Many-body physics of 2D microfluidic droplet ensembles

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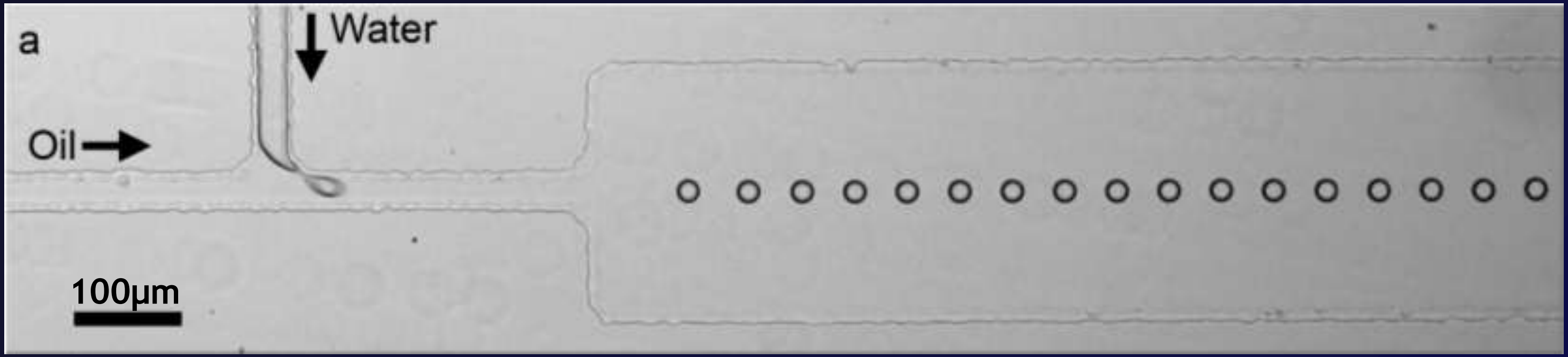
Squishy Physics, Harvard, 2012

Outline: many-body physics in 2D microfluidics

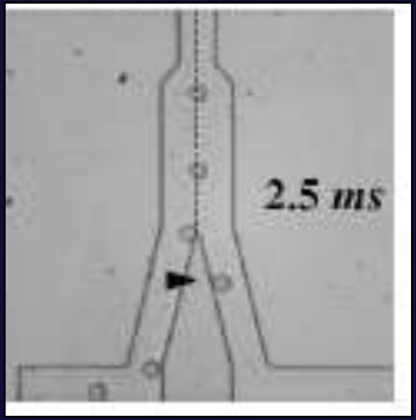
- μ-droplet ensembles in 2D: long-range, non-equilibrium dipoles.
- 1D crystals – “phonons” and instabilities.
- Long-range interactions and geometry: Phonons under confinement.
- 2D Disordered ensembles – Burgers shockwaves.

Nat Phys 2006, PRL 2007, PRL 2009, Phys Rep 2012

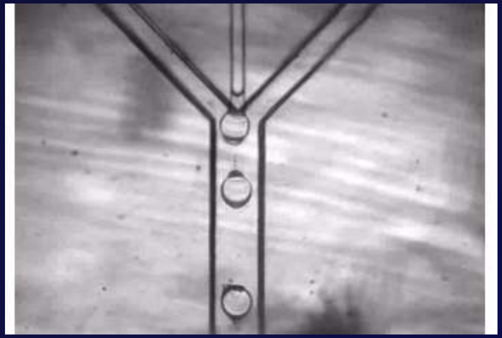
Engineering frontier with many applications: High-throughput droplet generators



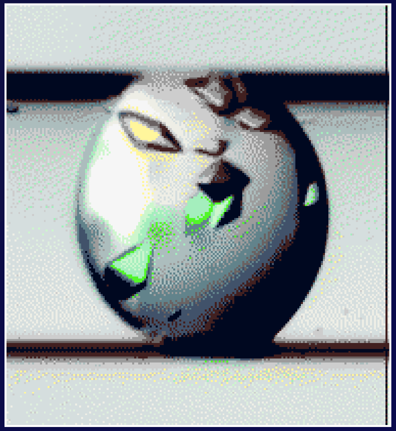
Mono-dispersed droplets



Droplet manipulation



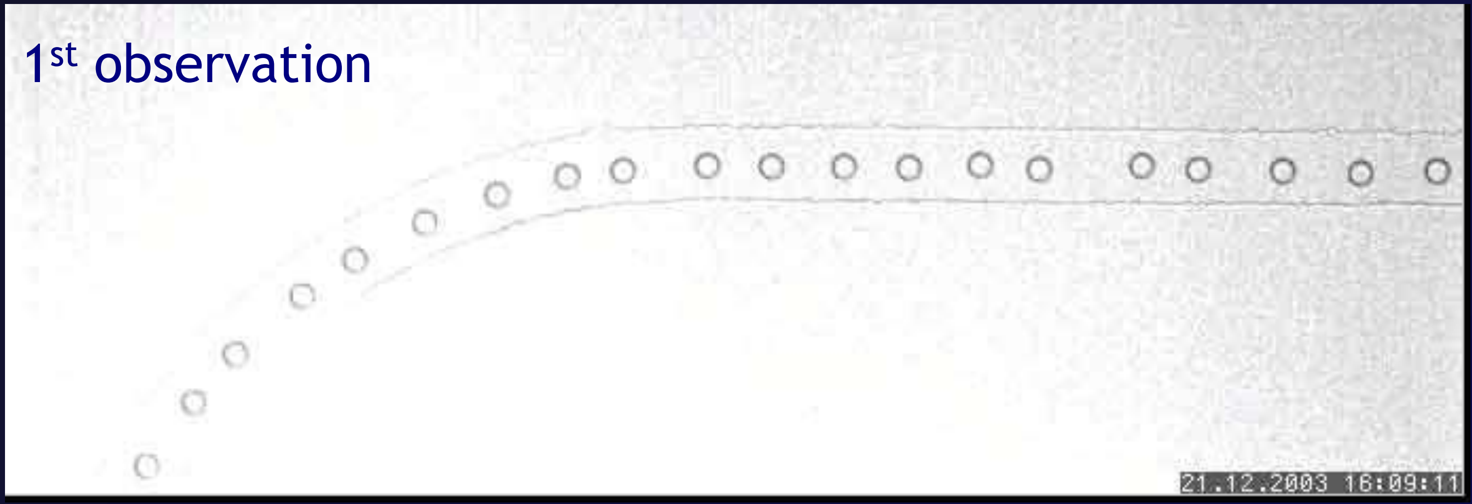
Double emulsions

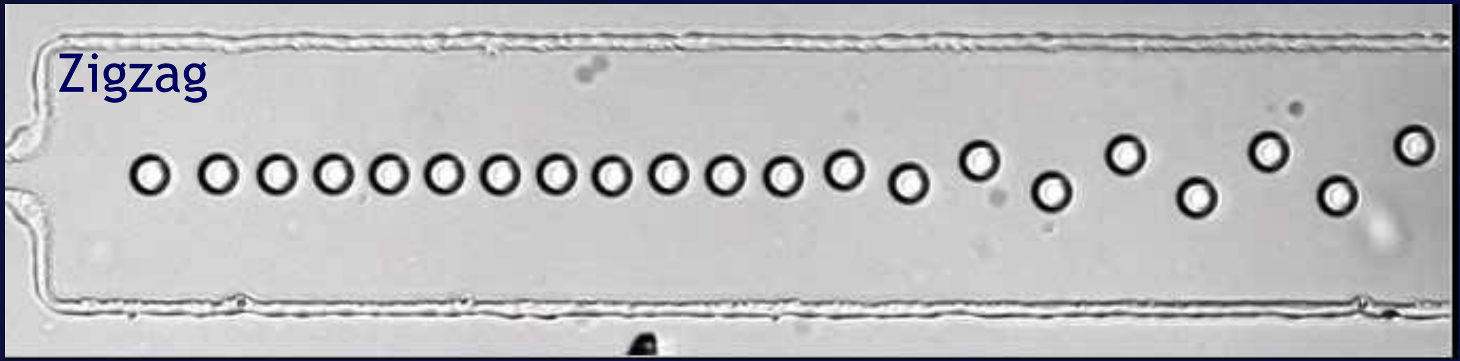


Bio-reactors

Quake, Stone, Tabeling, Weitz, Whitesides, Ismagilov…

Two experimental puzzles





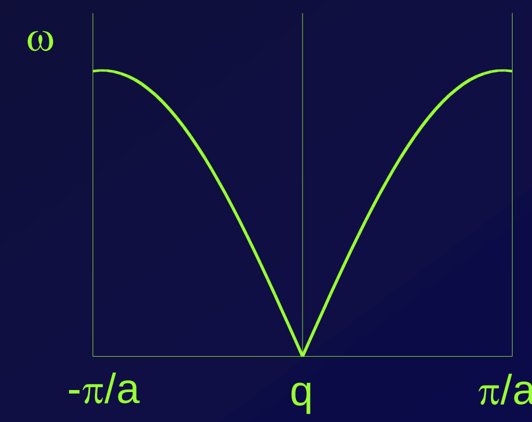
What is the underling physics?

Can waves persist in viscous media?

Harmonic crystal   Friction

$$m\cancel{\frac{\partial^2 r}{\partial t^2}} = \kappa \frac{\partial^2 r}{\partial x^2} + \mu \frac{\partial r}{\partial t}$$

$$\omega \sim \sqrt{\kappa / m}\,|q|$$



$m \to 0$, no waves

$$\omega \sim i(\kappa / \mu) q^2$$

can get waves if

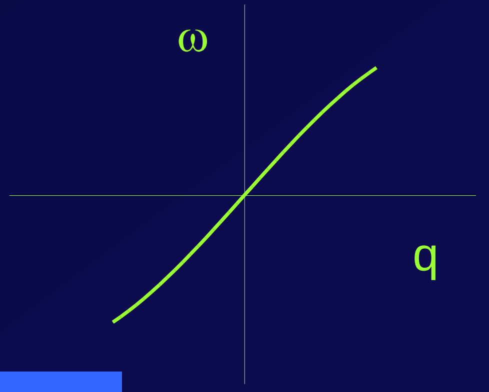
$$\mu^2 < \kappa m / a^2$$

No mass + symmetry-breaking field

$$0 = \lambda \frac{\partial r}{\partial x} + \mu \frac{\partial r}{\partial t}$$

Waves

$$c_s = \frac{\omega}{q} = \frac{\lambda}{\mu}$$



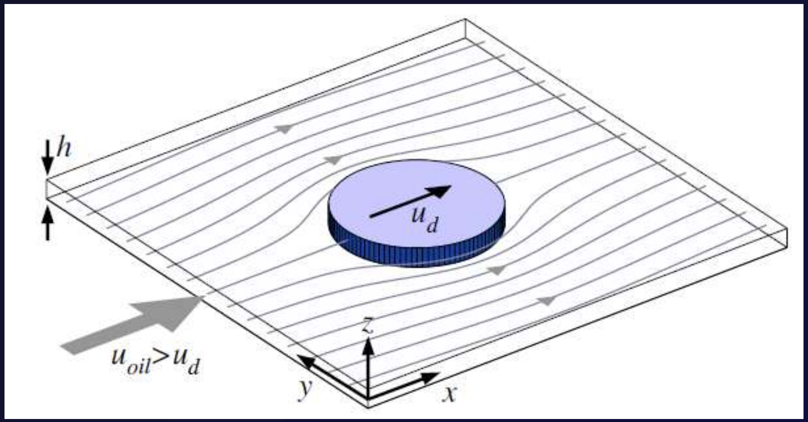
Symmetry-breaking → Waves

What force could break symmetry?

Suggestion: Hydrodynamics

Driven pancakes are hydrodynamic dipoles

Effective potential flow in 2D



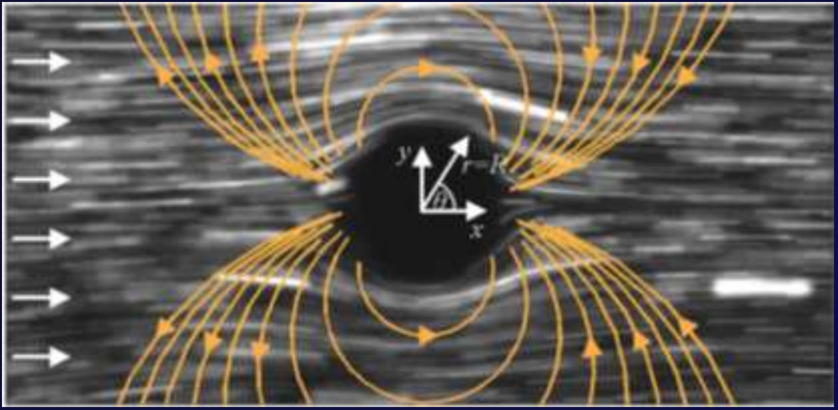
Stokes $\eta \nabla^2 \mathbf{v} = \nabla P$

Lubrication $\mathbf{v} = \left(1 - 4z^2/h^2\right) \cdot \mathbf{u}(x, y)$

Incompressible $\nabla \bullet \mathbf{v} = 0$

2D Laplace $\Rightarrow \nabla^2 \phi = 0, \quad \mathbf{u} = \nabla \phi$

Dipole field



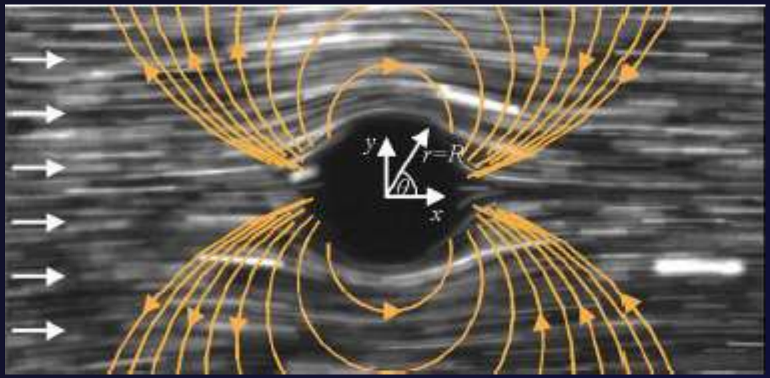
$$\phi(\mathbf{r}) = \phi_{\text{uniform}} + \phi_d(\mathbf{r})$$

$$\phi_d(\mathbf{r}) = \frac{\mathbf{d} \cdot \hat{\mathbf{r}}}{r}; \qquad \mathbf{d} = R^2 \delta u$$

$$\delta u = u_{oil}^{\infty} - u_d$$

complex potential is simple pole: $w(z) = \phi + i\psi = d\,\dfrac{1}{z}$

Hydrodynamic drag force on a single dipole



$$\xi \sim \eta h^{-1} R^2$$

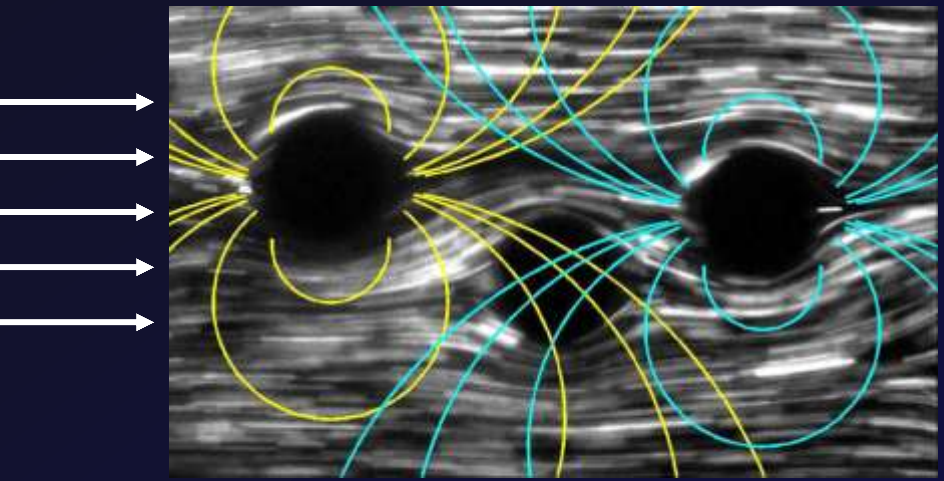
$$\mathbf{F}_{drag} = -\oint_D P dS = \frac{1}{2}\xi \mathbf{u}_d + \xi\left(\mathbf{u}_{\text{oil}}^{\infty} - \mathbf{u}_d\right)$$

Force of Poiseuille pressure gradient

Force due to dipolar field

$$\mathbf{F}_d = \xi R^{-2} \cdot \mathbf{d}$$

Hydrodynamic interaction is dipole-monopole



- Dipoles perturb flow field around other droplets.

→ Long range drag forces

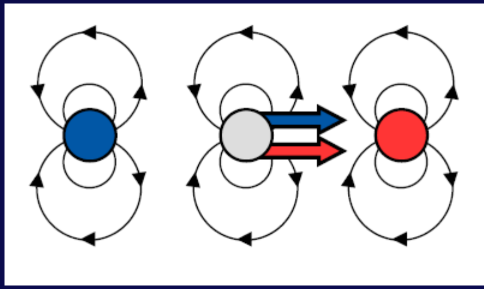
$$\mathbf{F}_{ij} = \xi \nabla \phi (\mathbf{r}_i - \mathbf{r}_j) \sim \xi \delta u_j \left(\frac{R}{r_{ij}} \right)^2$$

$$\xi \sim \eta h^{-1} R^2$$

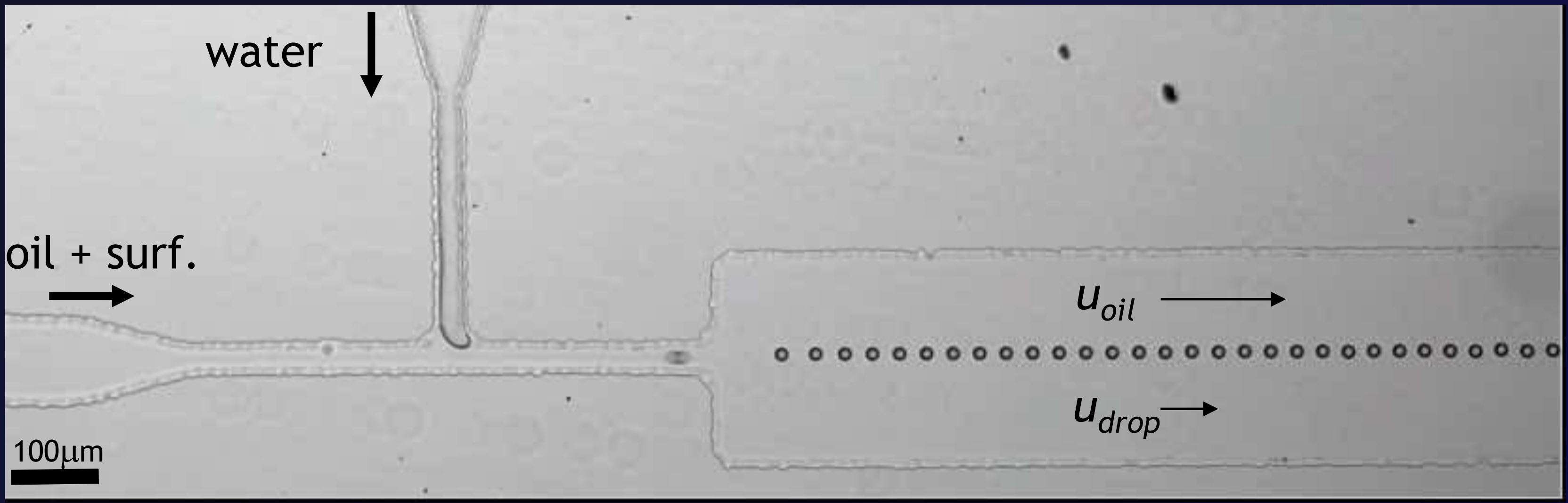
- Overall force is linear superposition $\mathbf{F}_i \propto \sum_j \mathbf{F}_{ij}$

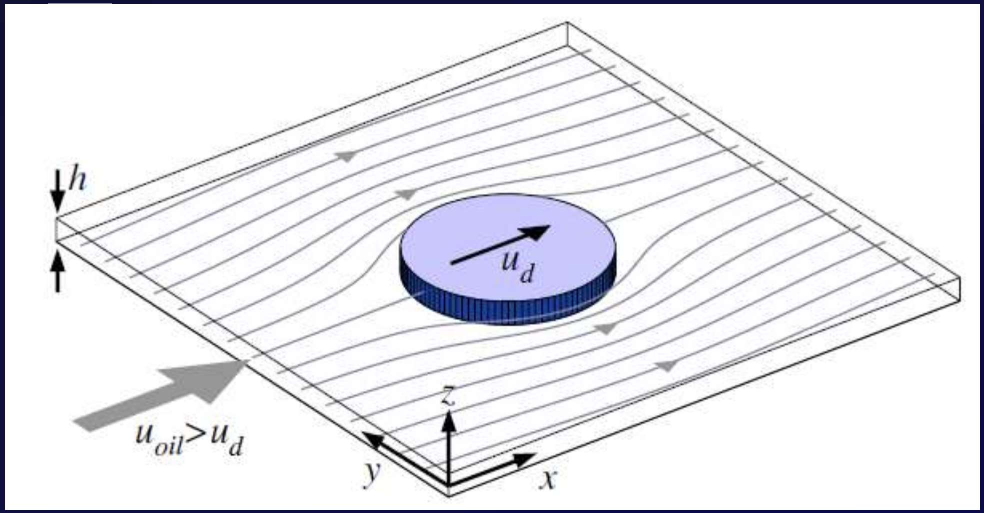
- In crystals symmetry-breaking forces: $\mathbf{F} \propto \frac{\partial r}{\partial x}$

$$\text{Waves:} \quad C_s \sim \frac{\lambda}{\mu} \sim \delta u \left(\frac{R}{a} \right)^2$$



Testing simple theory in 1D droplet crystal





Typical scales:

h = 10 μm

R = 10-15 μm

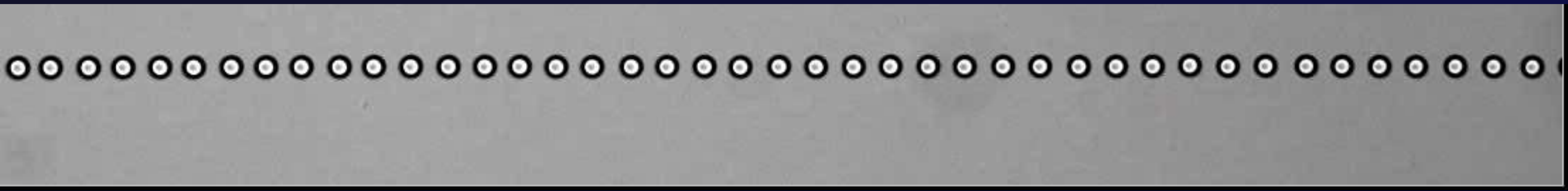
u_d = 100-500 μm/s

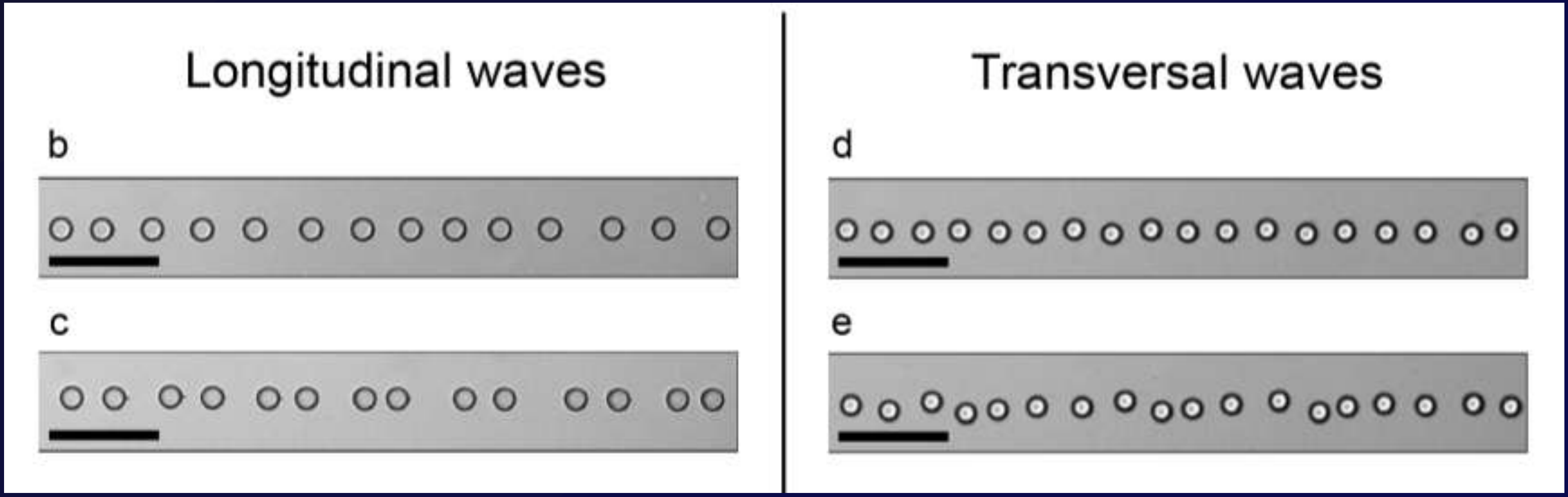
$u_{oil} \sim 4\, u_d$

Re = $uL\rho/\eta \sim 10^{-3}$

Crystals exhibit normal modes of vibration

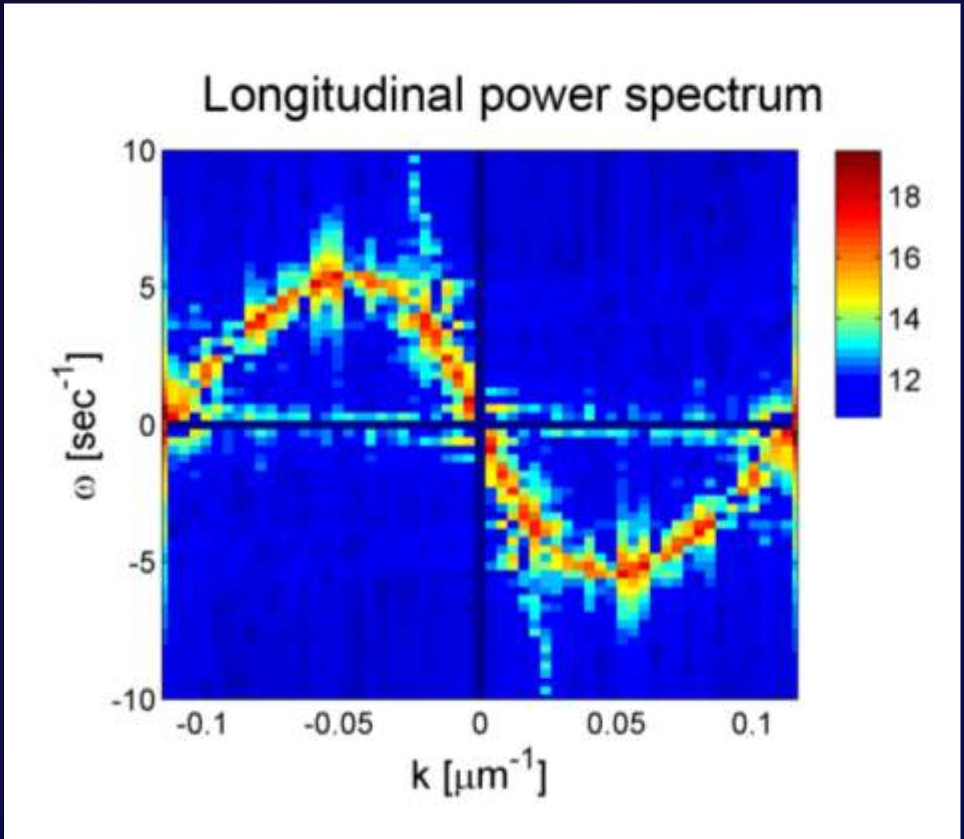
Moving with the crystal





‘Phonon’ spectra of the 1D droplet crystal

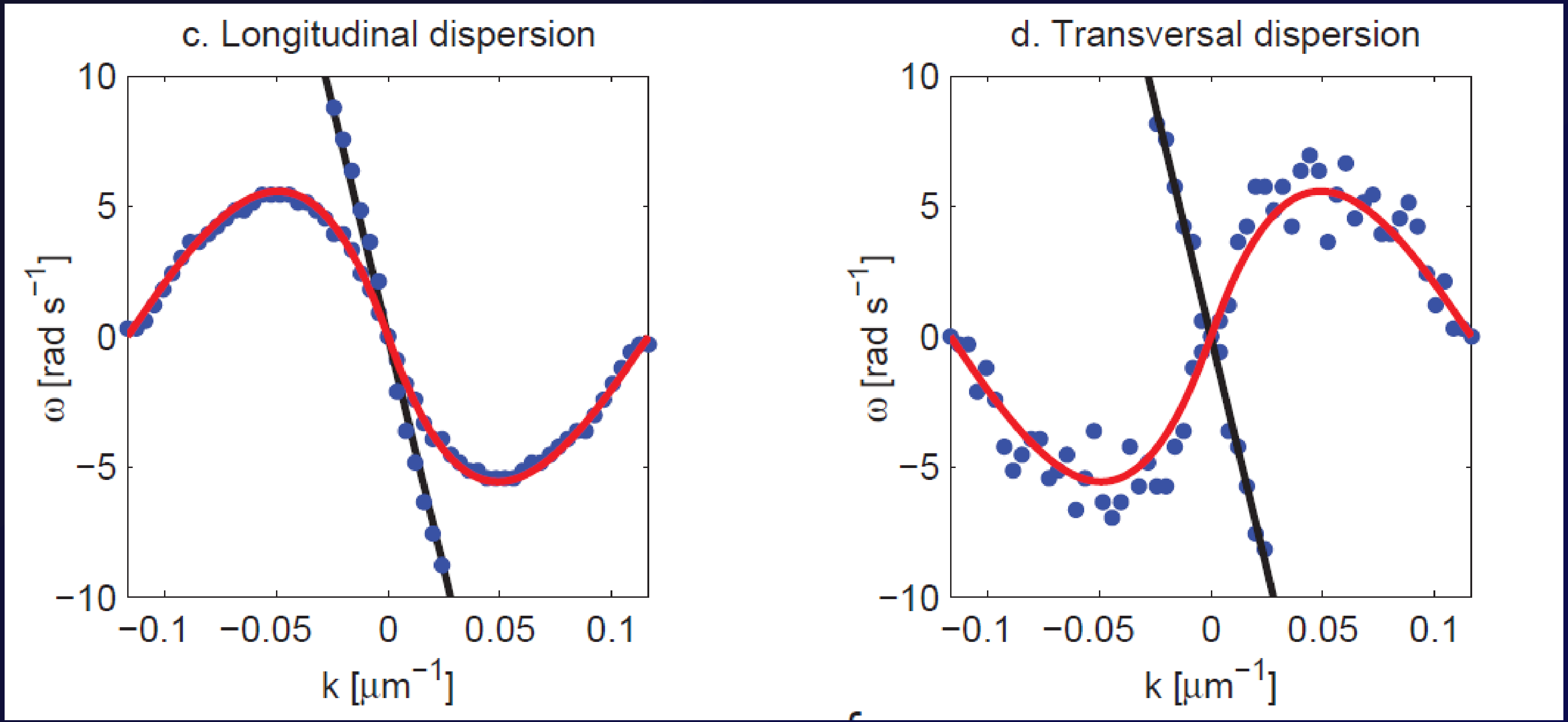
$$A_n(t) \rightarrow \left|A(k,\omega)\right|^2$$



N = 60
T = 20s
L = 1cm

Spectrum implies predcited symmetry breaking

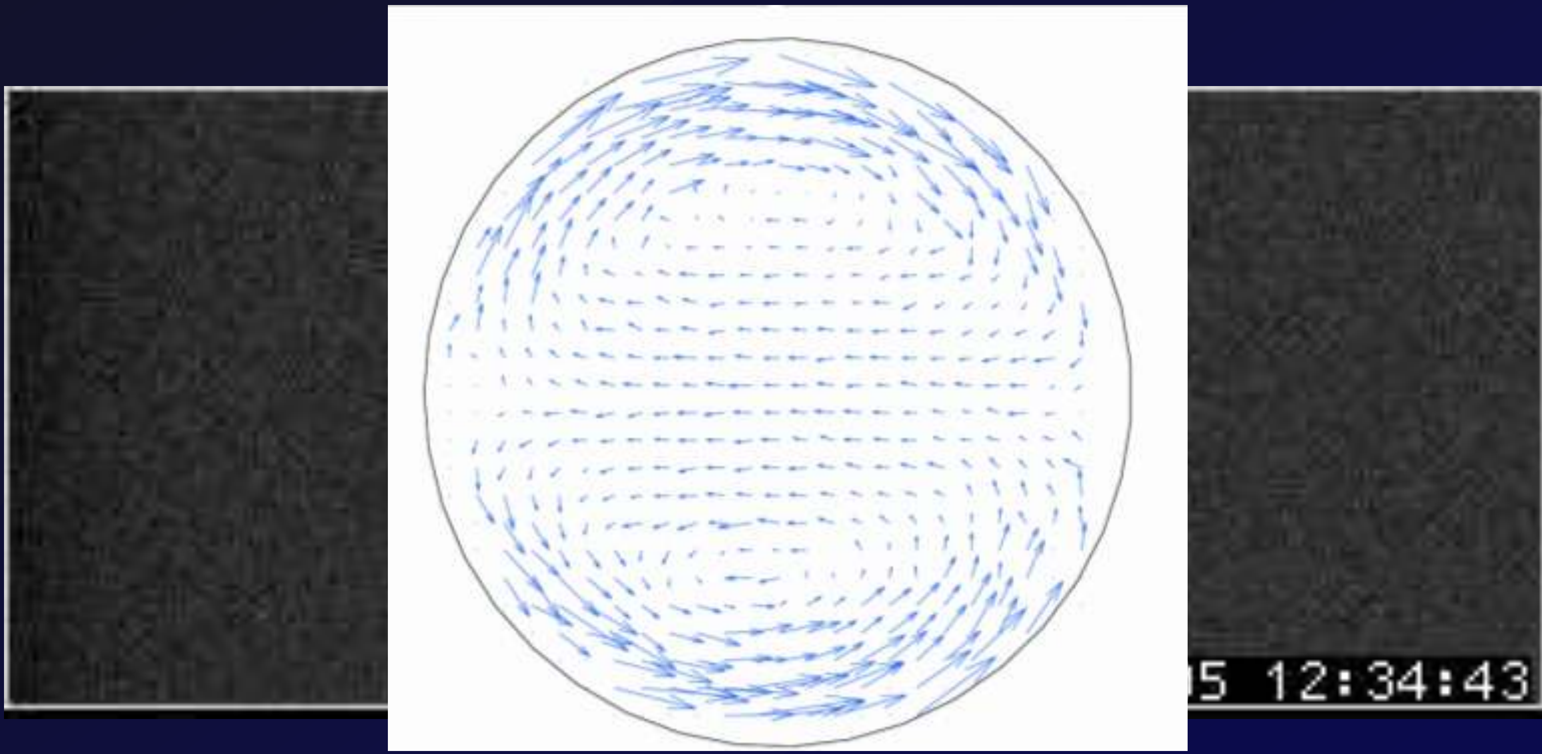
Dispersion relations hint symmetry breaking



$$\omega_x(k) = -\omega_y(-k)$$

$a = 27\mu m \quad R = 10\mu m \quad u_d = 360\mu m/\sec \quad u_{oil} = 1700\mu m/\sec$

Opposing force: friction



Flow inside droplets → Energy dissipation → Friction

$$\mathbf{F}_{friction} = \mu \mathbf{u}_d$$

Drag and friction balance out → eq. of motion

$$\mathrm{Re} << 1 \longrightarrow F = ma = 0$$

$$\mathbf{F}_{drag} = \tfrac{1}{2}\xi \mathbf{u}_d + \xi\left(\mathbf{u}_{oil} - \mathbf{u}_d\right) = \mathbf{F}_{friction} = \mu \mathbf{u}_d$$

Equation of motion of a droplet in an ensemble:

$$\dot{\mathbf{r}} = \mathbf{u}_d = K\,\mathbf{u}_{oil}(\mathbf{r}); \quad K = \left(\frac{1}{2} + \frac{\mu}{\xi}\right)^{-1}$$

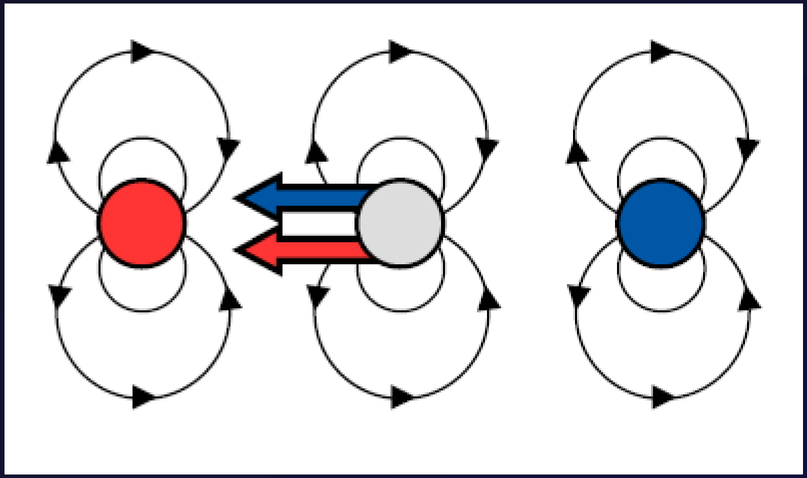
All the interactions are here

$$\mathbf{u}_{oil}(\mathbf{r}) = \nabla \phi_{\text{ensemble}}$$

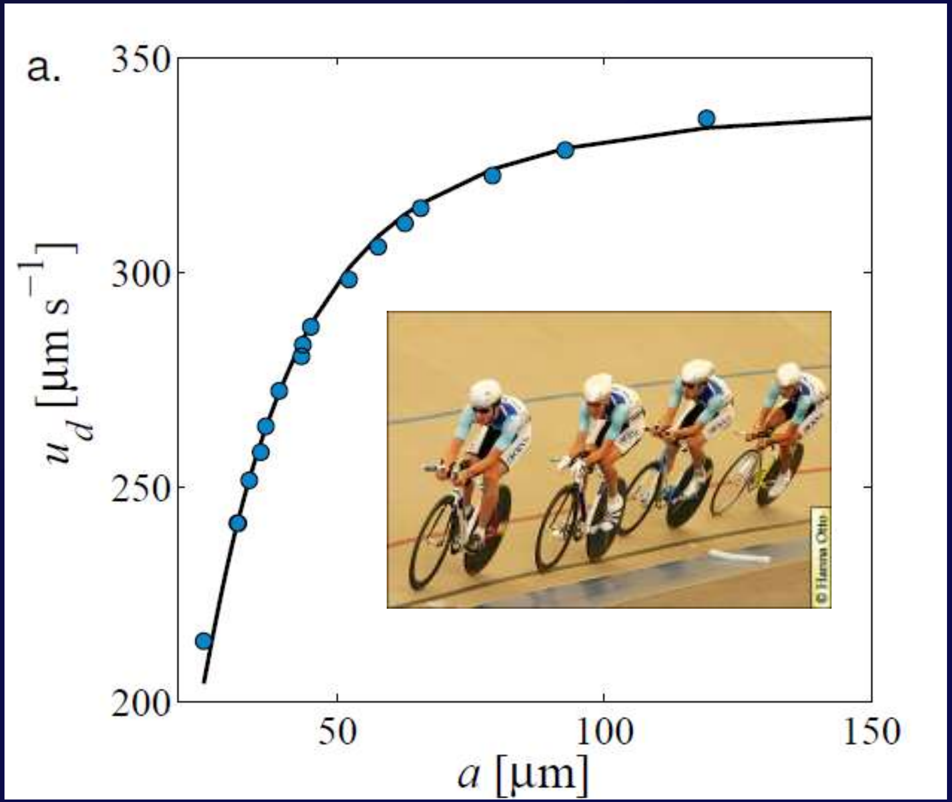
Calibrate for an isolated droplet

$$\left(\frac{u_d^\infty}{u_{oil}^\infty}\right) = K$$

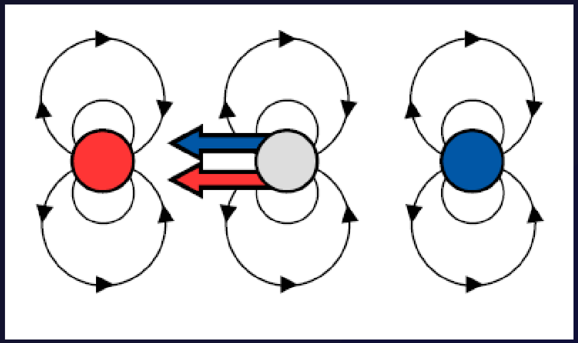
Droplet interaction: 'Peloton' effect



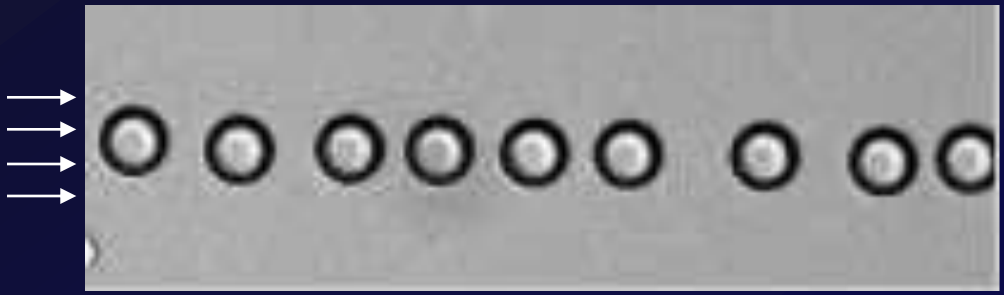
- Interactions slow down the crystal.
- Crystal moves faster w.r.t. oil
- Similar to sedimenting particles and cyclists → the 'Peloton' effect.

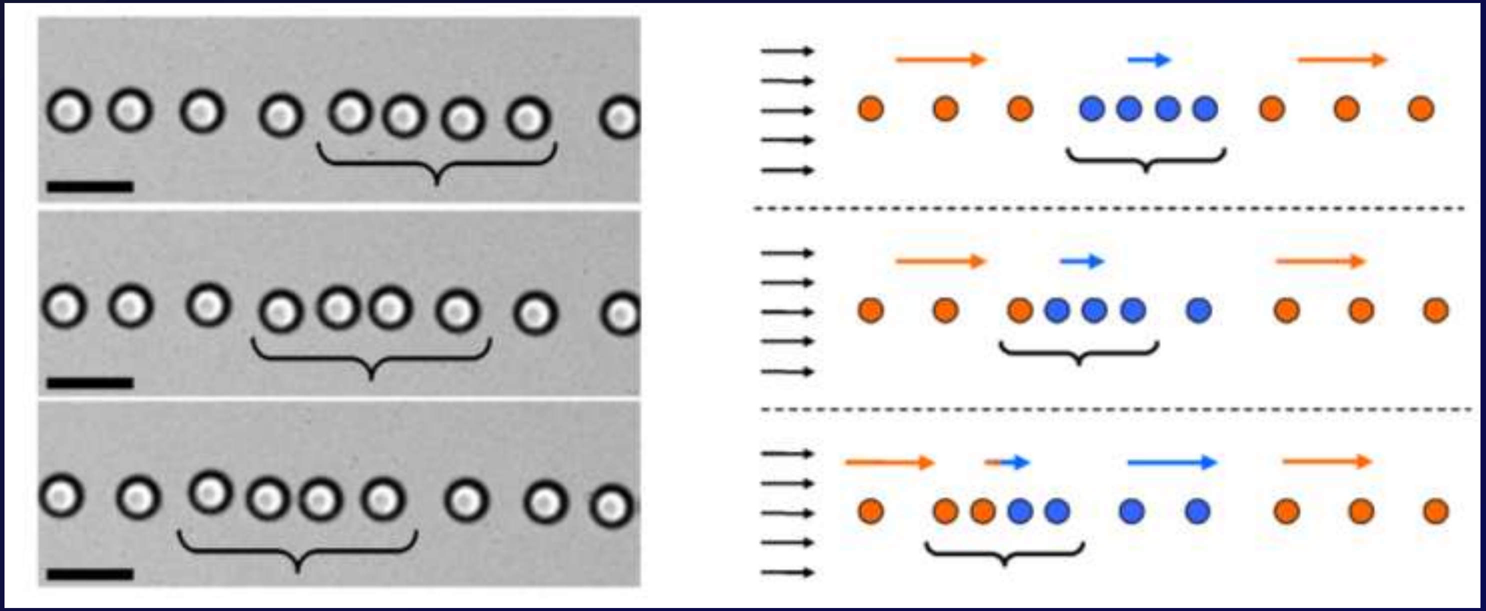


Waves intuition



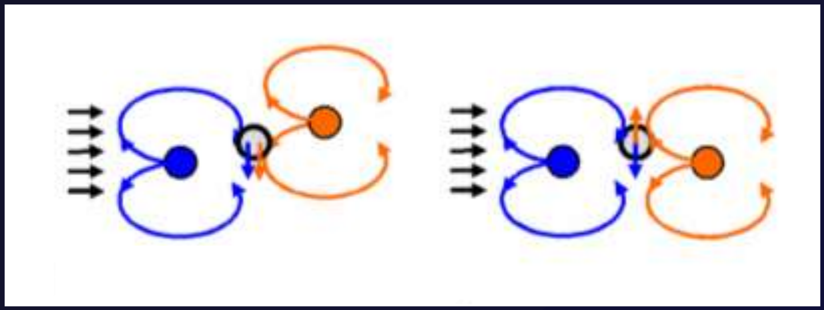
A dense crystal moves slower

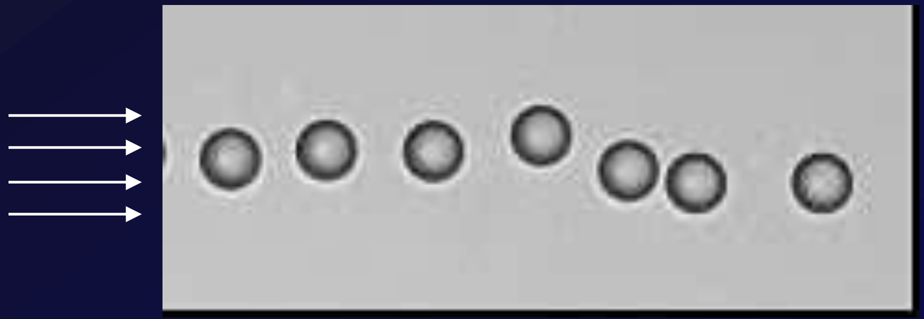


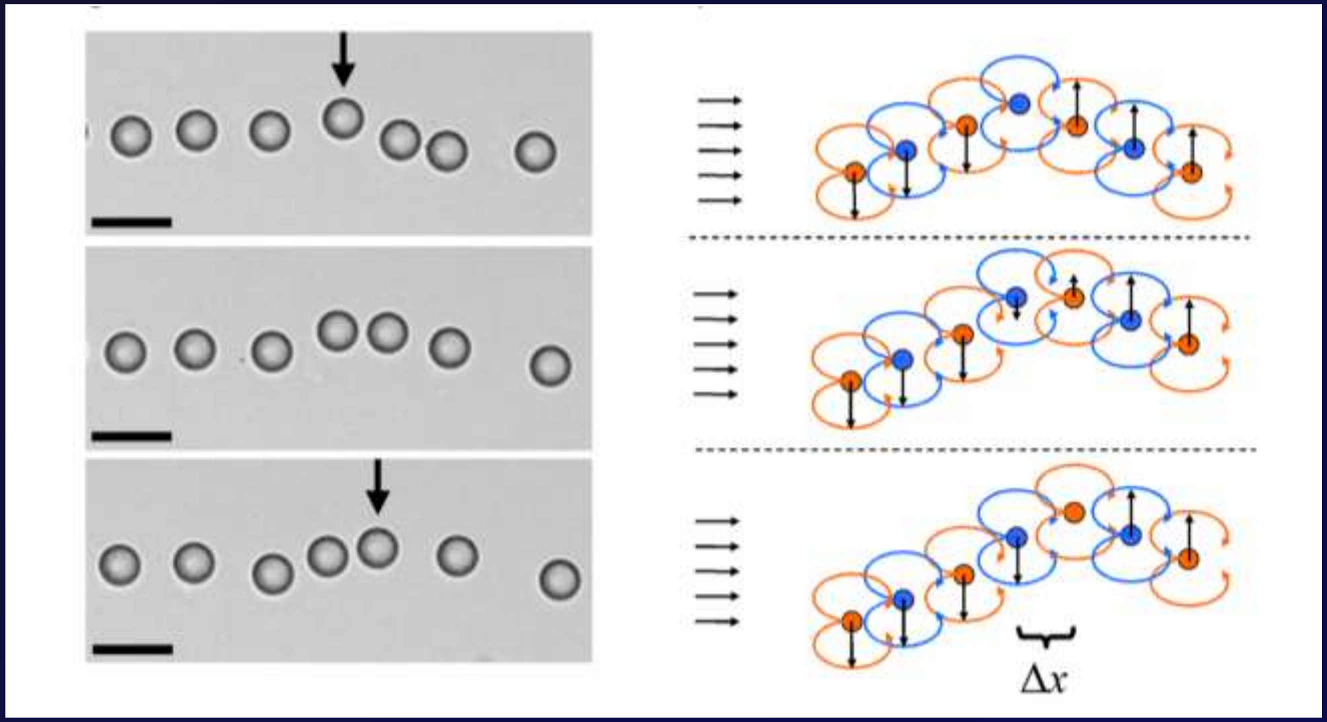


Long longitudinal waves travel **against** flow

Dipolar interaction leads to transversal waves







Long transversal waves travel **with** the flow

Wave equations and spectra agree with theory

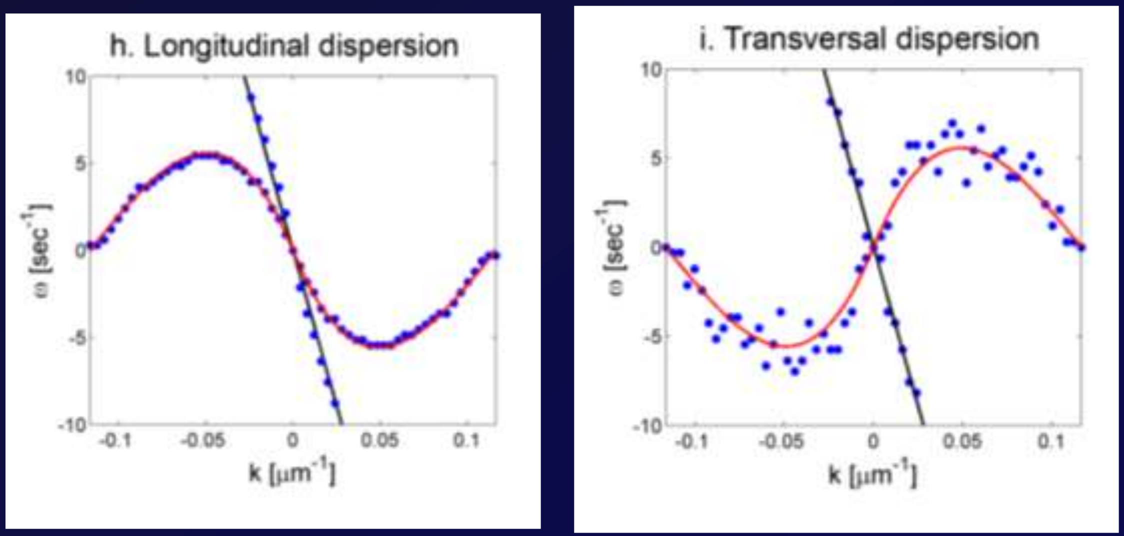
Superposition of dipole fields on a lattice $\mathbf{r}_n = (na, 0)$ + small vibrations

Eq. of motion $\dot{\mathbf{r}}_n = K\,\mathbf{u}_{oil}(\mathbf{r}_n)$ $\xrightarrow{x,\,y \,<<\, a}$

$$\omega_x(k) = -\frac{6C_s}{\pi^2 a}\sum_{j=1}^{\infty}\frac{\sin(jka)}{j^3}$$

$$\omega_y(k) = -\omega_x(k)$$

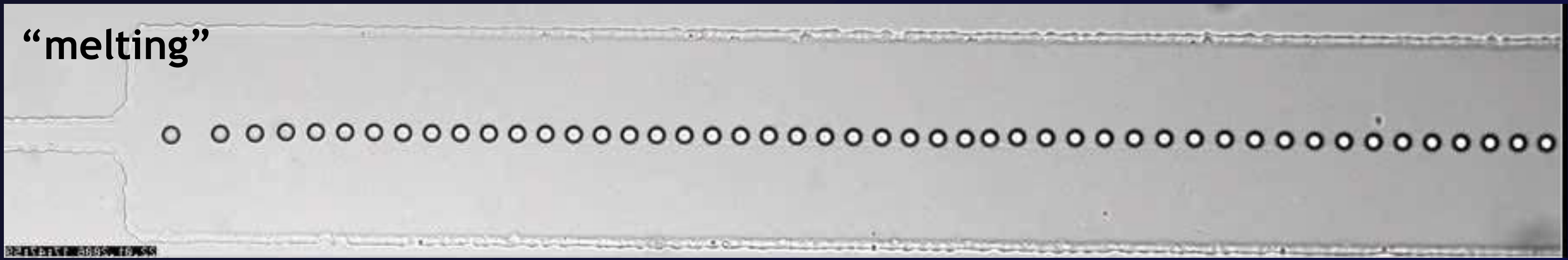
Sound velocity $C_s = \left(\frac{2\pi^2}{3}\right) K \left(\frac{R}{a}\right)^2 \left(u_{oil}^{\infty} - u_d^{\infty}\right)$

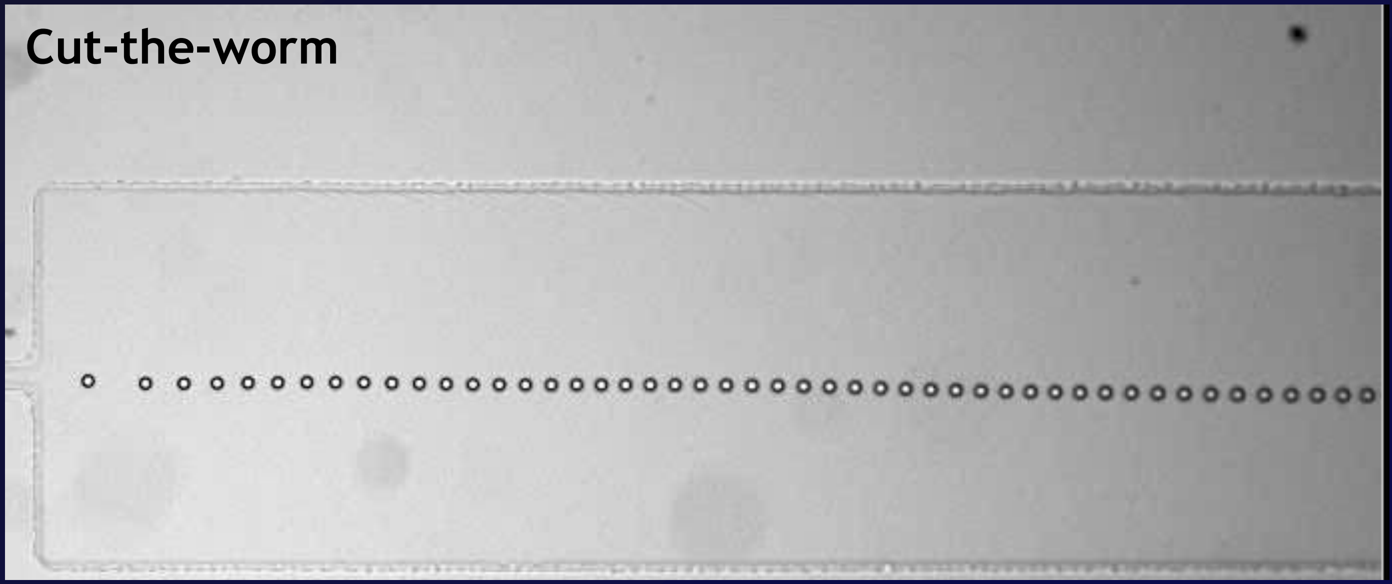


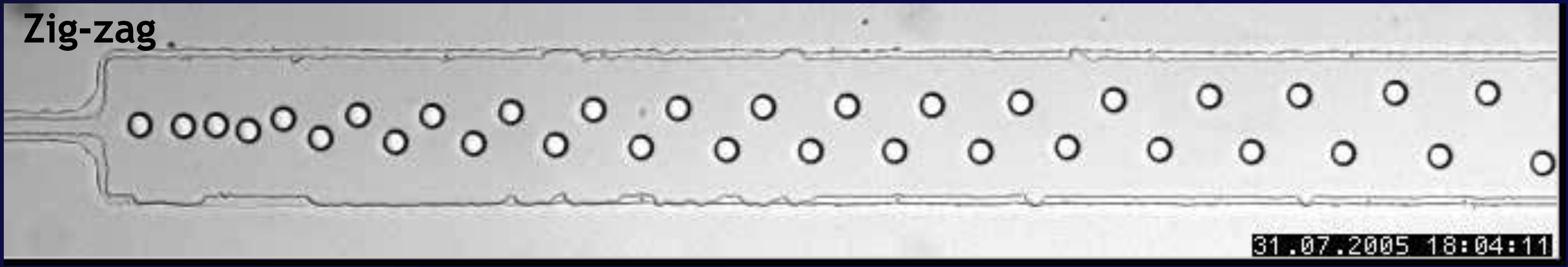
(No adjustable parameters)

Nat. Phys. **2**, *743*, (2006)

Non-linear instabilities



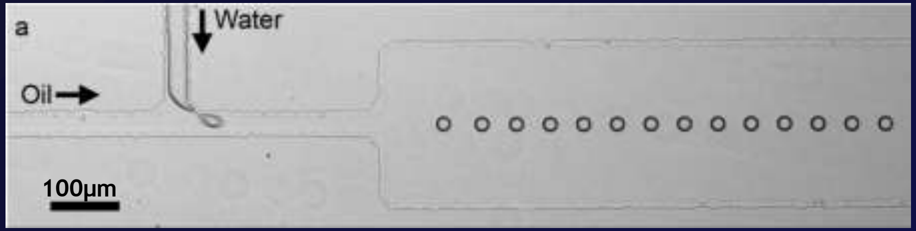




Geometry affects long-range dipoles: Confined crystal - from 2D to 1D

Confinement parameter $\gamma \equiv \frac{2R}{W}$

$\gamma = 0.1$



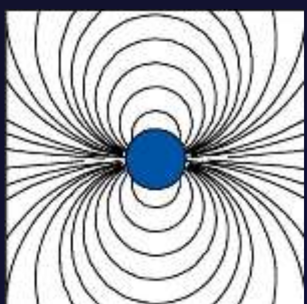
$\gamma = 0.4$

$\gamma = 0.63$

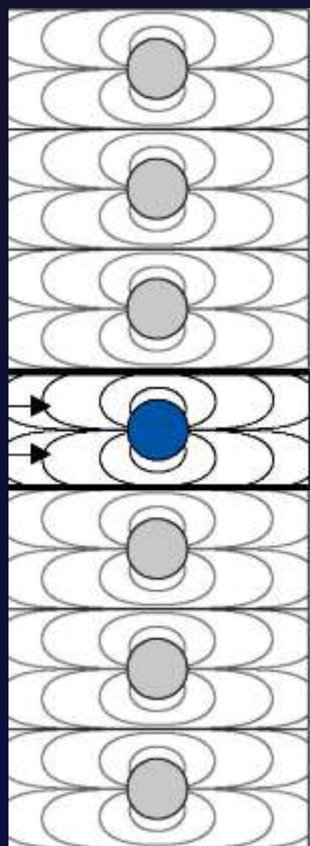
$\gamma = 0.8$

What is the effect on phonons?

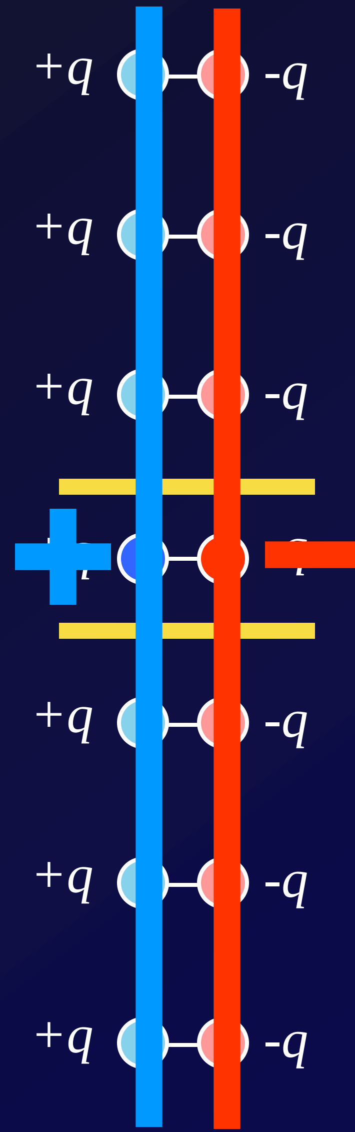
Confinement induces screening



unconfined droplet



confined droplets carries an array of mirror droplets



a dipole array as a plate capacitor

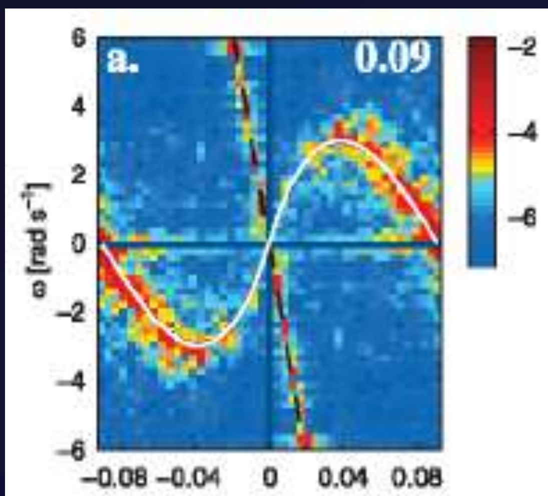
$$F \sim \partial_x \phi \sim e^{-2\pi x/W}$$

potential is screened

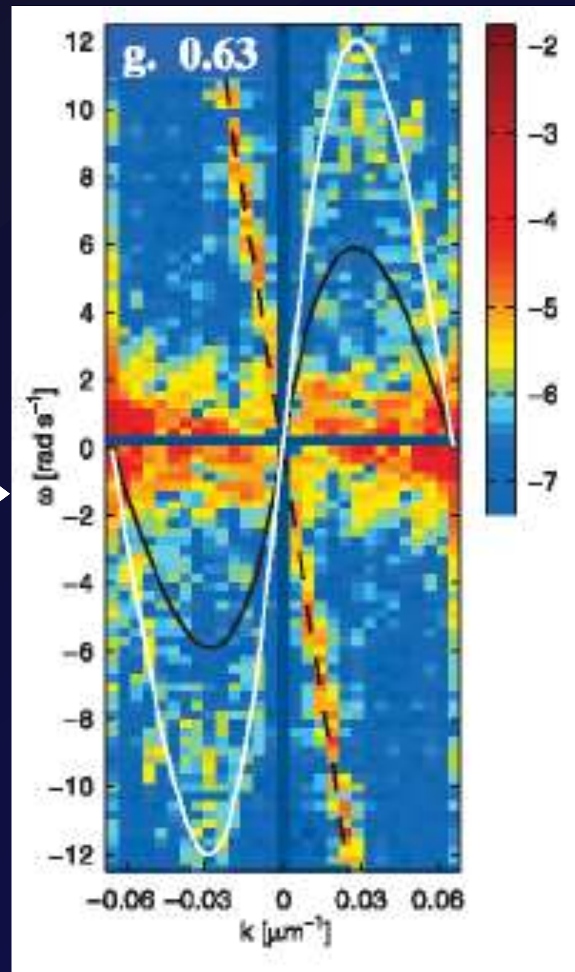
expect lower $\omega(k)$ and C_s

Movie

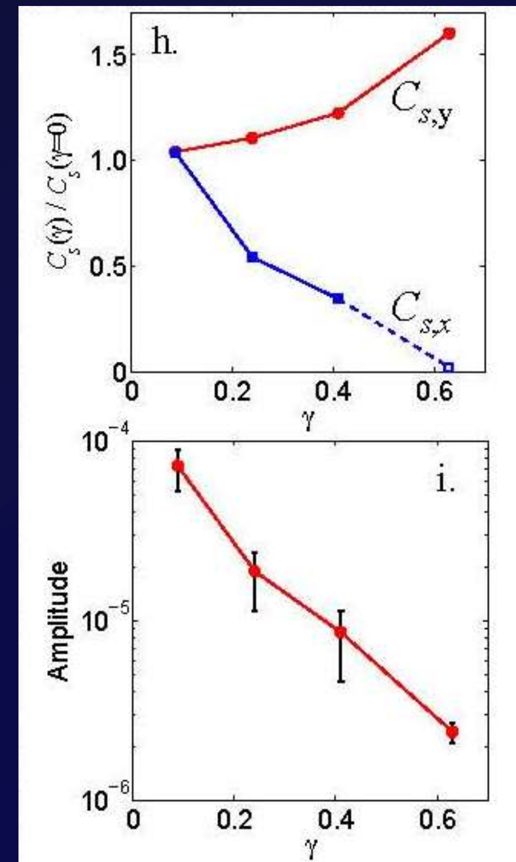
Anomaly: Sometimes phonons move faster



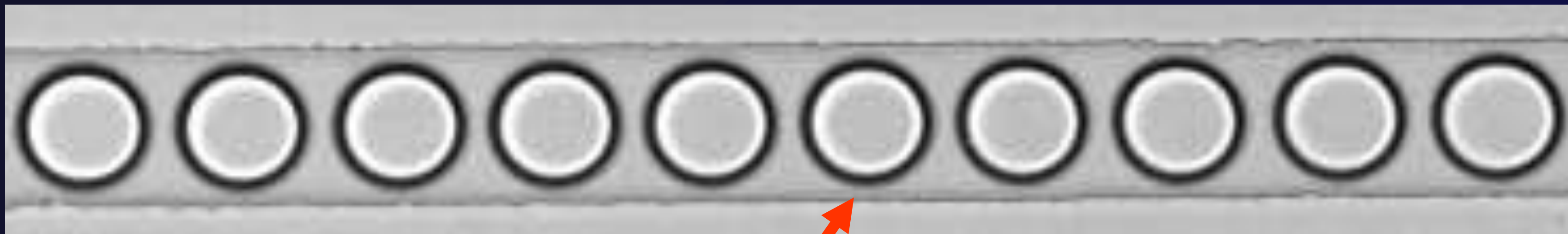
Unconfined transversal



confined



Screening against incompressibility

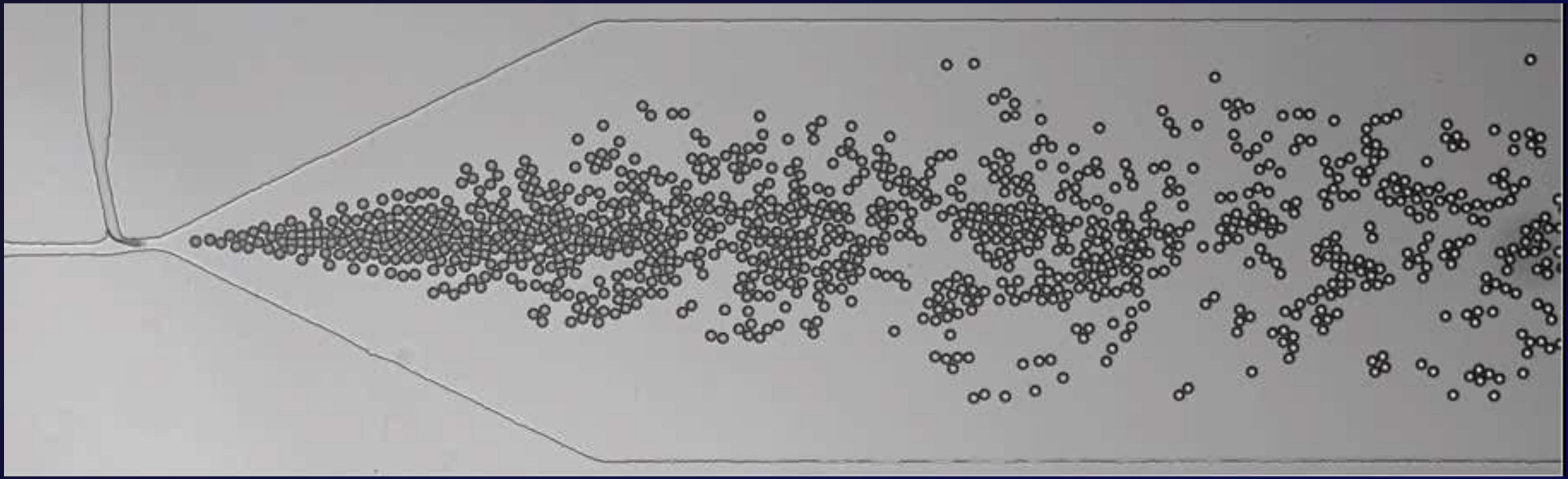


- 1D limit → small gaps → high resistance
- Crystal becomes ‘incompressible’
- Expect $C_s \to \infty$

$$F \sim \partial_x \phi \sim \gamma \underbrace{\tan\left(\pi\gamma / 2\right)}_{\text{incompressibility}} \underbrace{\exp\left(-2\pi x / W\right)}_{\text{screening}}$$

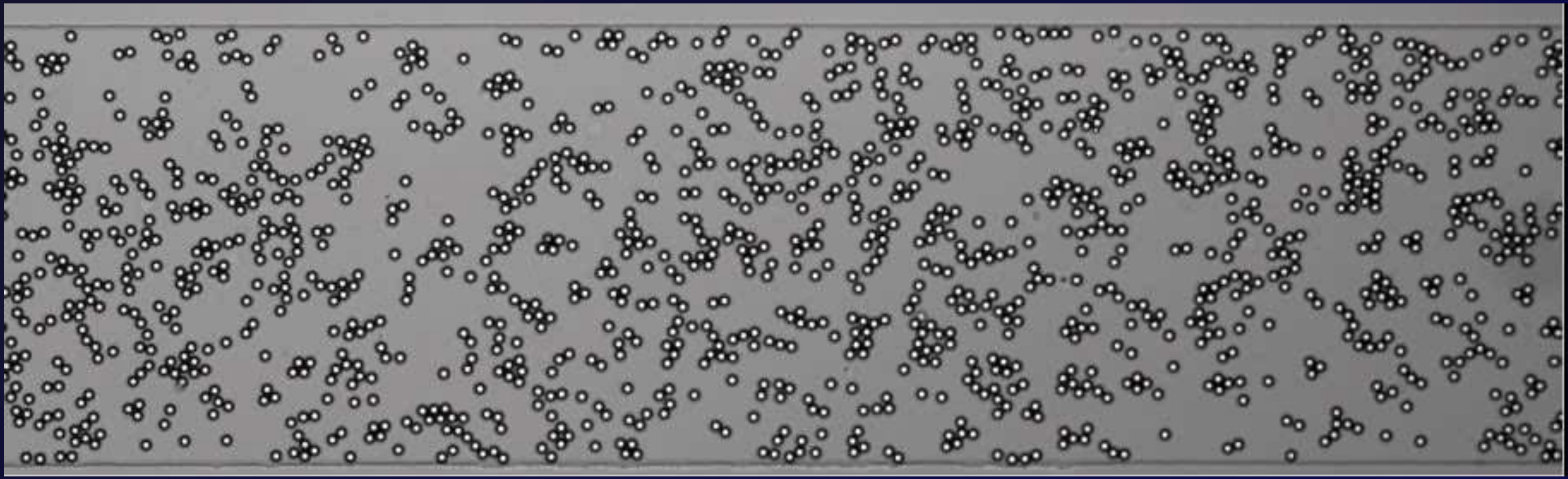
Phys. Rev. Lett., **99**, 124502 (2007)

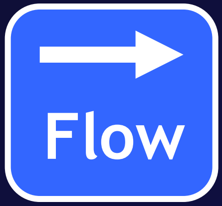
Go from 1D-order to 2D-Disorder



Flow

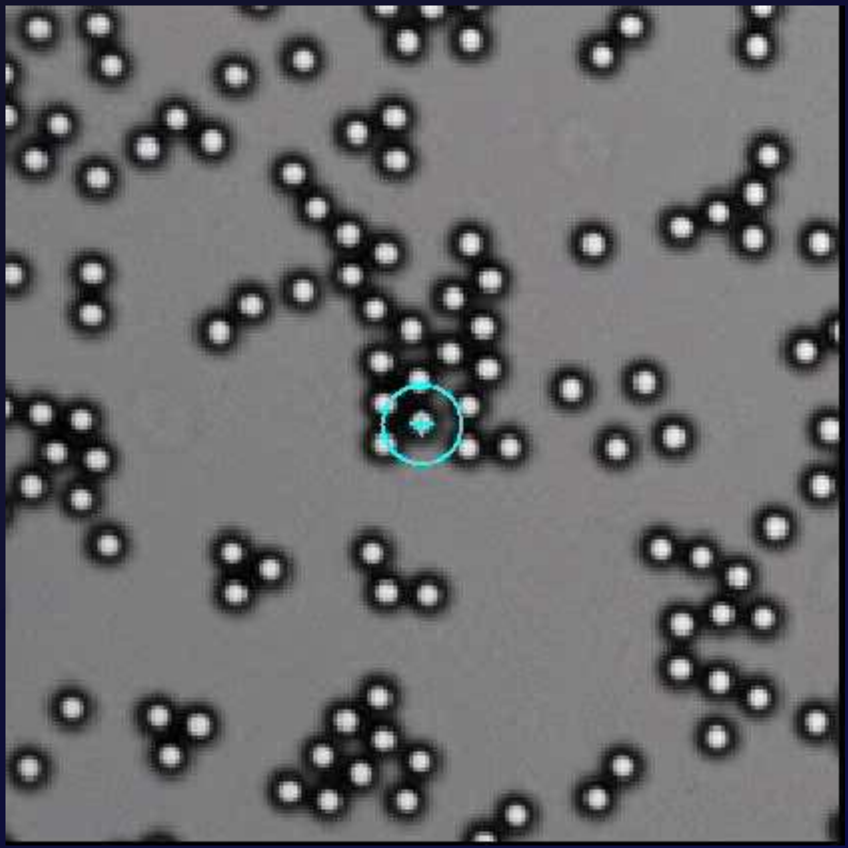
Move along with the droplets

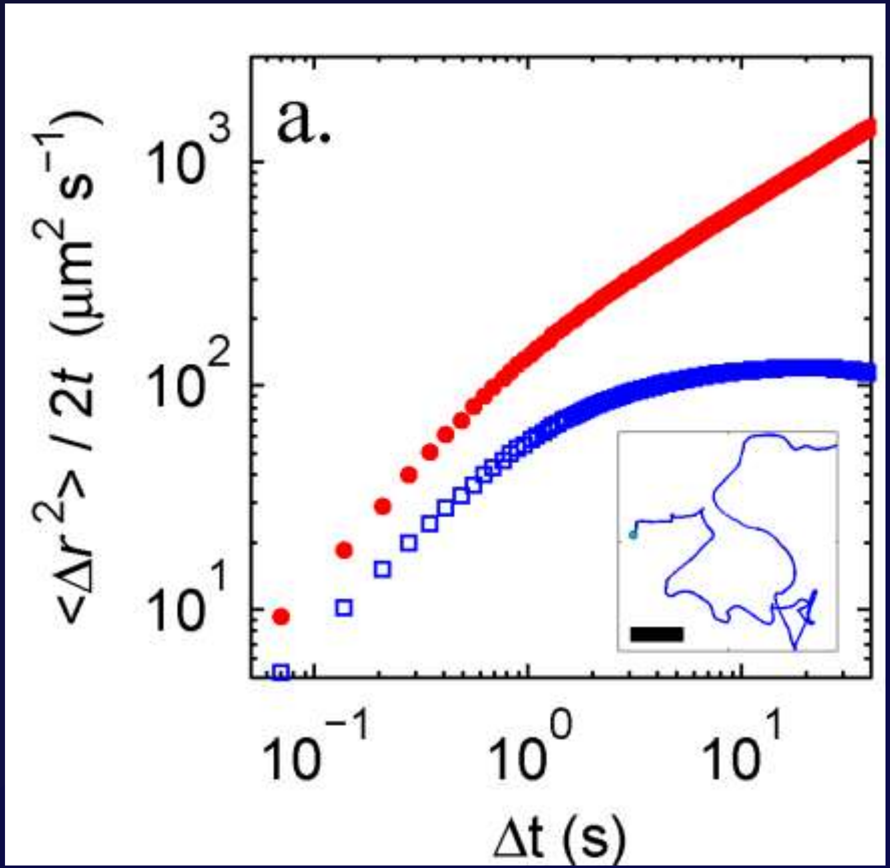




Complex collective motion

Random motion is complex: diffusion and clustering

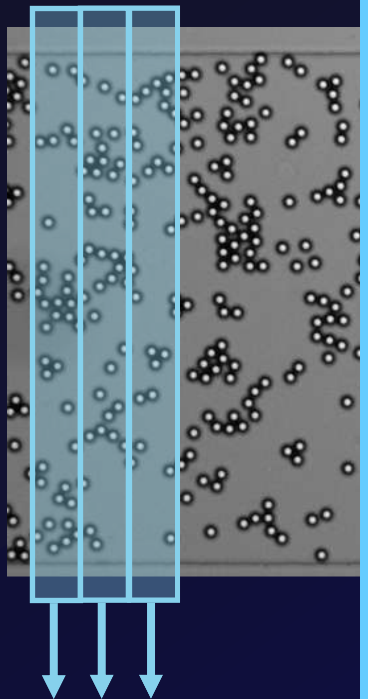




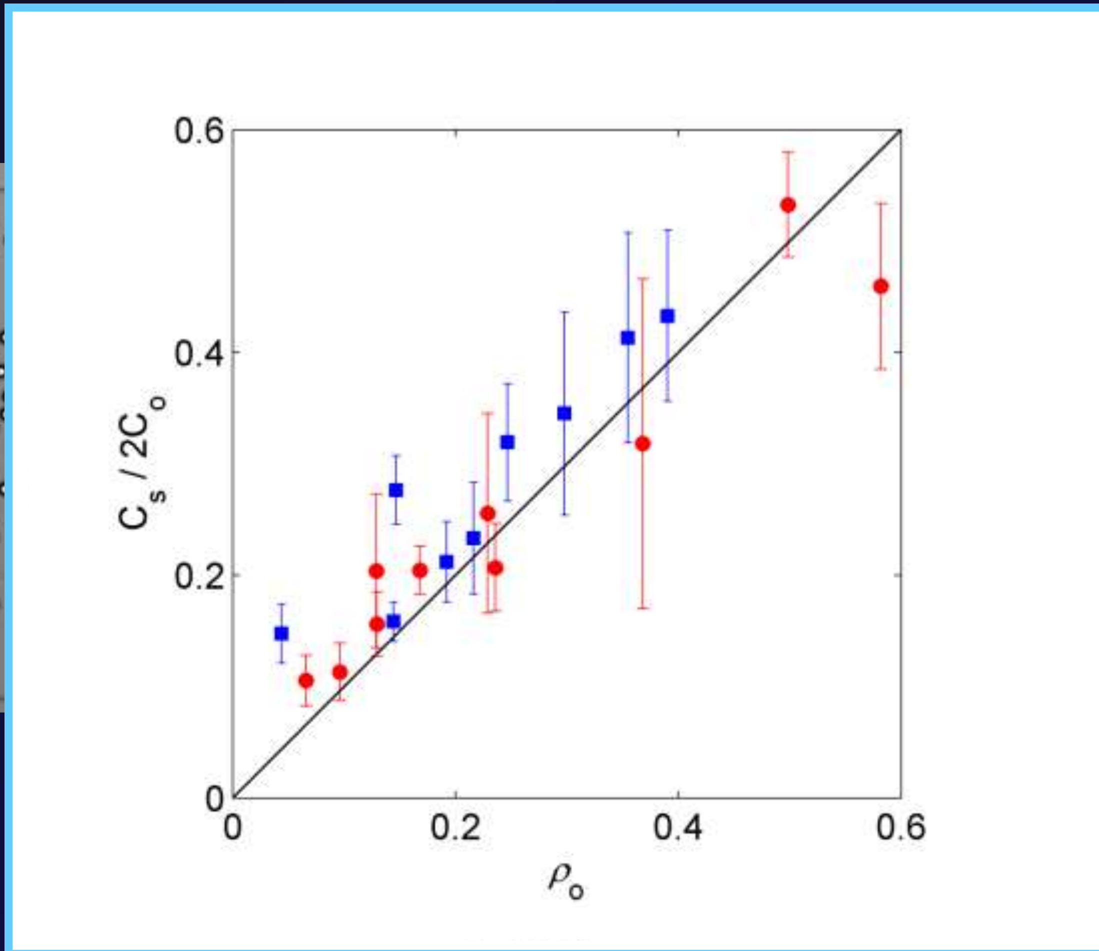
$$\Delta y \sim \sqrt{t}$$

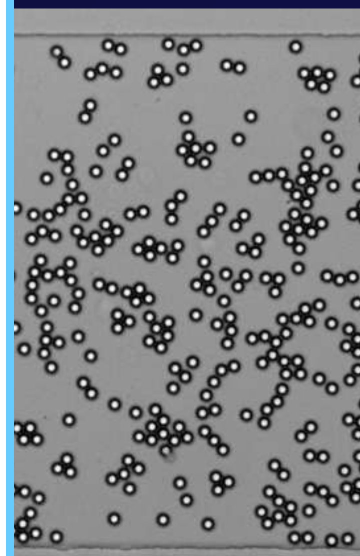
$$\Delta x \sim t^{0.7}$$

Density waves



$\rho(x,t)$

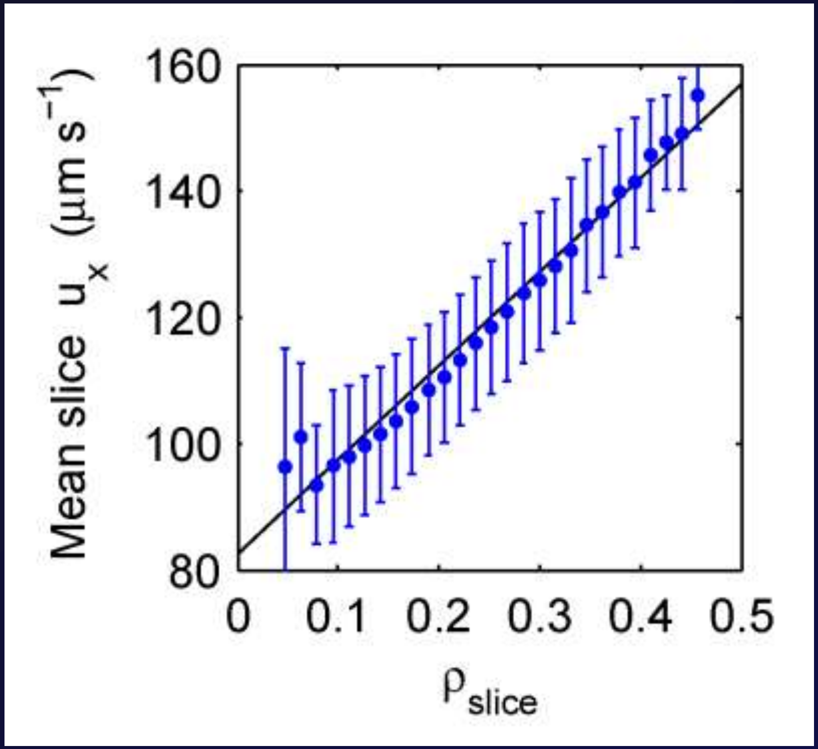




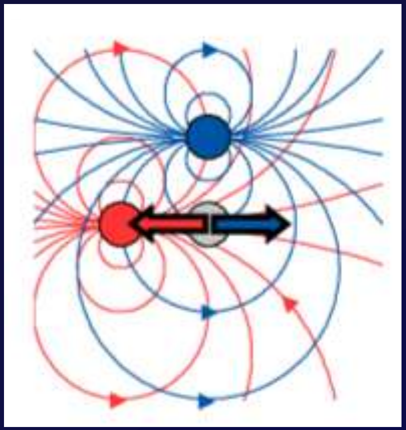
$$c_0 \equiv (u_d / u_{oil})(u_{oil} - u_d)$$

Density waves with velocity $c_s \sim 2\rho_0 c_0 \sim 10 - 100\,\mu m\, s^{-1}$

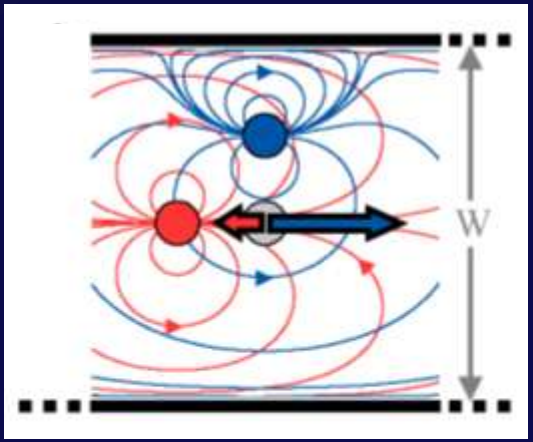
Local velocity-density coupling



$\Delta u = 0$



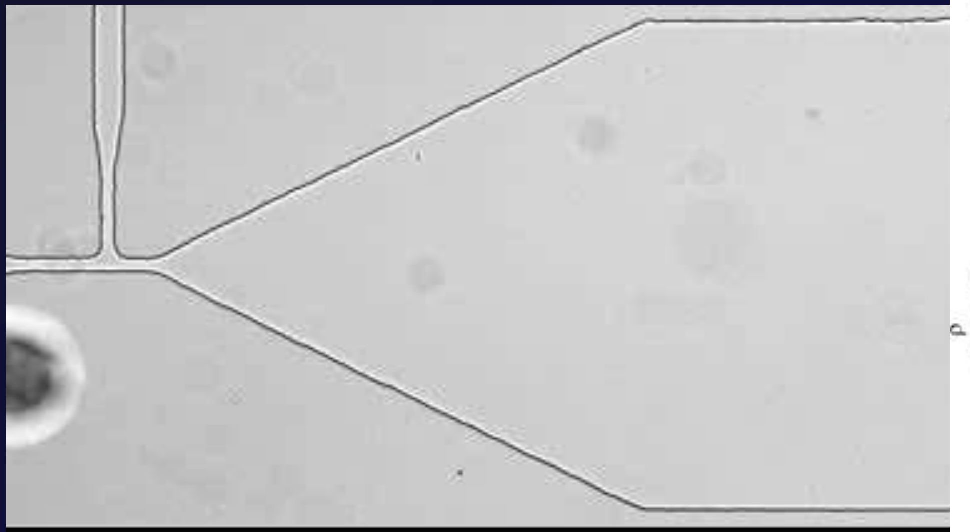
$\Delta u > 0$

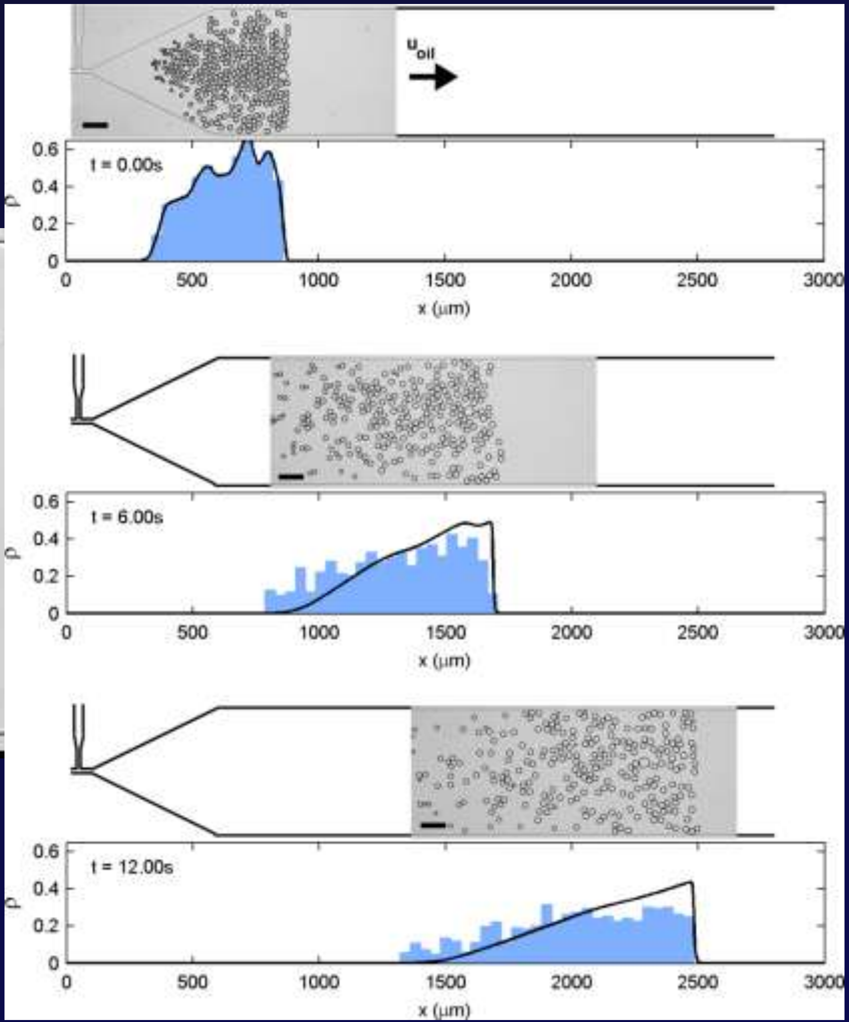


$$u(x,t) = u_o + c_o \cdot \rho(x,t)$$

Coupling stems from the sidewalls

Shockwave at $Re \sim 10^{-4}$





Coupling → Burgers Eq.

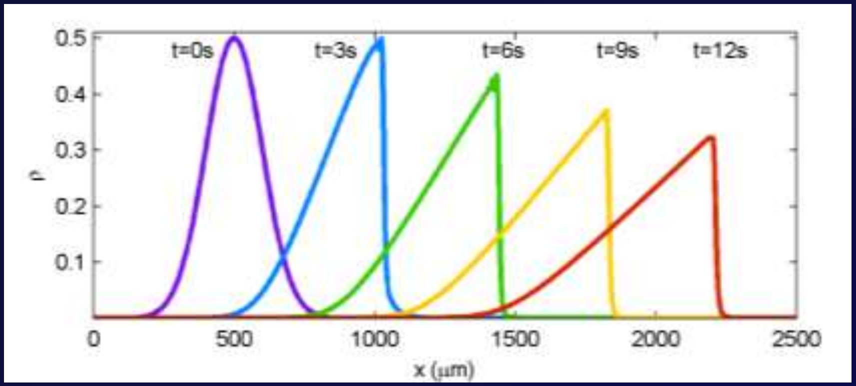
$$\begin{cases} u(x,t) = u_o + c_o \cdot \rho(x,t) \\ \dfrac{\partial \rho}{\partial t} + \dfrac{\partial}{\partial x}(u\rho) = D[\rho] \end{cases}$$

Coupling

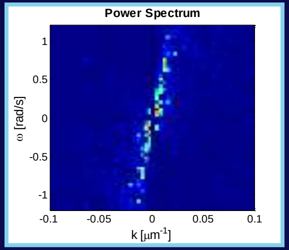
Mass conservation

$$\boxed{\frac{\partial \rho}{\partial t} + \rho \frac{\partial \rho}{\partial x'} = D[\rho]}$$

1D Burgers eq.

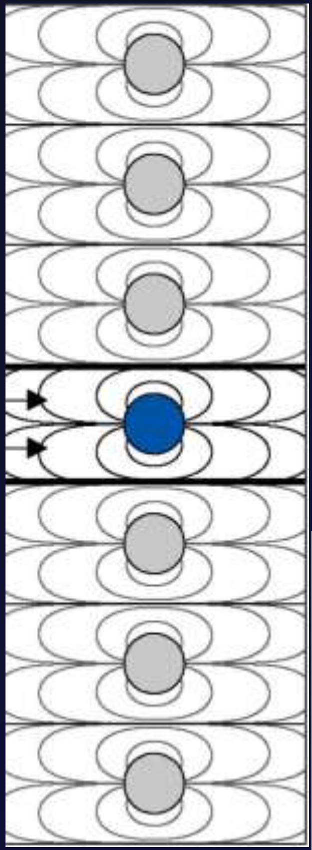


$\delta\rho \ll \rho_o \rightarrow$ Sound



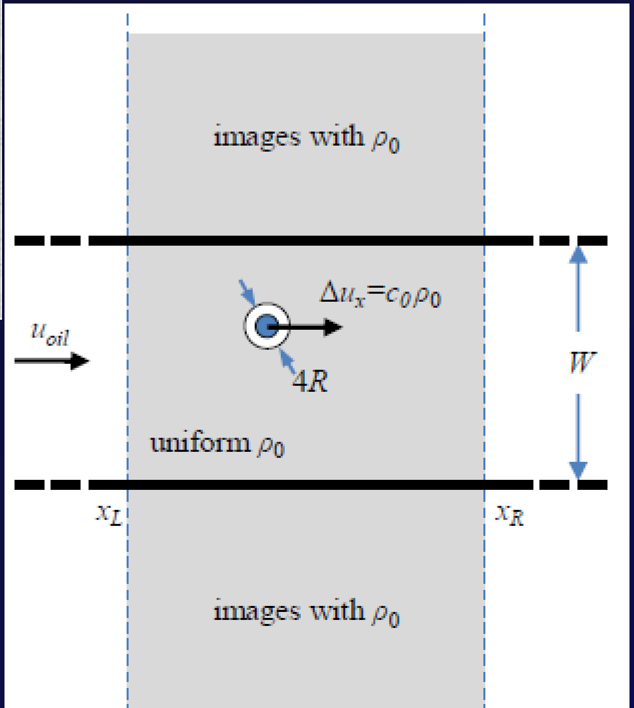
$\delta\rho \sim \rho_o \rightarrow$ Shockwaves !

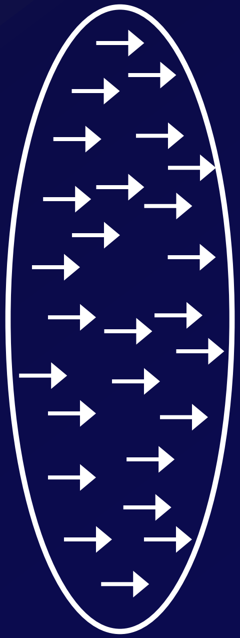
Shocks arise from symmetry breaking by walls



- Every droplet is reflected infinite times.
- Long mode is uniform in *y*.
  → Broken symmetry by boundaries.

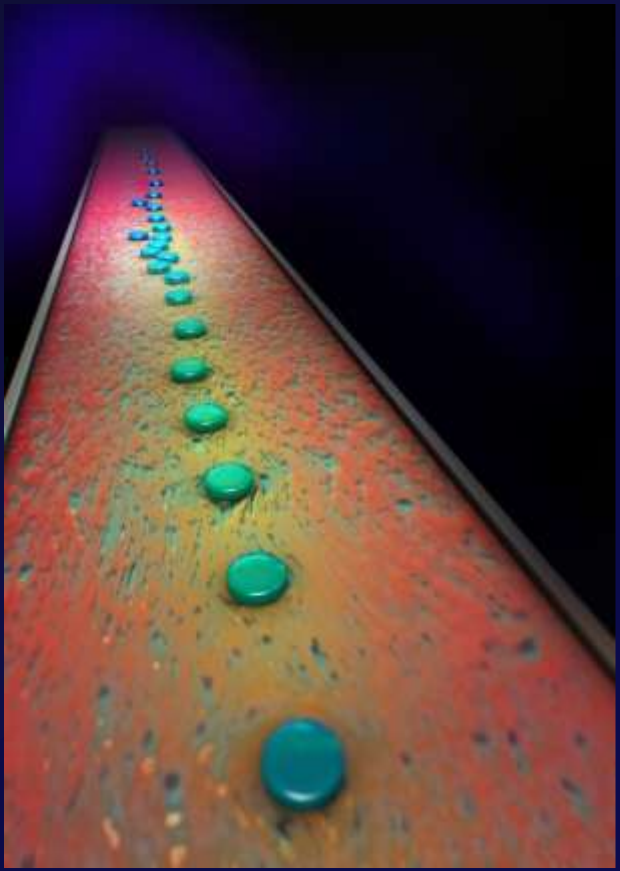
- Does not depend on width *W*.
- "Marginal divergence" of 2D dipoles in 2D:
- Shape effect (like electrostatic dipoles) not size.





Summary

- Many-body system
- Driven far from Equilibrium
- Highly dissipative (Re<<1)
- Broken symmetry
- Long-range interactions ~ r^{-2}
- Strong boundary effects
- Collective modes
  - Phonons in 1D (order)
  - Non-linear instabilities
  - Sound & shockwave in 2D (disorder)
- Simple theory: dipoles + boundaries
  - Crystal dispersion and dynamics
  - Burgers Equation
- Outlook – a model system for Non-Eq?



Nat. Phys. **2**, *743* (2006)

PRL, **99**, 124502 (2007)

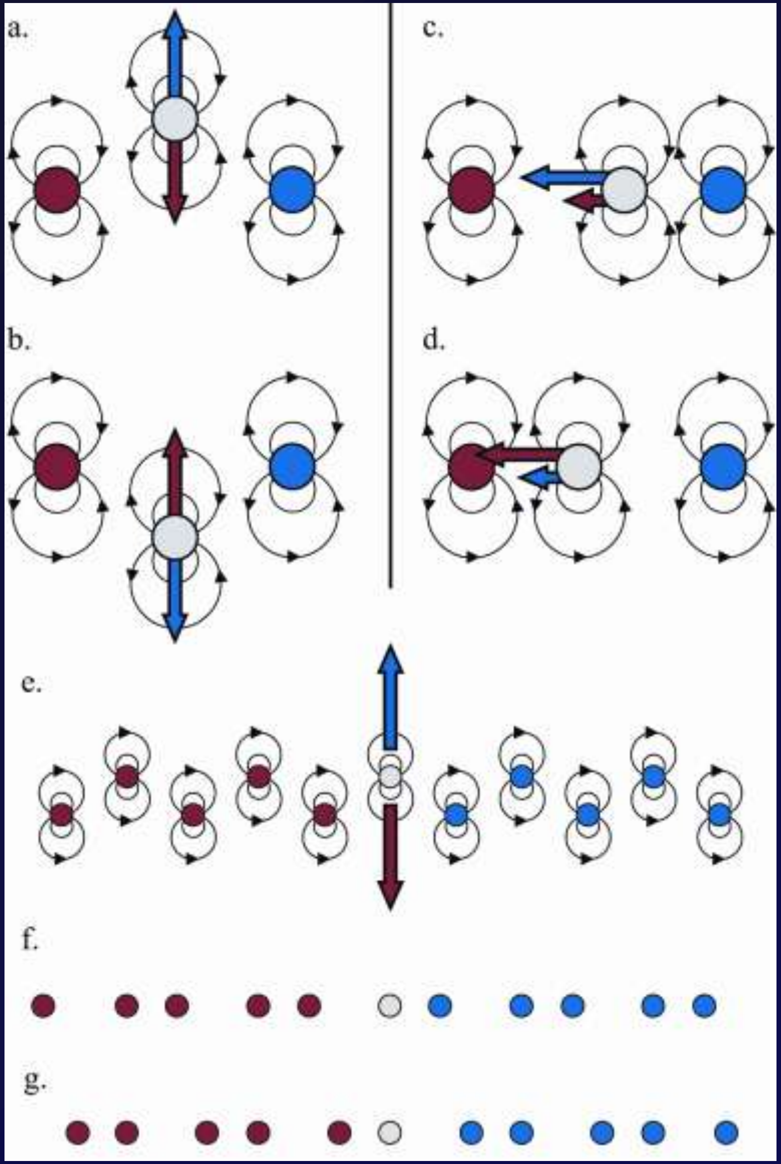
PRL, **103**, 114502 (2009)

Phys. Rep. (2012)

General Context

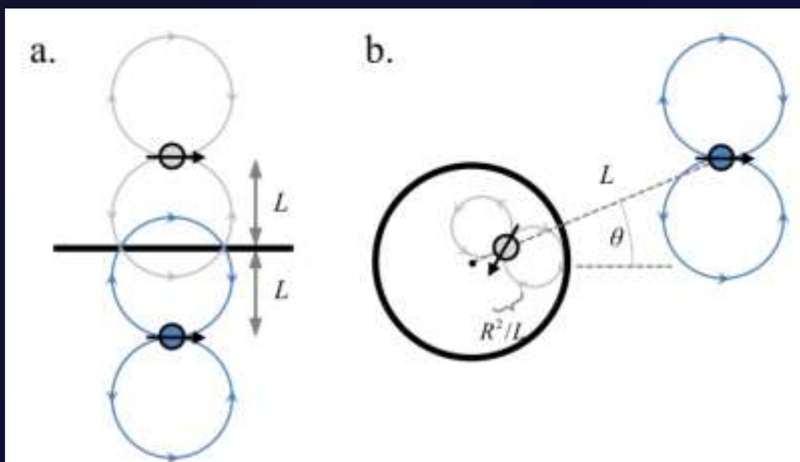
- Non-Equilibrium systems with:
  - Many bodies
  - Long range interactions
  - Dissipation and external drive (broken symmetry)
- Experimental examples:
  - Electrons conducted in a solid (1/r in 3D)
  - Dusty plasma (screened 1/r in 3D)
  - Sedimentation (1/r in 3D)
  - Microfluidic droplets ($1/r^2$ in quasi-2D)
- Complex collective dynamics, <u>no general theory</u>

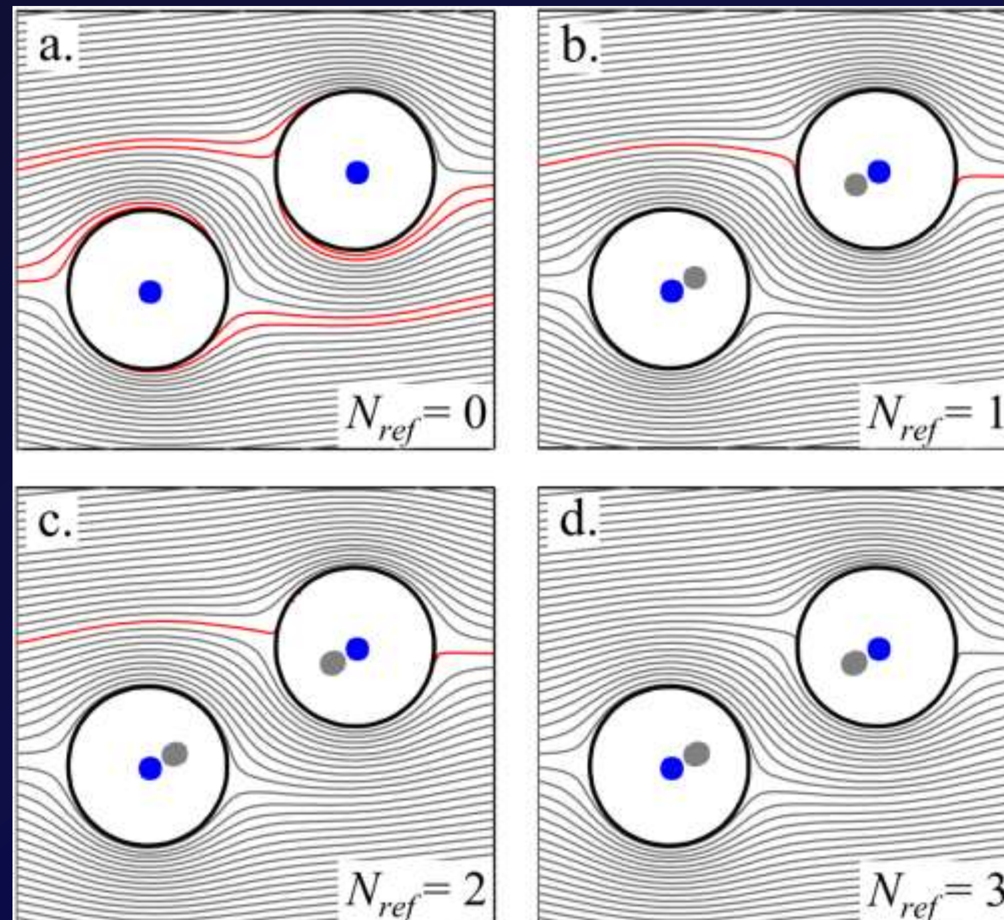
Zig-zag zero mode



Beyond the superposition approximation

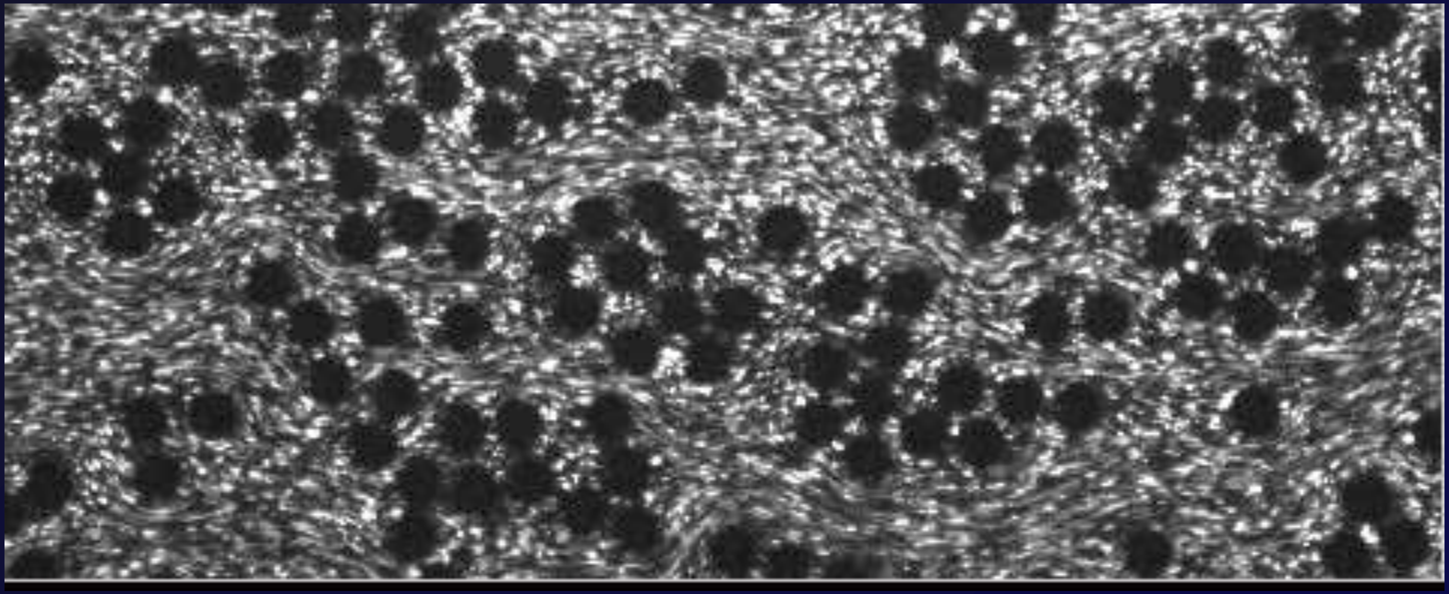
Use image dipoles to account for boundaries





- For more than 2 boundaries – hard enumeration of images
- Use lowest order (no reflections): accurate to ~4%

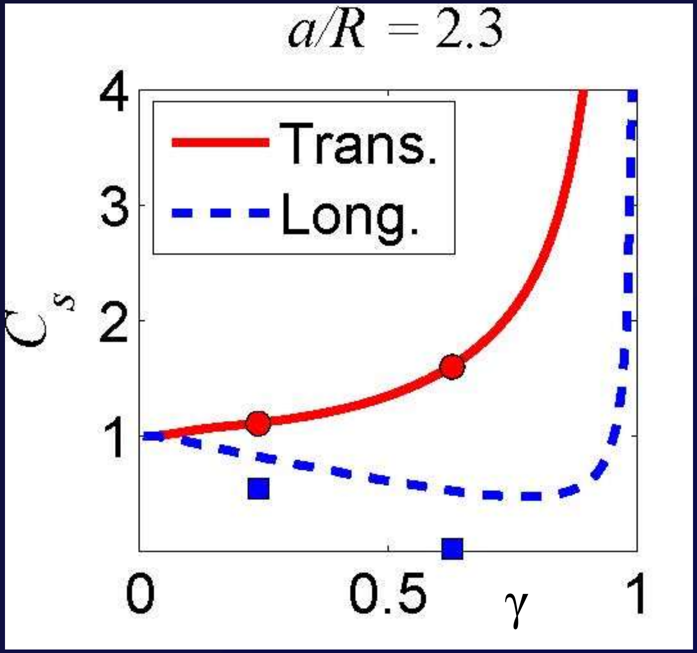
Outlook 3 - Measure Dissipation

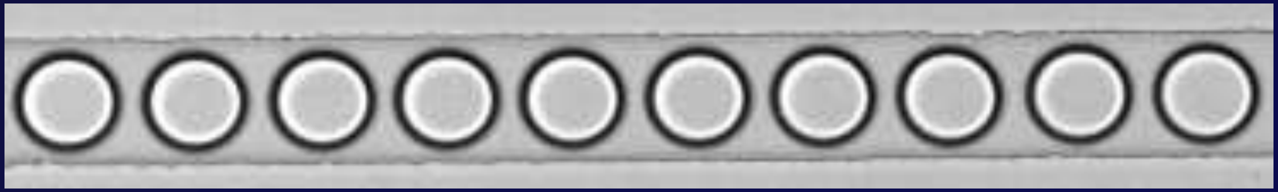


- Dissipation of oil and inside droplets
- Does the system arranges to minimize dissipation or related function?
- Can perturb and measure if dissipation decreases
- Dissipation in shockwaves…

Screening against incompressibility

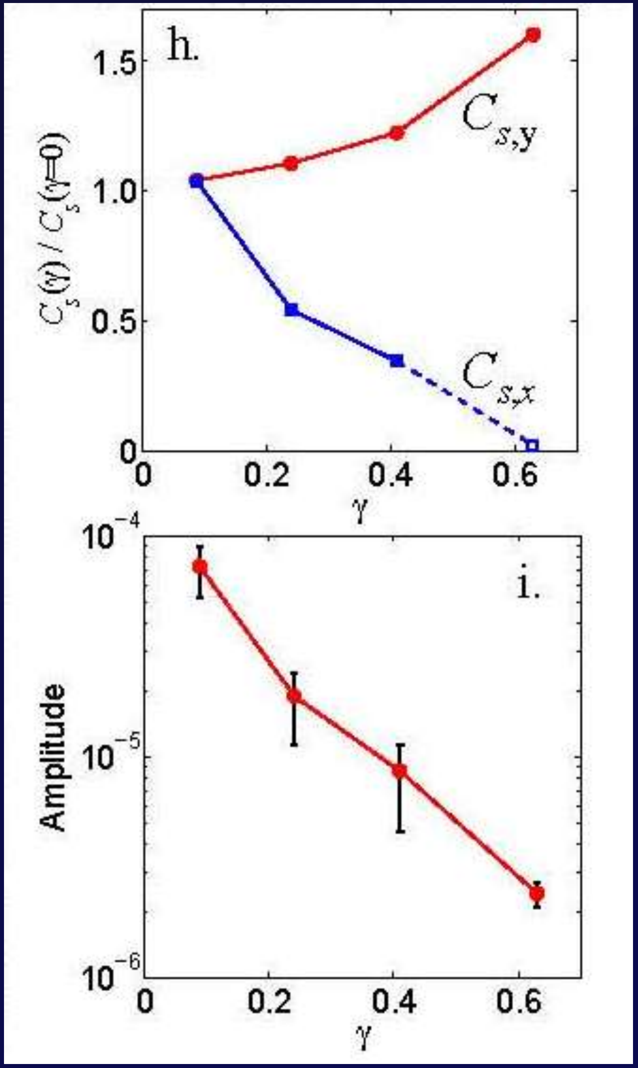
$$\partial_x \phi \sim \left(u_{oil}^{\infty} - u_d^{\infty}\right)\begin{cases}\gamma \tan\left(\pi\gamma/2\right)\exp\left(-2\pi x/W\right), & x \gg W \\ R^2/x^2 & , R < x \ll W\end{cases}$$





Breaking of xy anti-symmetry

- Breaking of x-y anti-symmetry
- in x : $\omega_y(k)$ and $C_{s,x}$ decrease
- in y : $\omega_x(k)$ and $C_{s,y}$ increase
- "Sub-sonic transition": $C_{s,y} > u_d$
- Phonon amplitude decays



The reason – an interplay between

- Screening of interactions
- Incompressibility

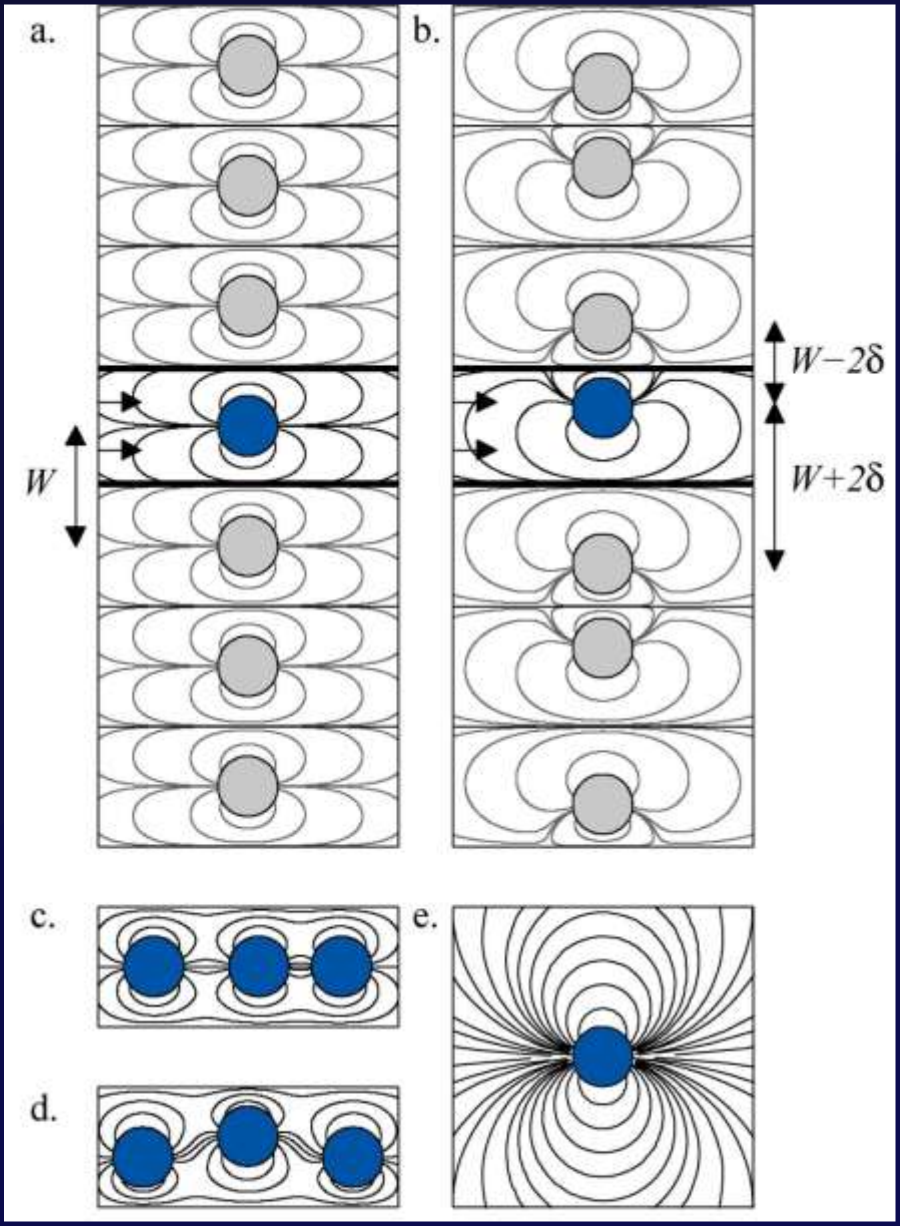
Confined flow potential

$$w(z) = \phi(z) + i\psi(z)$$

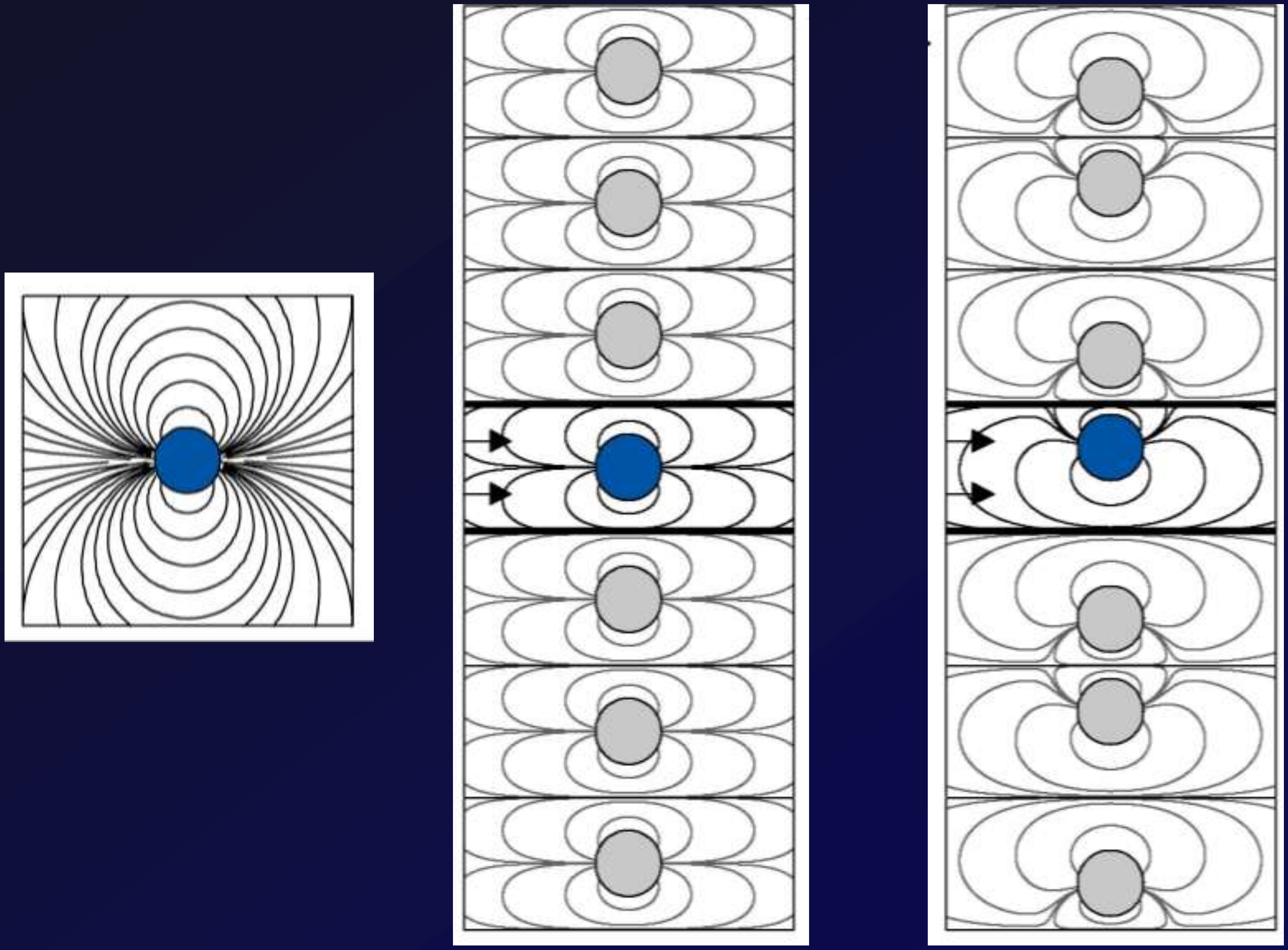
$$\phi \sim \coth(\pi z / 2W)$$

$$F \sim \partial_x \phi \sim e^{-2\pi x / W}$$

- Interactions are screened
- Expect lower $\omega(k)$ and C_s



Confined flow potential by mirror dipoles



General Context

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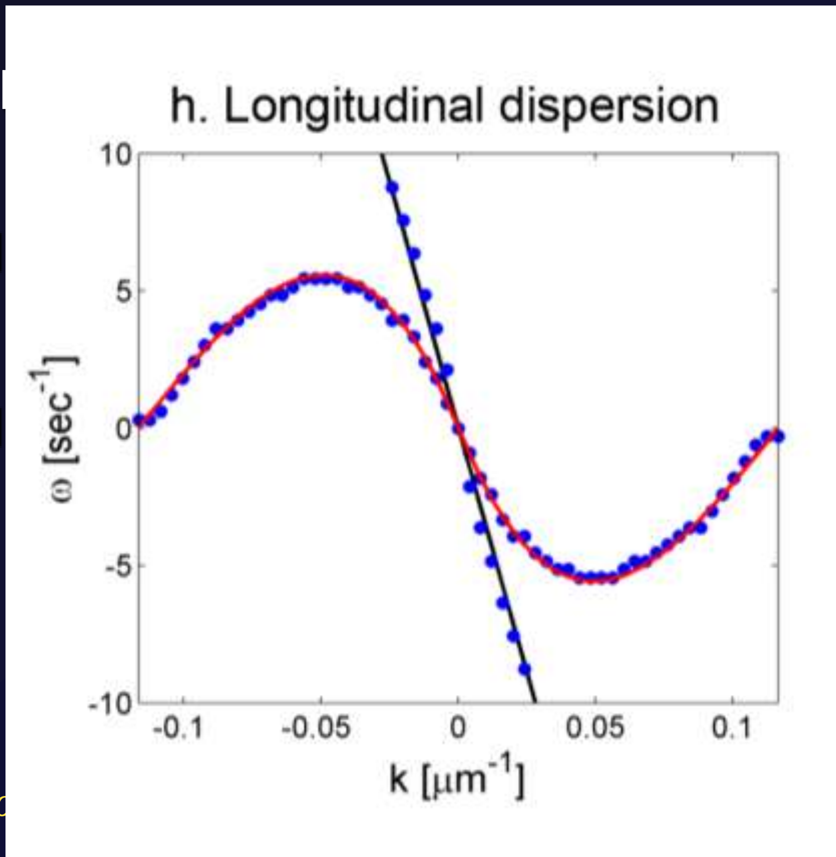
  - Microfluidic droplets ($1/r^2$ in quasi-2D)
- Complex collective dynamics, no general theory

Derive wave equations and $\omega(k)$

Su



tice + small vibrations

Eq $= \xi\left(\mathbf{u}_{oil} - \mathbf{u}_d\right) \qquad \mathbf{F}_{friction} = \mu \mathbf{u}_d$

Eq

$<< a$

$\dot{\mathbf{r}}_n$

$\mathbf{u}_d$ $)$

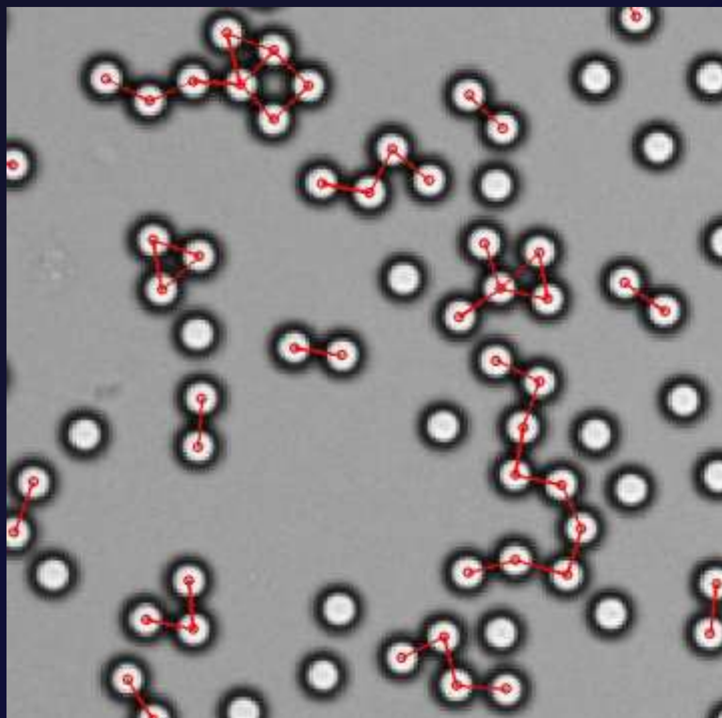
$$\omega_x(k) = -\frac{6C_s}{\pi^2 a}\sum_{j=1}^{\infty}\frac{\sin(jka)}{j^3}$$

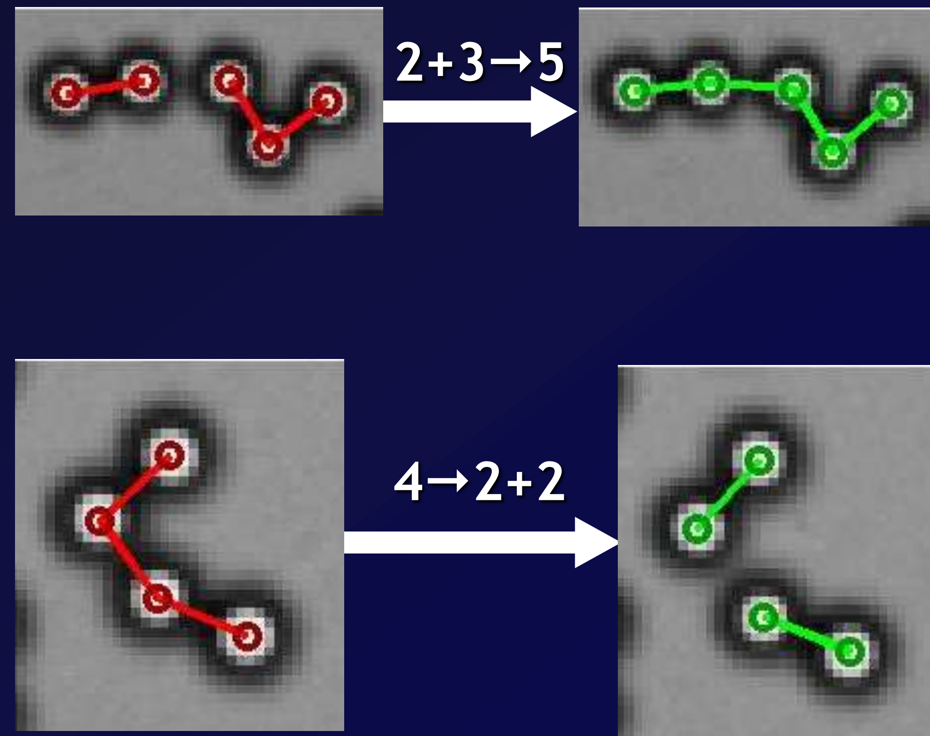
$$\omega_y(k) = -\omega_x(k)$$

Sound velocity

$$C_s = \left(\frac{2\pi^2}{3}\right)\left(\frac{R^2}{a^2}\right)\left(\frac{u_d^{\infty}}{u_{oil}^{\infty}}\right)\left(u_{oil}^{\infty} - u_d^{\infty}\right)$$

Outlook 1 - “Droplecules” dynamics





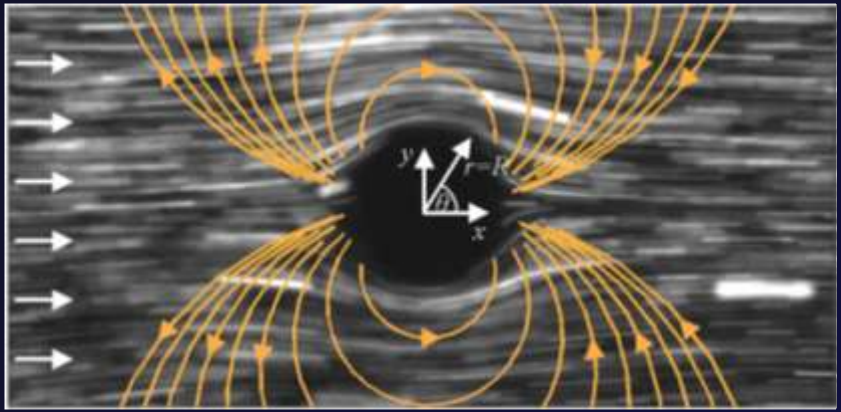
“Reactions” A+B ⇆ C
close to detailed-balance

Movie 1

Movie 2

Droplets are interacting dipoles

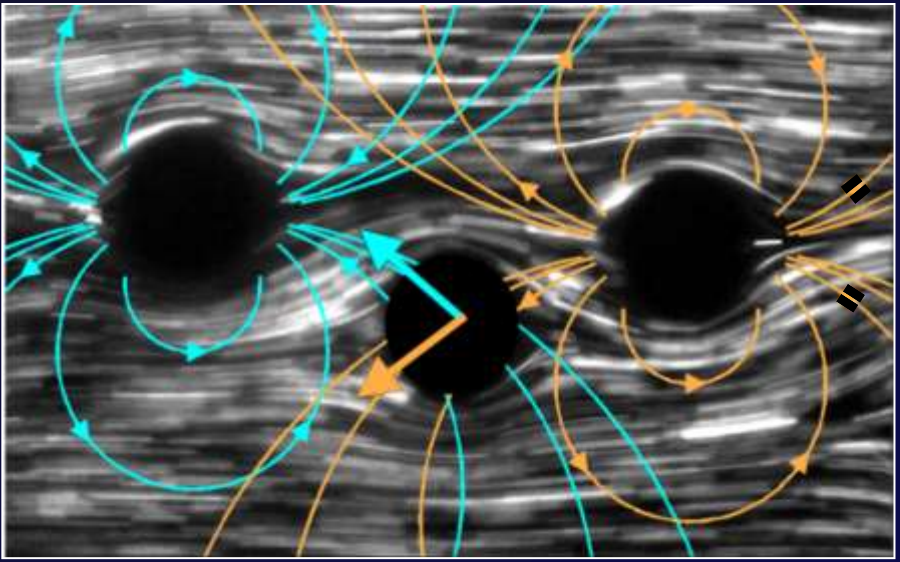
Dipole field



$$\eta\nabla^2\mathbf{v} = \nabla P$$ Navier-Stokes $Re<<1$

$$\mathbf{v}(\mathbf{r}) = \left(1-4z^2/h^2\right)\cdot\nabla\phi_d$$ Hele-Shaw

$$\begin{cases}\mathbf{v}(x,y) = \nabla\phi_d \\ \nabla\mathbf{v} = 0\end{cases} \Rightarrow \boxed{\nabla^2\phi_d = 0}$$ Laplace Eq



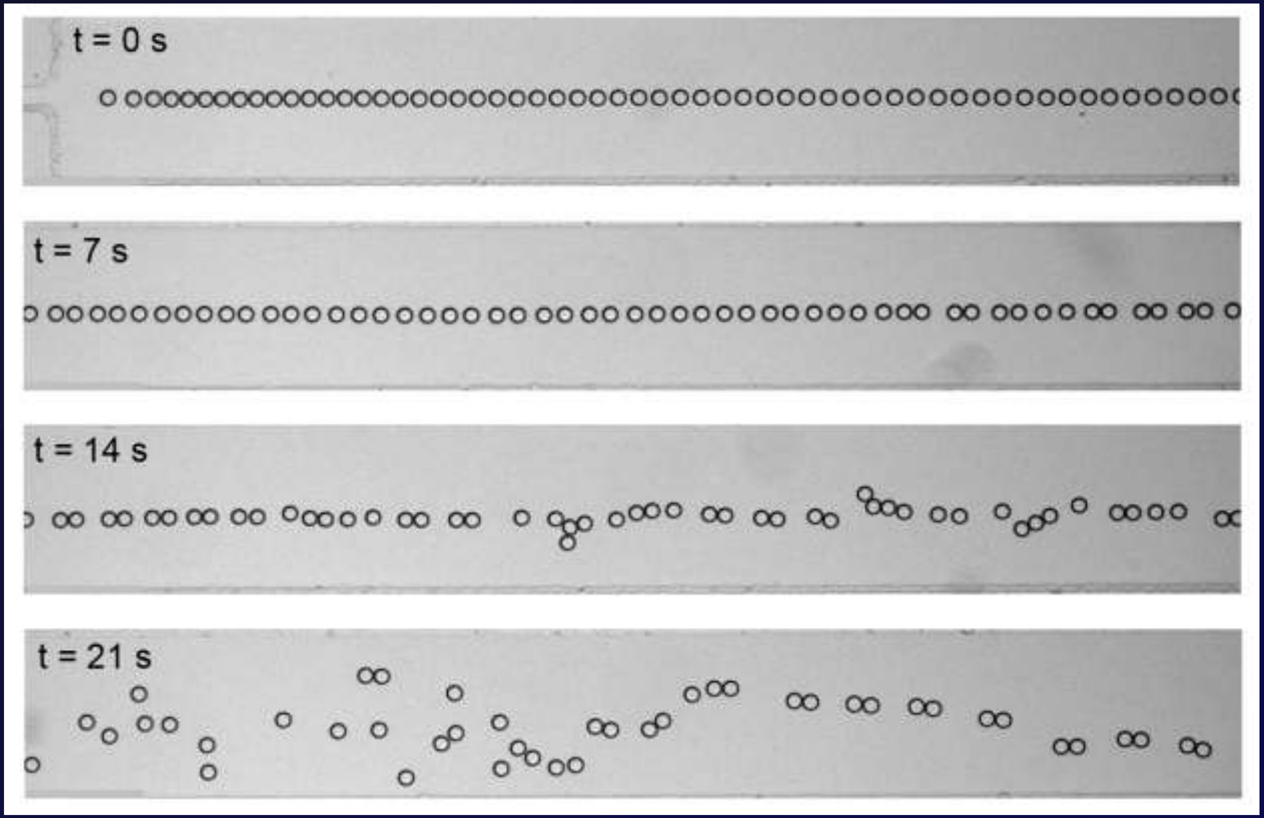
$$\phi(\mathbf{r}) = R^2\left(u_{oil}^{\infty} - u_d\right)\frac{\mathbf{r}\cdot\hat{x}}{r^2}$$ Dipole

Drag force: Long-range interaction

$$\boxed{\mathbf{F}_{drag} = \xi_d\,\nabla\phi\,(\mathbf{r}_i - \mathbf{r}_j) \sim r^{-2}}$$

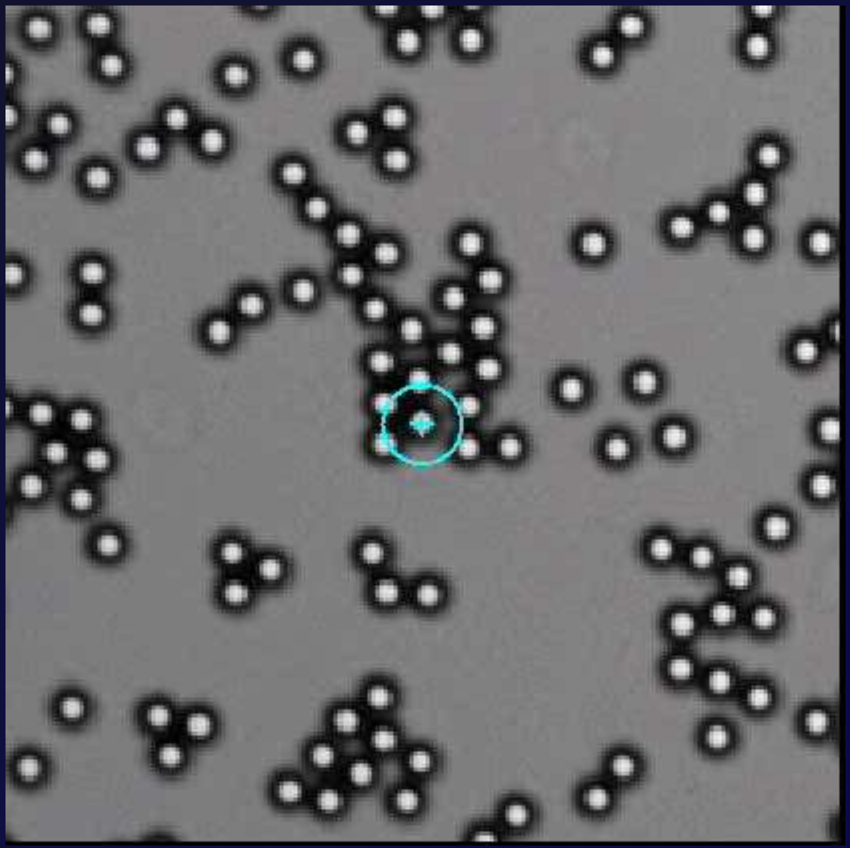
$$\xi_d = \frac{8\pi\eta R^2}{h}$$

Outlook 2 - Crystal Melting



- Characterize melting in terms of
  - Non-linear interactions (dynamics)
  - Crystal parameters (statistics)

Random trajectories

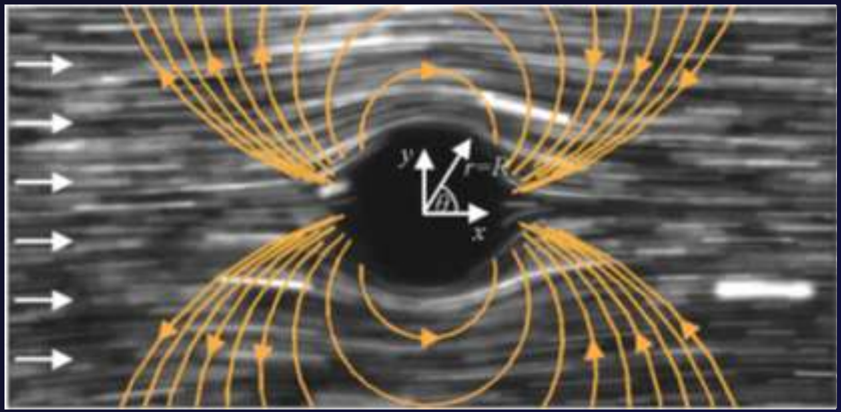


Outline

- Microfluidic droplet ensembles
- 1D crystal – phonons and instabilities
- Phonon spectra under confinement
- 2D Disordered ensemble – Burgers shockwaves

Droplets are interacting dipoles

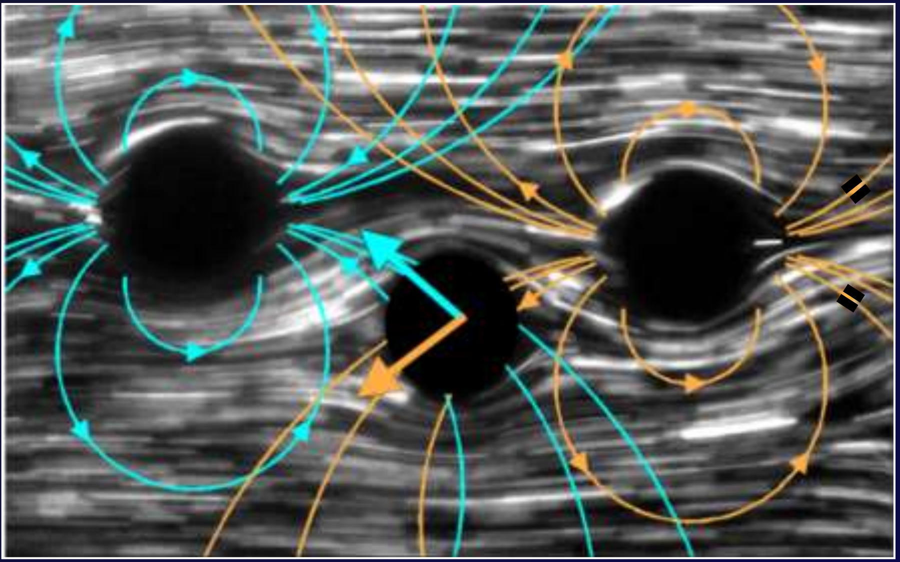
Dipole field



$$\eta \nabla^2 \mathbf{v} = \nabla P$$ Navier-Stokes $Re<<1$

$$\mathbf{v}(\mathbf{r}) = \left(1-4z^2/h^2\right)\cdot\left(\nabla\phi_d\right) = \left(1-4z^2/h^2\right)\cdot\left(-\frac{h^2}{8\eta}\nabla P\right) \qquad \phi_d \equiv -\frac{h^2}{8\eta}P$$

$$\begin{cases} \mathbf{v}(x,y) = \nabla\phi_d \\ \nabla \mathbf{v} = 0 \end{cases} \Rightarrow \boxed{\nabla^2\phi_d = 0}$$ Laplace Eq



$$\phi(\mathbf{r}) = R^2\left(u_{oil}^{\infty} - u_d\right)\frac{\mathbf{r}\cdot\hat{x}}{r^2}$$ Dipole

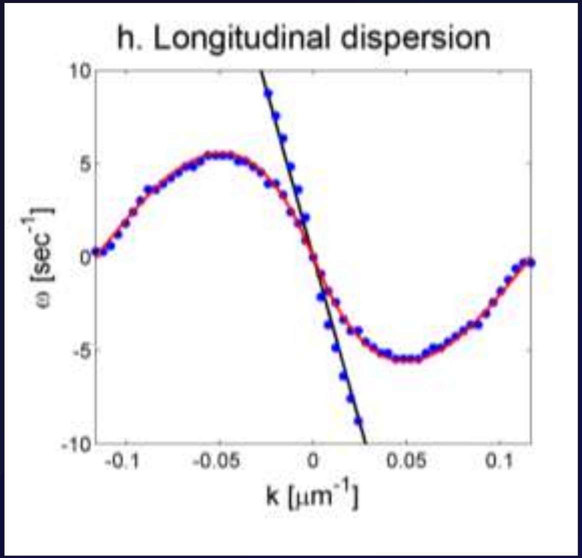
Drag force: Long-range interaction

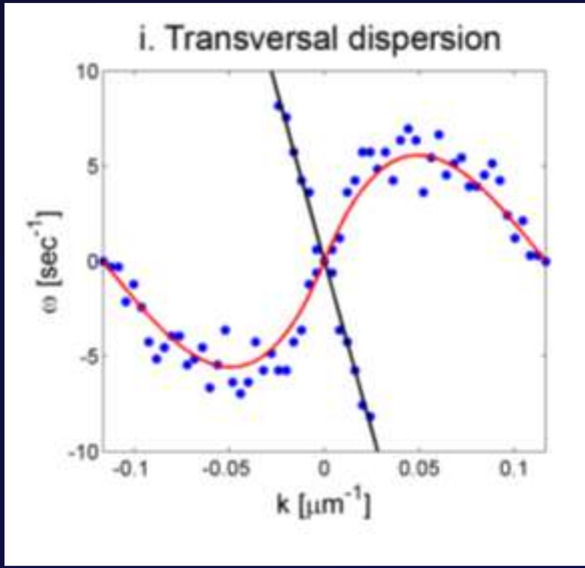
$$\boxed{\mathbf{F}_{drag} = \xi_d \nabla\phi(\mathbf{r}_i - \mathbf{r}_j) \sim r^{-2}}$$

$\Downarrow$

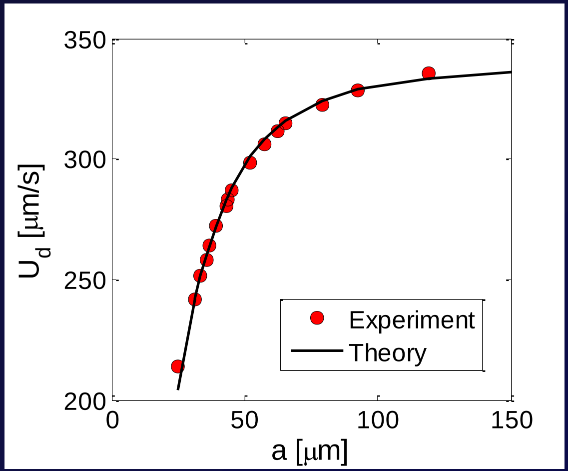
Collective modes !

Theory vs. Experiments

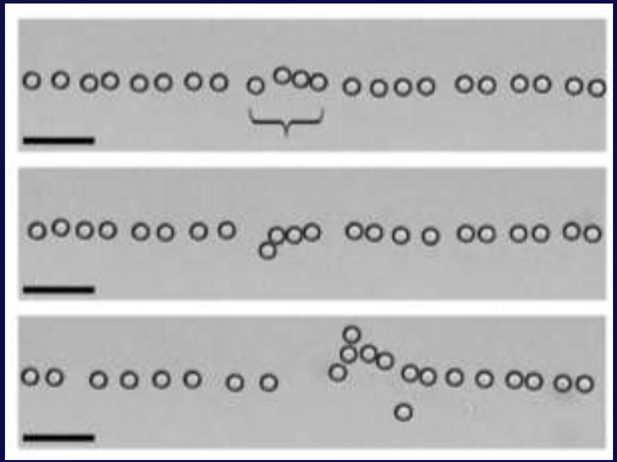




'Peloton' effect



Non-linear instabilities



Nature Physics, **2**, 743 (2006)

Derive wave equations and *ω(k)*

Superposition of dipole fields on a lattice + small vibrations

The only forces are: $\mathbf{F}_{drag} = \xi\left(\mathbf{u}_{oil} - \mathbf{u}_d\right)$ $\mathbf{F}_{friction} = \mu\mathbf{u}_d$

Equate forces, get equation of motion:

$$\dot{\mathbf{r}}_n = \left(\frac{1}{1+\mu/\xi}\right)\mathbf{u}_{oil}(\mathbf{r}_n)$$

$$\mathbf{u}_{oil}(\mathbf{r}_n) = u_{oil}^{\infty}\hat{\mathbf{x}} + \sum_{i\neq n}\nabla\phi(\mathbf{r}_i - \mathbf{r}_n)$$

Derive wave equations and $\omega(k)$

$$\dot{\mathbf{r}}_n = \left(1+\mu/\xi\right)^{-1}\mathbf{u}_{oil}(\mathbf{r}_n)$$

$$\mathbf{u}_{oil}(\mathbf{r}_n) = u_{oil}^{\infty}\hat{\mathbf{x}} + \sum_{i\neq n}\nabla\phi(\mathbf{r}_i-\mathbf{r}_n)$$

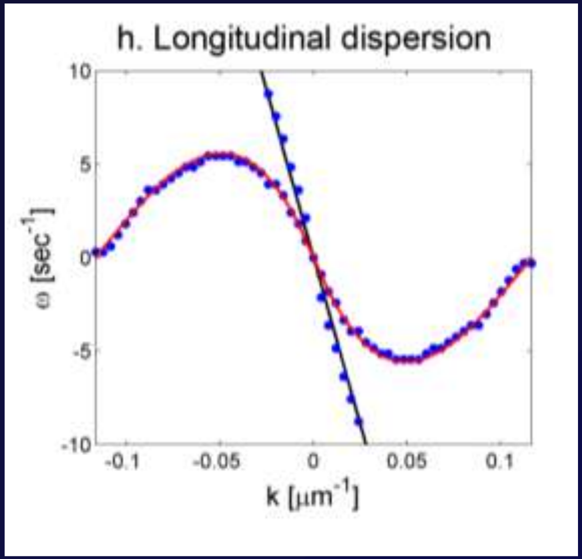
$x, y << a$

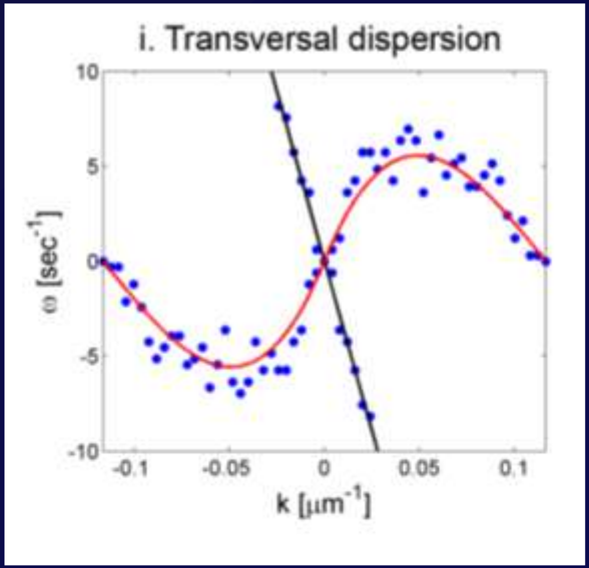
$$\omega_x(k) = -\frac{6C_s}{\pi^2 a}\sum_{j=1}^{\infty}\frac{\sin(jka)}{j^3}$$

$$\omega_y(k) = -\omega_x(k)$$

Sound velocity $$C_s = \left(\frac{2\pi^2}{3}\right)\left(\frac{R^2}{a^2}\right)\left(\frac{u_d^{\infty}}{u_{oil}^{\infty}}\right)\left(u_{oil}^{\infty}-u_d^{\infty}\right)$$

(No adjustable parameters)

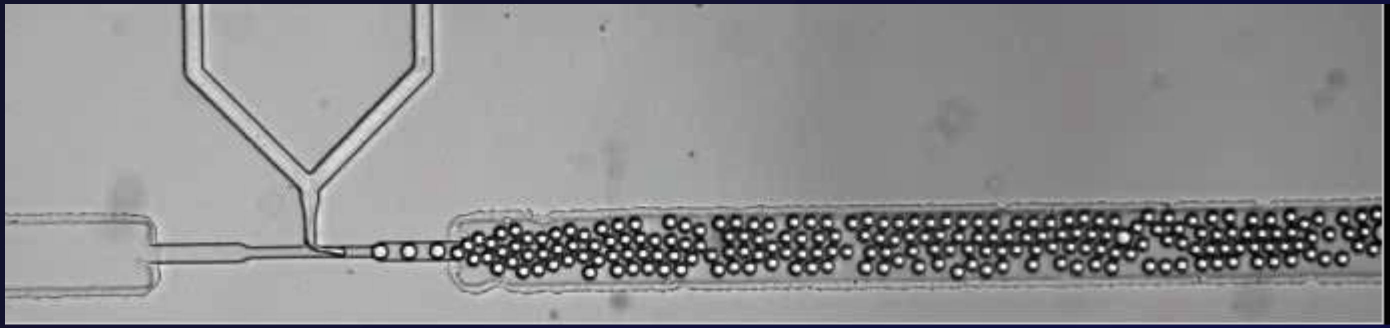


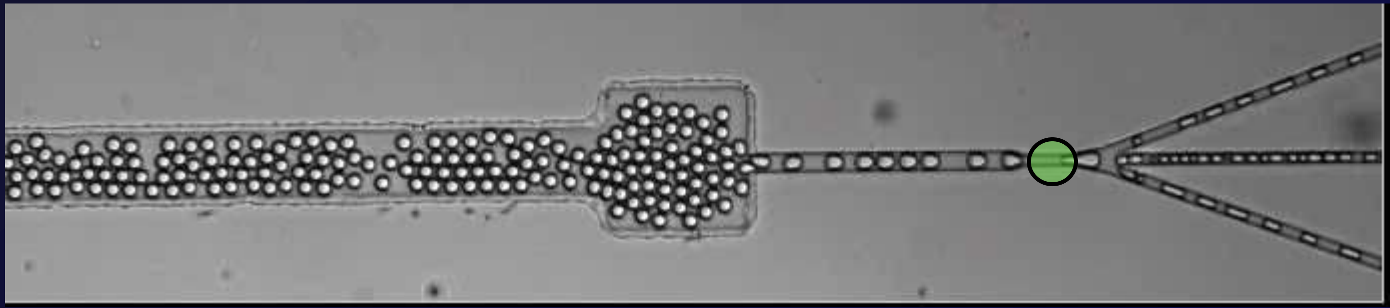


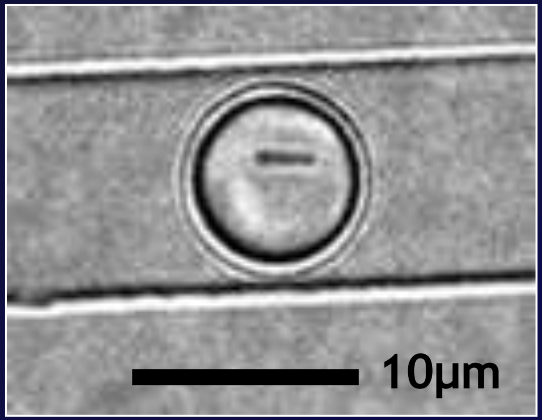
Full Eq.

Nature Physics, **2**, 743 (2006)

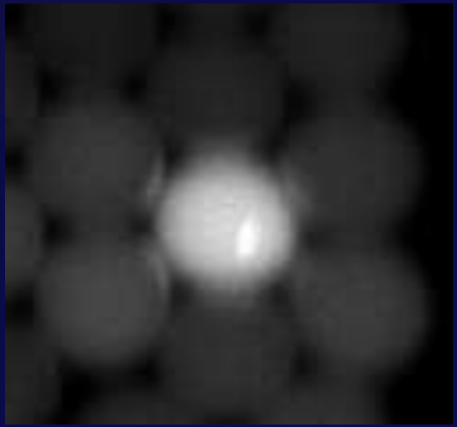
Droplets as reactors for gene expression







single E.Coli



T. Beatus