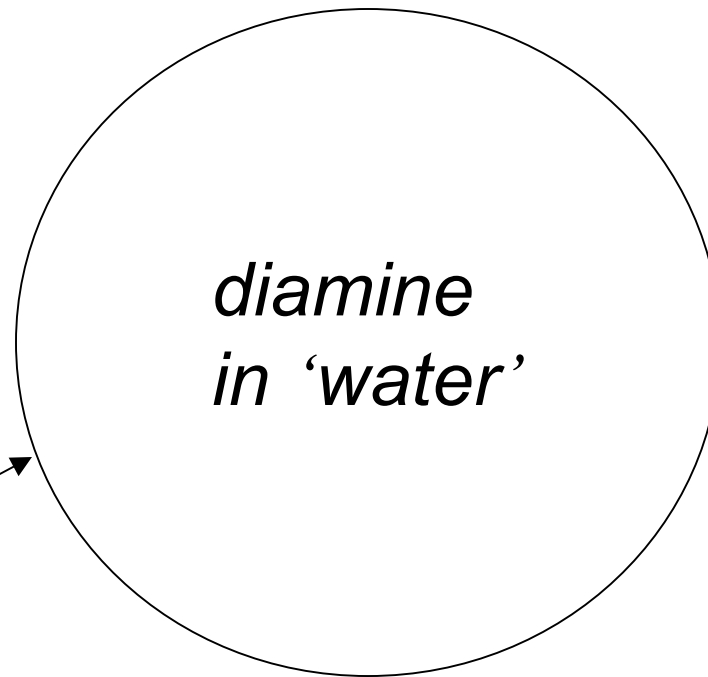
spheres, disks, rods, elipsoinds



polyTPGDA: (a) microspheres, (b) a colloidal crystal of microspheres (c) rods, (d) disks, (e) ellipsoids, (f) spherical capsules, and (g) truncated microspheres. (h) agarose disks and (i) bismuth alloy ellipsoids

interfacial polymerization

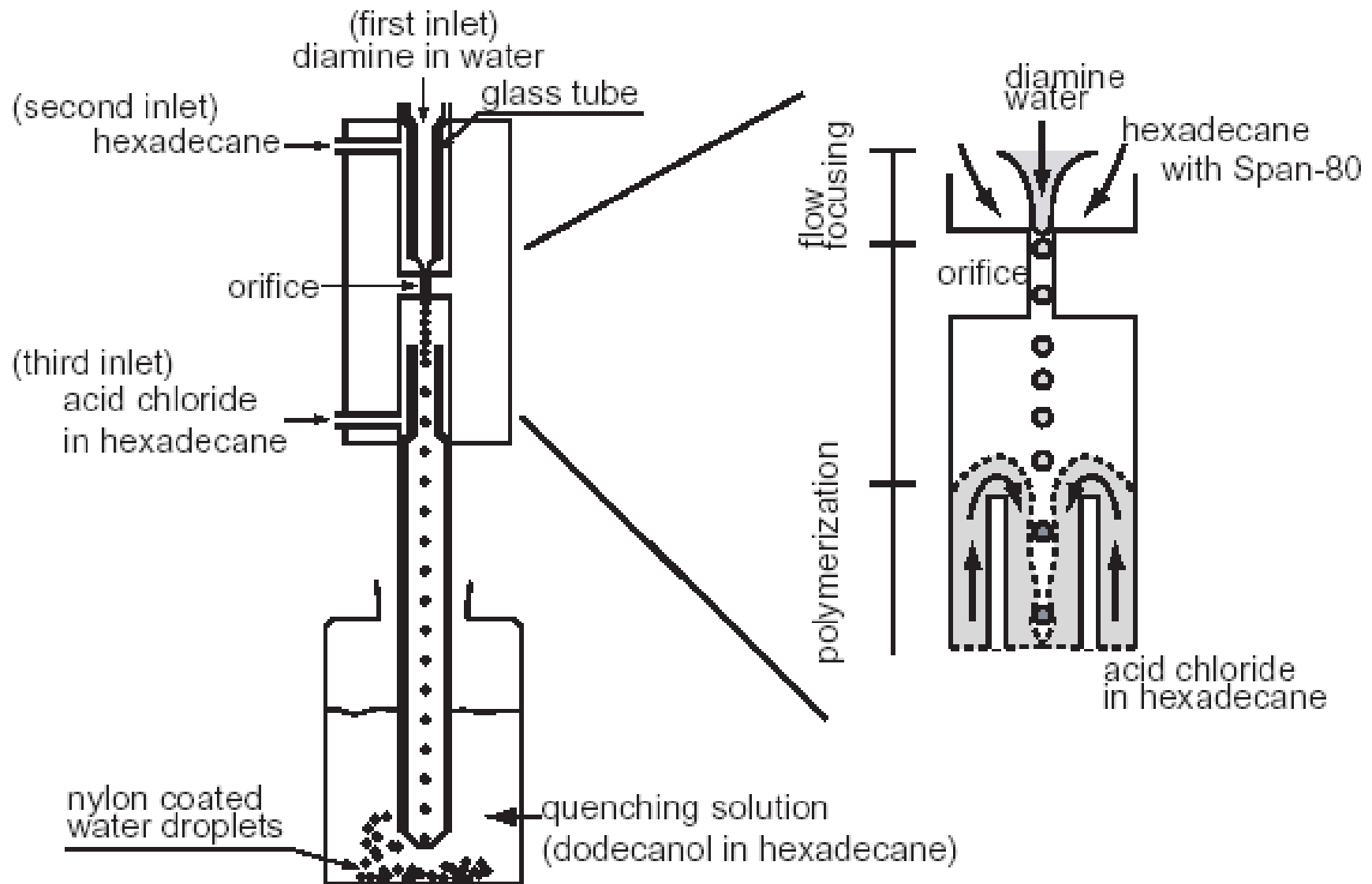
acid chloride in 'oil'



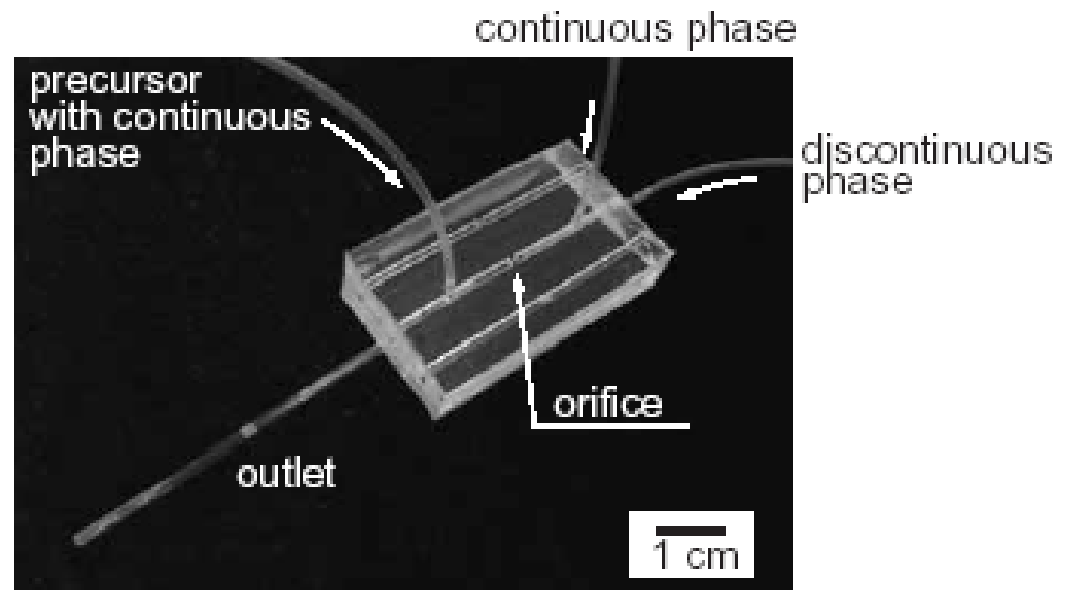
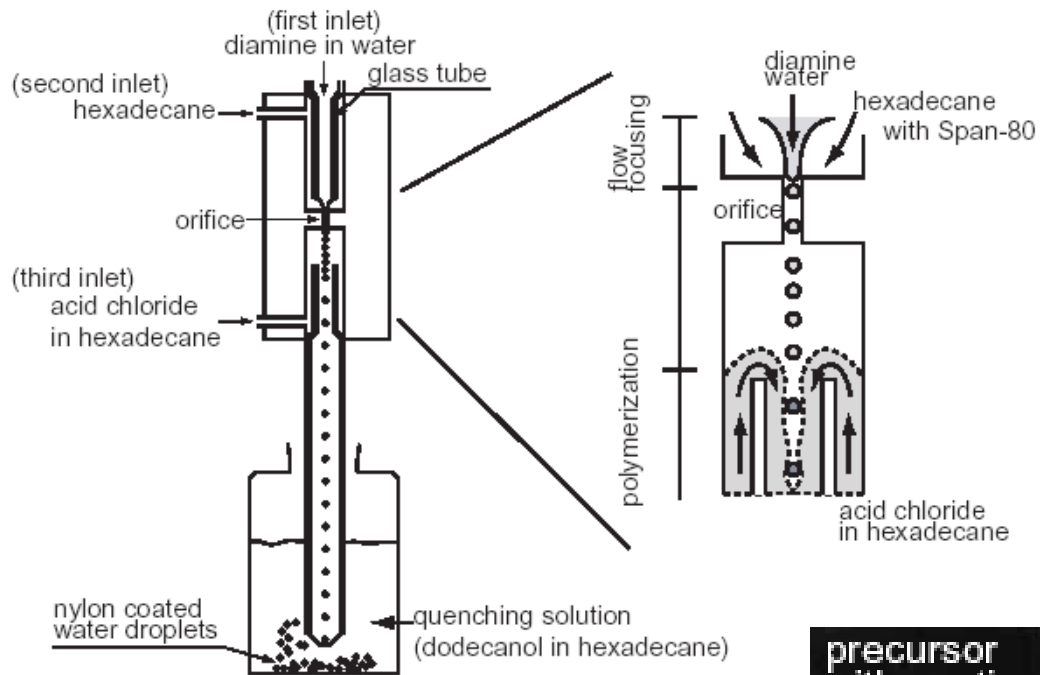
interfacial
polymerization

→ **semi-permeable
microcapsules**

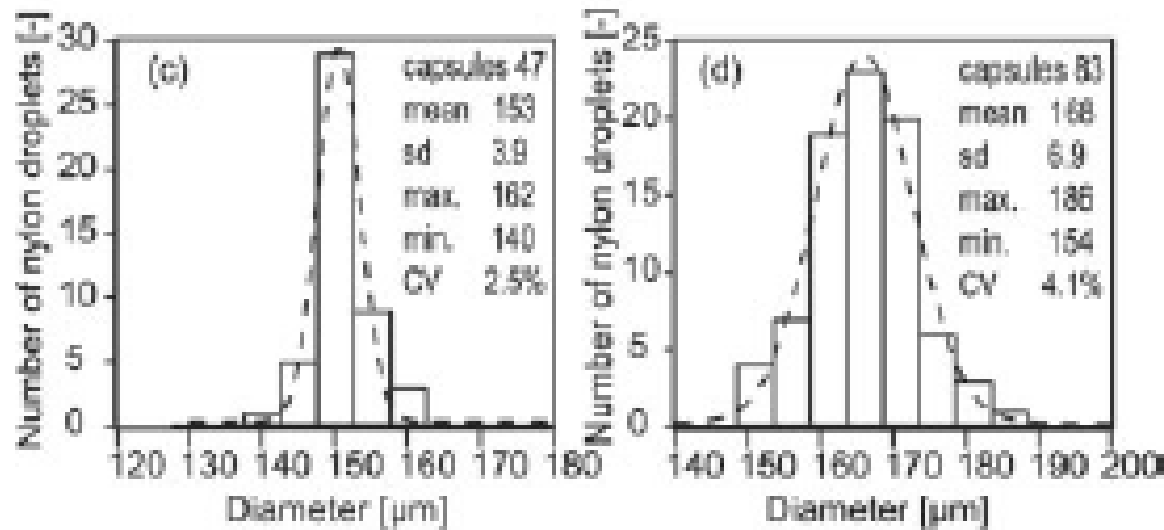
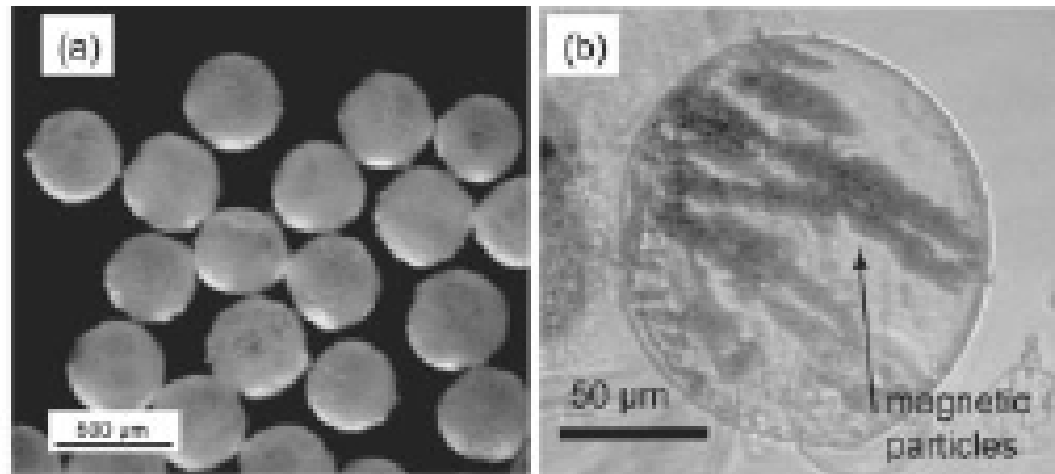
in-situ interfacial polymerization



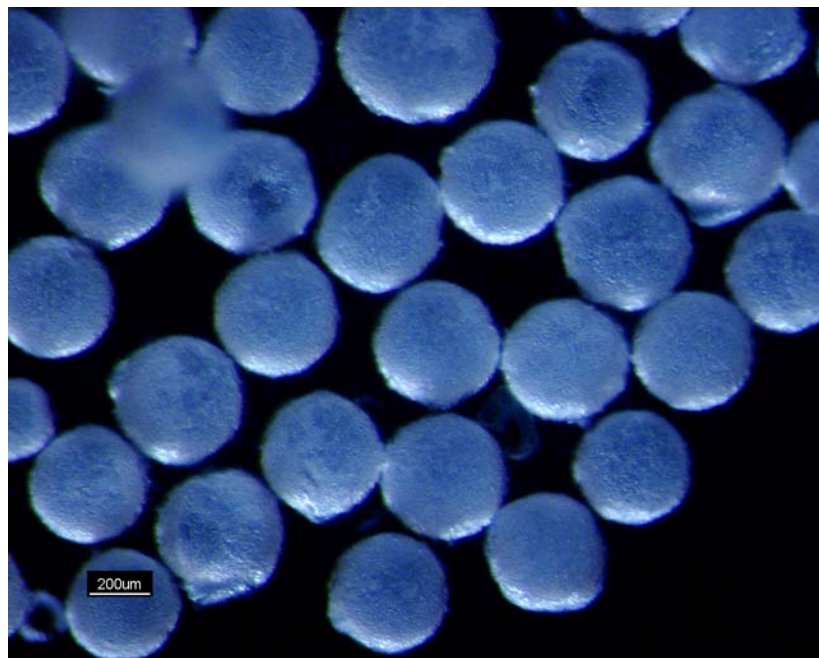
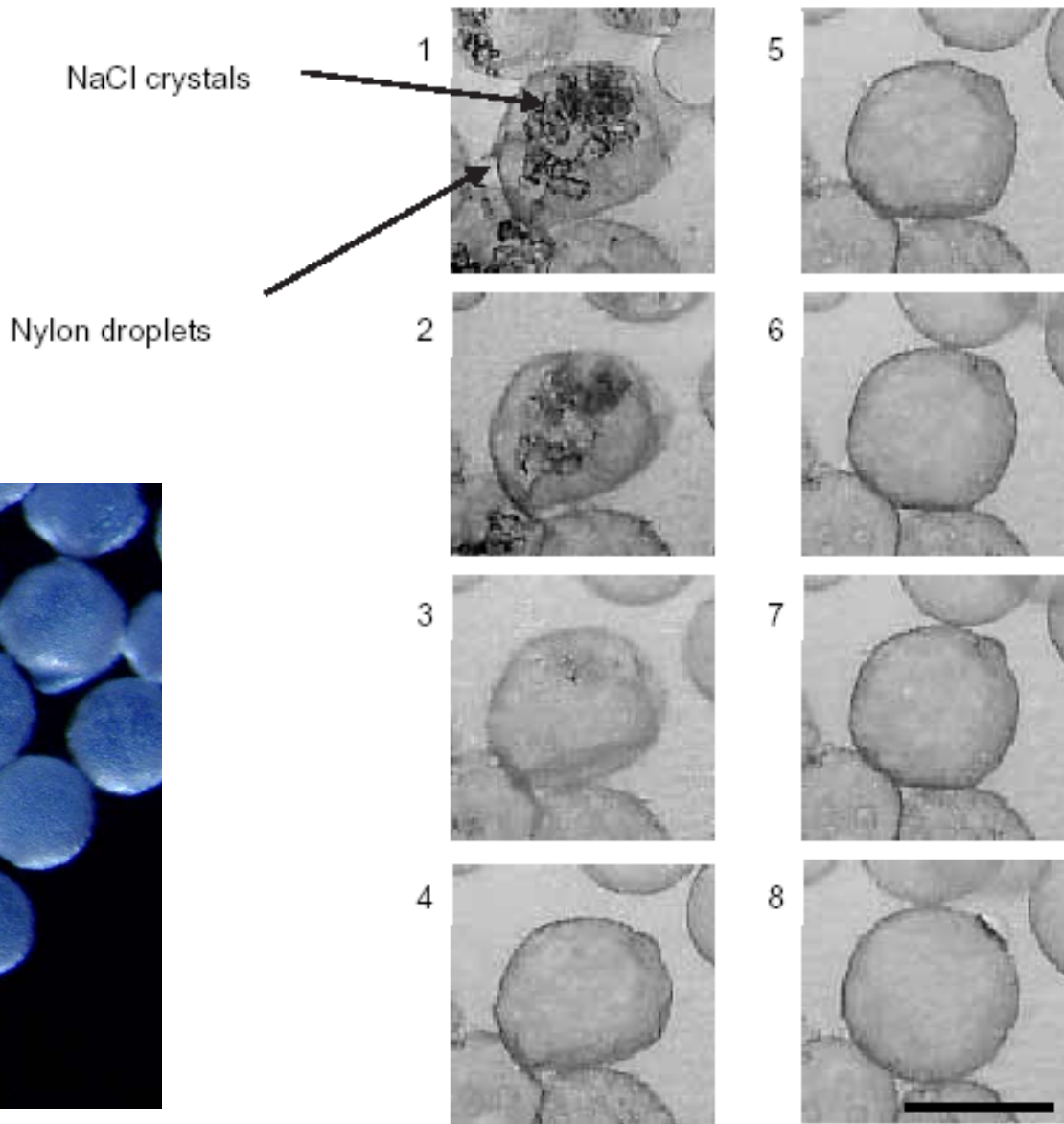
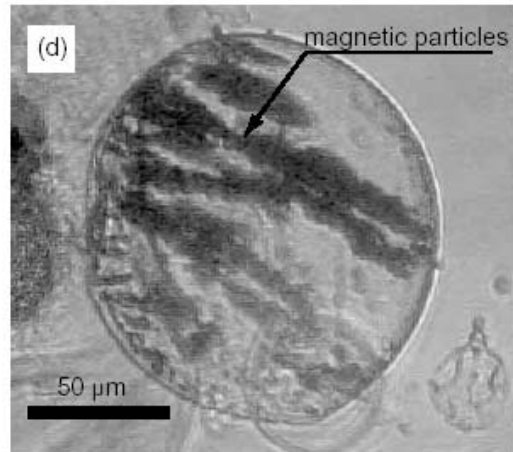
micro-encapsulation



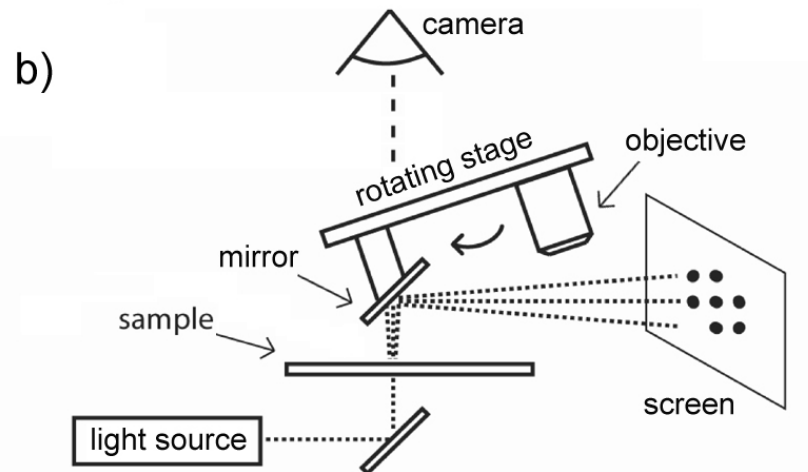
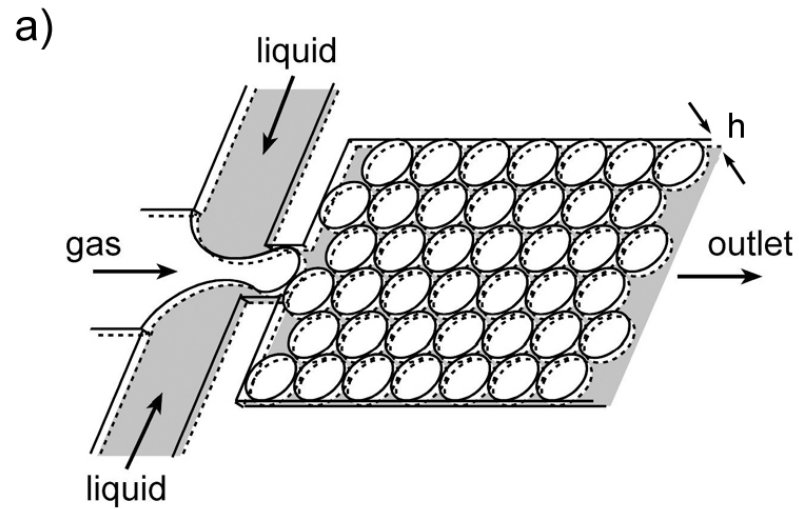
nylon capsules



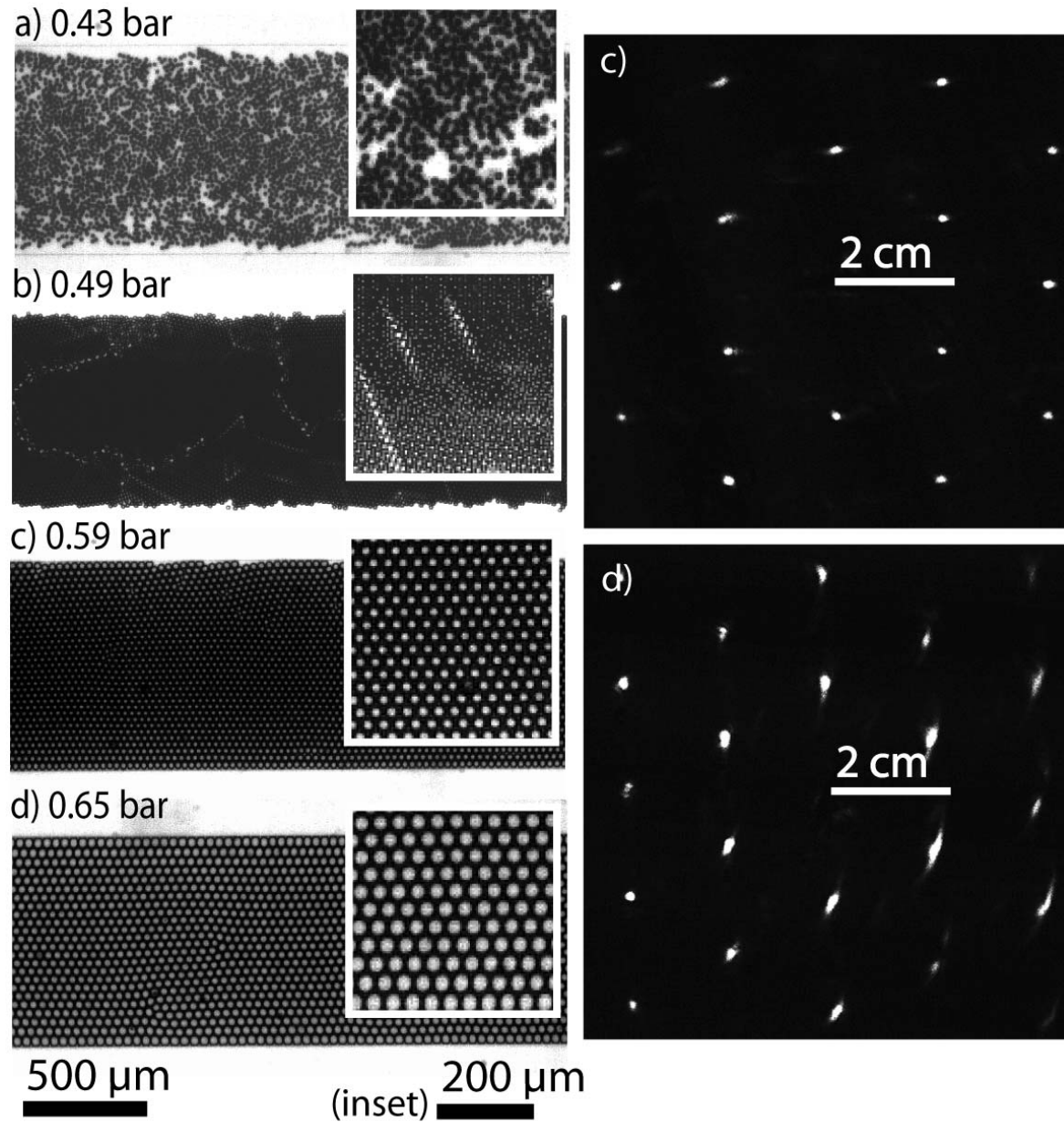
nylon capsules



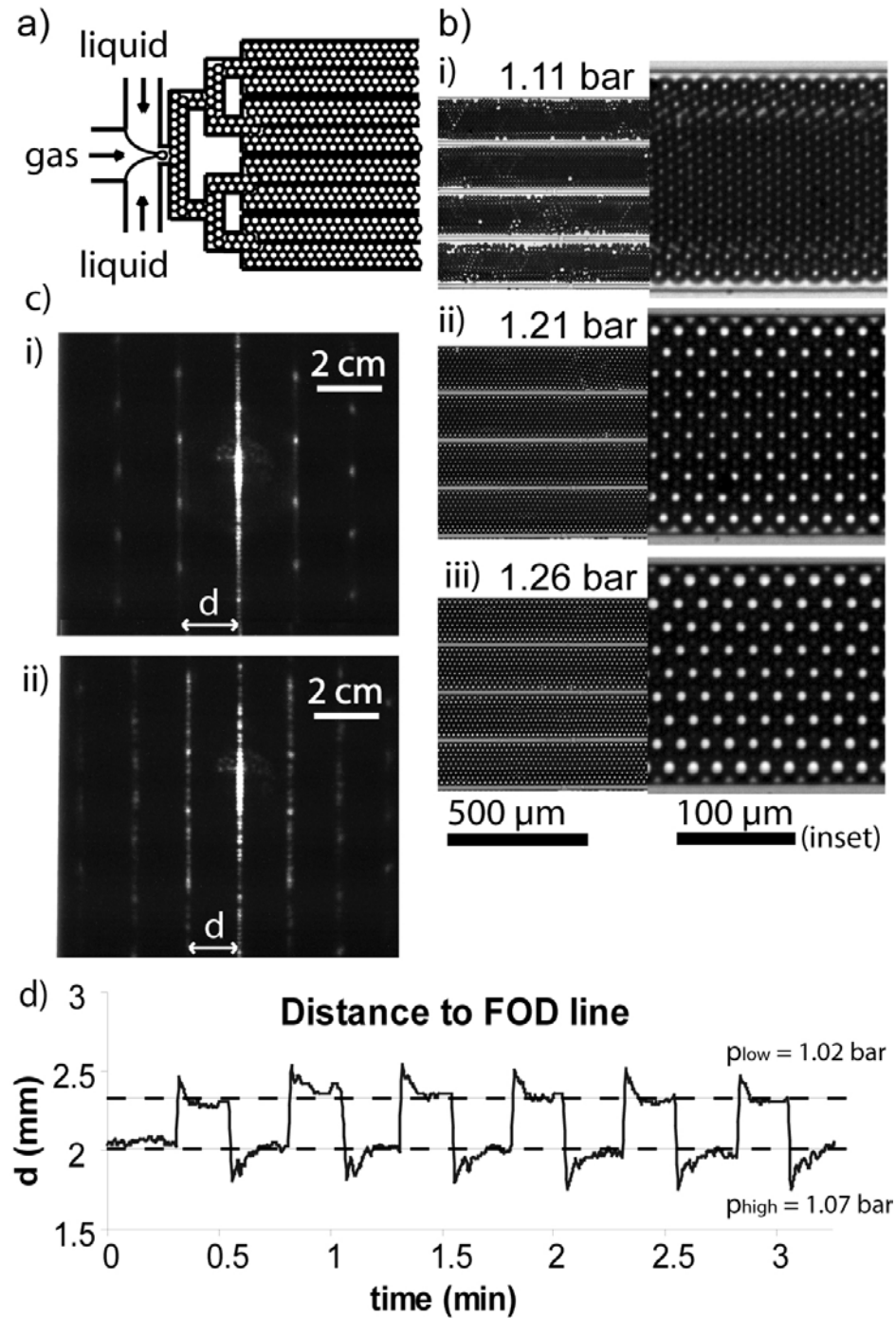
tunable diffraction gratings



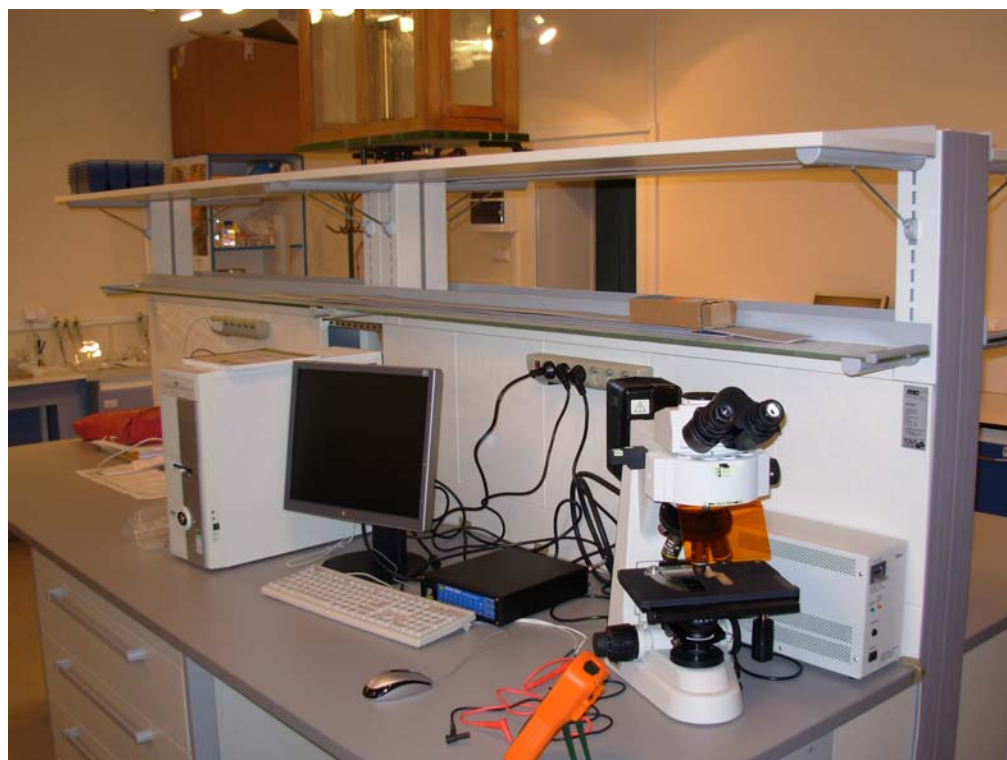
tunable diffraction gratings



tunable diffraction gratings



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- **applications**
 - micromixing, portable assays
 - micro-particles and micro-capsules
 - diffraction gratings



garst@ichf.edu.pl

acknowledgement

Prof. George Whitesides

- controlled formation of bubbles and droplets

**Irina Gitlin, Willow DiLuzio, Michael Furstman, Prof. Eugenia Kumacheva,
Prof. Howard Stone, M. De Menech, F. Jousse & Unilever**

- periodic bubbling

Michael Furstman, Michinao Hashimoto, Prof. Mahadevan

- stability

Prof. A. M. Ganan-Calvo & Sevilla

- mixing and simple solutions

Michael Fuerstman, Michael Fishbach, Sam Sia

- particles and capsules

Doug Weibel, Prof. Shoji Takeuchi, Irina Gitlin, Prof. Kumacheva & Toronto

- digital microfluidics

Adam Siegel, Derek Bruzewicz

- interfacial instabilities in a HeleShaw

Michinao Hashimoto, Prof. Howard Stone

- fluidic optics

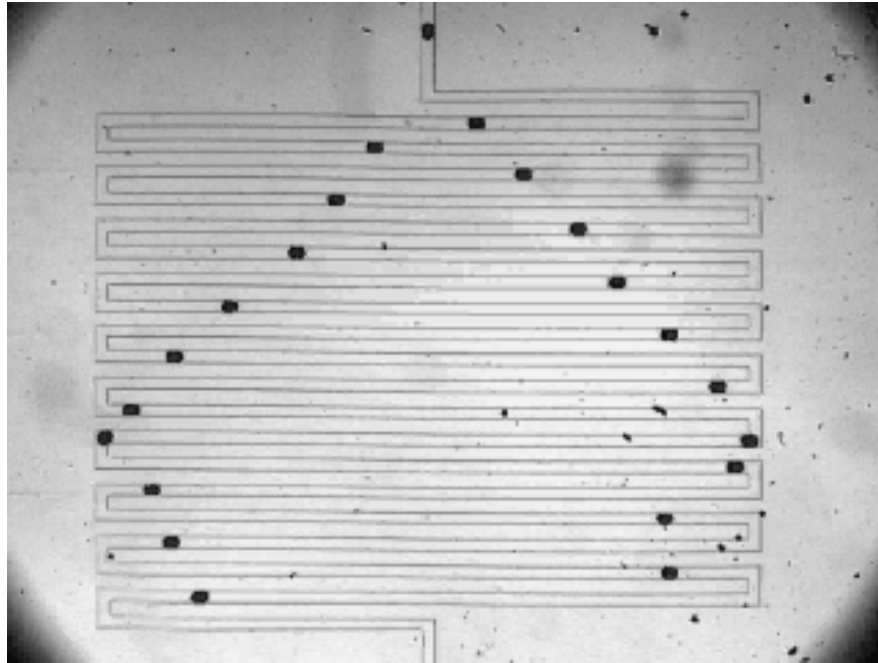
Brian Mayer, Michinao Hashimoto

- discussions

Weitz and Stone Groups, A. Ganan-Calvo

- funding

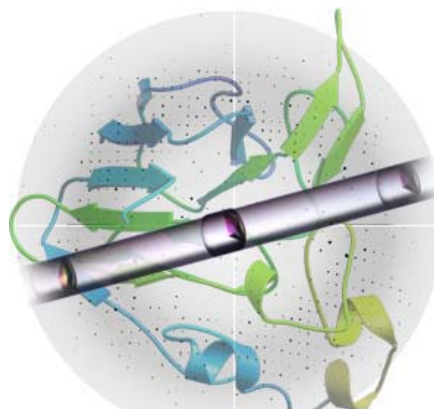
Foundation for Polish Science, U.S. Department of Energy



thank you

drops and bubbles in microfluidics

- control, reproducibility
- size, size distribution
- preparation of emulsions
- phenomena
- chemical kinetics, synthesis, materials
- analytical and portable systems



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a NanoReactor company

PRODUCTS

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RainDance Technologies proudly presents the DG4114 NanoReactor Platform.
RainDance Technologies NanoReactor Platform may be operated in conjunction with either K or S-type Chips ([see pdf](#)).

K-type Chips.
These chips mount in the DG4114 NanoReactor Platform to measure reaction kinetics.

S-type Chips.
These chips mount in the DG4114 NanoReactor Platform to perform sorting applications.