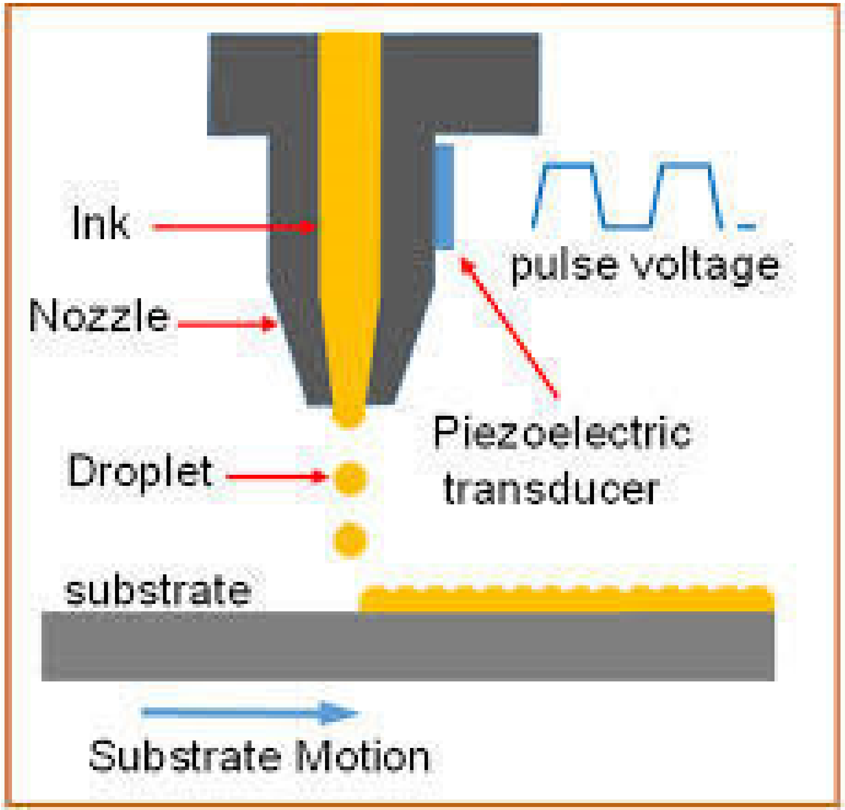
Mathematical problems in inkjet printing technology

Marco A. Fontelos
Instituto de Ciencias Matemáticas
Consejo Superior de Investigaciones Científicas
Madrid

Joint work with S. K. Hong, H. J. Kim, H. J. Hwang and Y. M. Oh



Motivation: Cover (as uniformly as possible) a plate with semiconducting material. Essential for mobile phone industry.

Varios problems: ejection and vibrations of the piezo device, drop impact and drop evaporation.

Motivation

The evaporation of droplets is a classical problem in fluid mechanics: Maxwell(1877) and Langmuir(1918)

The main assumption of their works are

- the evaporation process is diffusion-controlled
- quasi-stationary
- isothermal
- the interface of the drop is at local equilibrium

the simplifying hypotheses

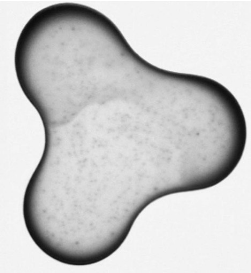
They computed the evolution of a spherical drop undergoing evaporation

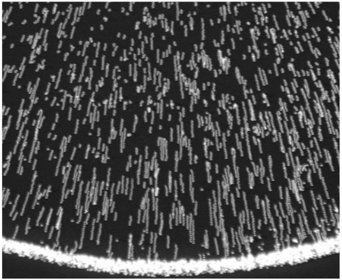
Motivation

After about one century later, the problem of evaporating drops has attracted a great deal of attention in the physics society again:

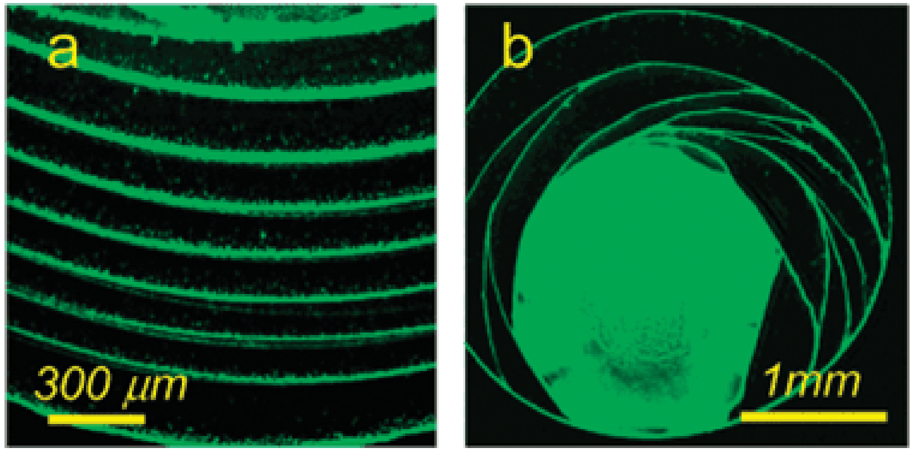
the pioneering work is

Deegan, R.D. et al.: Capillary flow as the cause of ring stains from dried liquid drops. Nature. 389, 827–829 (1997)





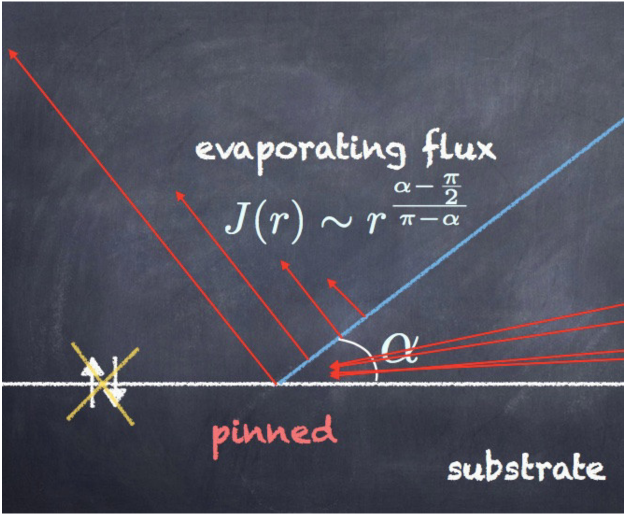
Stick-slip motion of the contact line (pinned-depinned) and the formation of deposit ring patters



Fluorescent micrographs of typical DNA multiple-ring pattern formed by evaporation of DNA droplets
H. Ma, J. Hao, ***Chem. Soc. Rev.***, 2011,**40**, 5457-5471

Motivation

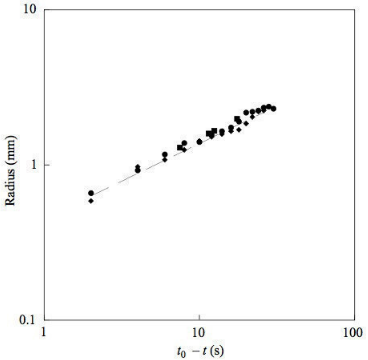
The key analysis by Deegan et al.(1997)'s work is based on **the simplifying hypotheses** and the wedge geometry near the pinned contact line



Motivation: the shrinking-droplet problem

- the evaporation of a completely wetting liquid on a perfectly flat surface

Many experiments reveal that the radius of such a drop goes tozero in finite time, with a characteristic scaling exponent closeto 1/2



Evaporation of hexane droplets deposited on mica. $R \propto (t_0 - t)^a$; $a = 0.47$.

Motivation: the shrinking-droplet problem

The simple argument based on Deegan et al.(1997)'s work:

1) the evaporation rate is proportional to the perimeter of the droplet

$$\frac{dV}{dt} \propto -2\pi R$$

2) assume that the contact angle θ is constant in time

3) by using

$$V \propto R^2 \cdot \text{height} \propto R^3\theta,$$

we finally deduce

$$R \propto (t - t_0)^{1/2}$$

A question is thus on the origin of the exponent $1/2$

We now give our study on the coffee-stain problem and the shrinking-droplet problem

The shrinking-droplet problem

First, we address a simple case by neglecting a substrate

This case can be reminiscent of the classical works of Maxwell(1877) and Langmuir(1918)

the simplifying hypotheses and the spherical symmetry lead to the classical D^2 law, i.e., the square of the drop-diameter D decreases linearly in time

$$R \propto (t - t_0)^{1/2}$$

Our result on the shrinking-droplet problem
Evaporating drops with arbitrary shapes close to a sphere also obey the D^2 law and the asymptotic shape of the drop is generically an ellipsoid

The shrinking-droplet problem: the model

$$
\begin{cases}
-\nabla p + \Delta \mathbf{u} = 0 & \text{in } \Omega(t), \\
\operatorname{div} \mathbf{u} = 0 & \text{in } \Omega(t), \\
\mathbf{T}[\mathbf{u}, p]\mathbf{n} = -\sigma\kappa\mathbf{n} & \text{on } \Gamma(t), \\
\int_{\Omega(t)} \mathbf{u}\, dV = 0, & \\
\int_{\Omega(t)} (\mathbf{u} \times \mathbf{x})\, dV = 0, & \\
\Delta\phi = 0 & \text{in } \mathbb{R}^3 \setminus \overline{\Omega}(t), \\
\phi = 1 & \text{on } \Gamma(t), \\
\phi \to 0 & \text{as } |\mathbf{x}| \to \infty,
\end{cases}
\tag{1}
$$

$$
v_{\mathbf{n}} = \mathbf{u} \cdot \mathbf{n} + \frac{1}{2}\frac{\partial \phi}{\partial \mathbf{n}} \quad \text{on } \Gamma(t), \tag{2}
$$

$$
\Gamma(0) = \Gamma^0, \tag{3}
$$

The shrinking-droplet problem: the model

where $\Gamma(t)$ denotes the boundary of $\Omega(t)$, Γ^0 is the initial shape of the free boundary, $\mathbf{T}[\mathbf{u}, p] = -p\mathbf{I} + \nabla\mathbf{u} + (\nabla\mathbf{u})^T$ is the stress tensor, σ is a given positive constant, κ is the mean curvature of $\Gamma(t)$ (κ is positive for a sphere), and $v_{\mathbf{n}}$ is the velocity of the free boundary in the direction of the outward normal

The shrinking-droplet problem: the self-similar solution

The system (1)-(3) has a self-similar solution given by

$$\begin{cases} p = \sigma(1-t)^{-1/2}, \\ \mathbf{u} = \mathbf{0}, \\ \phi = |\mathbf{x}|^{-1}(1-t)^{1/2}, \\ \Gamma(t) = \{|\mathbf{x}| = (1-t)^{1/2}\}. \end{cases} \tag{4}$$

The only self-similar solutions of (1)-(3) with a power-law structure are the ones given by (4)

The D^2 law is satisfied by (4)

The main aim of this study is therefore to prove **the stability of the self-similar solution (4)**

The shrinking-droplet problem: stable ellipsoidal collapse

Fontelos, M.A., Hong, S.H., Hwang, H.J.: Arch. Ration. Mech. Anal.(2015)

Theorem

Assume that the initial shape Γ^0 *is given by* $\{r = 1 + \epsilon g^0(\theta, \varphi)\}$*, where* $g^0 \in H^6(\mathbb{S})$*. Then, for a sufficiently small* $\epsilon > 0$*, there exists a unique solution to the free-boundary problem (1)-(3) such that*

$$\frac{\Gamma(t) - \epsilon \mathbf{x}_0}{R(t)} = \left\{ r = 1 + \epsilon g^{\xi}(\theta, \varphi, t) \right\},$$

$$R(t) = \left[1 - \frac{t}{1 + \epsilon t_0 (2\sqrt{\pi})^{-1}} \right]^{1/2}$$

on the time interval $0 \le t < 1 + \epsilon t_0 (2\sqrt{\pi})^{-1}$ *for some* $(\mathbf{x}_0, t_0) = (\mathbf{x}_0(\epsilon), t_0(\epsilon)) \in \mathbb{R}^3 \times \mathbb{R}$*, and we have:*

The shrinking-droplet problem: stable ellipsoidal collapse

Theorem

(a) The solution $\left\{r = 1 + \epsilon g^{\xi}(\theta, \varphi, t)\right\}$*, as well as the corresponding* $(\mathbf{u}, p, \phi)$*, of the transformed problem is unique in the function space* $\mathscr{X}$ *to be defined in the corresponding theorem*

(b) The quantity (x_0, t_0) *satisfies*

$$|\mathsf{x}_0 - \tilde{\mathsf{x}}_0| = O(\epsilon), \quad |t_0 - \tilde{t}_0| = O(\epsilon),$$

where $(\tilde{\mathsf{x}}_0, \tilde{t}_0) \in \mathbb{R}^3 \times \mathbb{R}$ *merely depends on the* $l \leq 2$ *terms in the spherical harmonic expansion of* g^0*.*

The shrinking-droplet problem: stable ellipsoidal collapse

Theorem

(c) There exist constants $C > 0$ and $0 < \lambda_0 < 1$ such that

$$\sup_{(\theta,\varphi)\in\mathbb{S}} |g^{\xi}(\theta,\varphi,t) - g^{\mathfrak{E}}(\theta,\varphi)| \leq CR^{\lambda_0}(t),$$

for all $t \in \left[0, 1+\epsilon t_0(2\sqrt{\pi})^{-1}\right)$, *where* $\left\{r = 1+\epsilon g^{\mathfrak{E}}(\theta,\varphi)\right\}$ *is an ellipsoid which satisfies*

$$\sup_{(\theta,\varphi)\in\mathbb{S}} |g^{\mathfrak{E}}(\theta,\varphi) - g^{0}_{0,0}Y_{0,0} - \sum_{|m|\leq 2} e^{-\sigma\frac{20}{19}} g^{0}_{2,m} Y_{2,m}| = O(\epsilon).$$

The shrinking-droplet problem: remark on Theorem

If neglecting the Stokes equations, i.e., $\sigma = 0$, then it is known that the system (1)-(3) has a shrinking self-similar solution whose asymptotic shape near extinction is an ellipsoid (the Hele-Shaw problem)

Surface tension usually plays the role of a stabilizing force $\Longrightarrow$ one may suspect that, if $\sigma > 0$, (1)-(3) has a shrinking solution whose shape converges to a sphere or ...

The shrinking-droplet problem: remark on Theorem

Our result confirms that, for the case of evaporating drops, surface tension merely produces a shifting on the center, orientation, and semi-axes of a "$\sigma = 0$ ellipsoid", i.e., in Theorem,

$$\sup_{(\theta,\varphi)\in\mathbb{S}} |g^{\mathfrak{E}}(\theta,\varphi) - g^0_{0,0} Y_{0,0} - \sum_{|m|\leq 2} e^{-\sigma\frac{20}{19}} g^0_{2,m} Y_{2,m}| = O(\epsilon),$$

but not modification in the ellipsoidal structure near extinction

The reason for this ellipsoidal structure is that **the evaporative flux $\frac{\partial \phi}{\partial \mathbf{n}}$ overwhelms $\mathbf{u} \cdot \mathbf{n}$ near extinction**

Indeed, this is the key point of the proof!

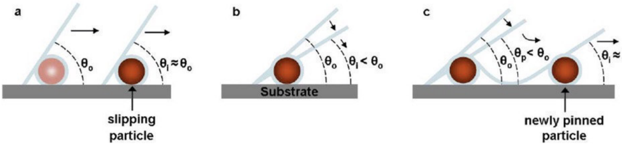
The coffee-stain problem

We also address a simple case for the coffee-stain problem

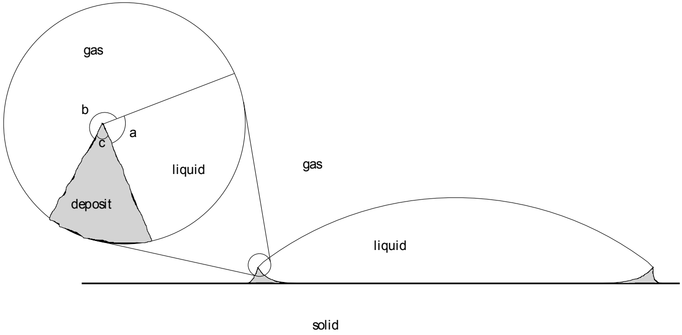
Some common features of deposit-patterns are now apparent: the formation of periodic or quasi-periodic deposit-patterns, due to stick-slip motion of the contact line

The contact line remains pinned (stick) at the growing deposit up to a moment when it unpins

After unpinning, the contact line moves (slips) towards a different location where the contact line pins again and a new deposit starts to grow



The coffee-stain problem: the model

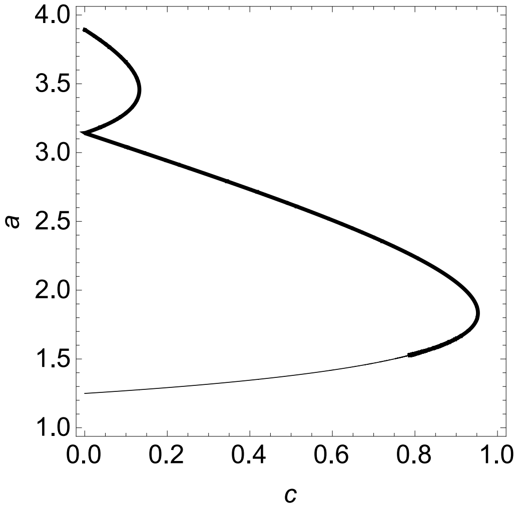


The deposit shape is idealized by a wedge

Our analysis yields criteria for the stability/instability of the contact line, where instability represents a transition from pinned to unpinned contact line representative of stick-slip motion

Our formulation relies on the introduction of suitable energies

The coffee-stain problem: the pinned-unpinned transition



The coffee-stain problem: the pinned-unpinned transition

The plot for the equilibrium configurations

In the evaporation dominated limit, the threshold of the stability is the point between the thick line and the thin line, where $c = 0.78706752635 \simeq 45.1^\circ$

The thick line represents stable configurations

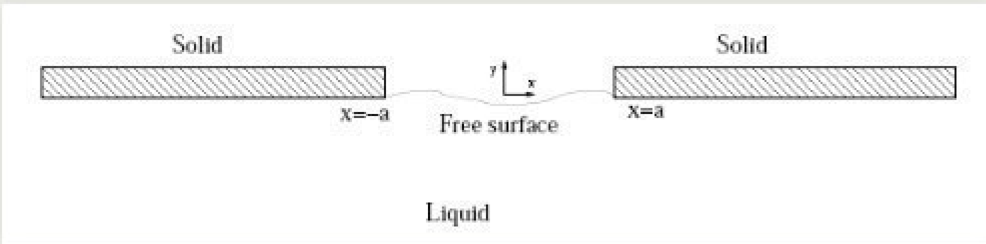
The coffee-stain problem: remark

Our criteria for pinning and depinning of the contact line are based on the energy dissipation/supply mechanism

When the energy supply exceed some point of $V_e + V_\sigma > 0$, depinning occurs

$$\widetilde{\frac{\partial^2 f}{\partial t \partial r}}^{\alpha+2} = CM(\lambda)(\widetilde{r^{\frac{\pi}{2b}-1}\frac{\partial f}{\partial r}})^{\alpha+1} + \frac{\sigma}{2\mu} N(\lambda) \widetilde{\frac{\partial f}{\partial r}}^{\alpha+1} \quad \text{on} \quad \theta = 0$$

Capillary oscillations at the exit of a nozzle



Capillary oscillations at the exit of a nozzle,

H.J.Kim, M.A.Fontelos & H.J.Hwang

IMA Journal of Applied Mathematics (2014)

Surface Waves

Capillary / Gravity / Gravity-capillary

Capillary waves

dominated by the surface tension

Gravity waves

dominated by the gravity/the buoyancy

Gravity-capillary waves

dominated by the both effects of surface tension and gravity

Capillary waves

Frequencies

Capillary water waves:

$$\omega = \sqrt{\frac{\sigma}{\rho_w + \rho_a} |k|^3},$$

- ρ_w, ρ_a : the densities of water, air, resp.,
- σ : the surface tension coefficient,
- k : the wavenumber of the capillary wave.

Gravity-capillary waves:

$$\omega = \sqrt{\left(\frac{\rho_w - \rho_a}{\rho_w + \rho_a} g + \frac{\sigma}{\rho_w + \rho_a} k^2 \right) |k|},$$

Mathematical model

a perfect incompressible fluid

Euler equation for inviscid incompressible flow:

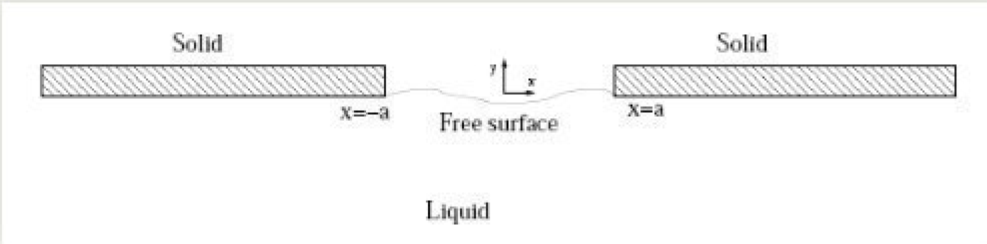
$$\nabla \cdot \mathbf{u} = 0, \text{ in } D(t),$$
$$\rho \left(\mathbf{u}_t + (\mathbf{u} \cdot \nabla)\mathbf{u} \right) = -\nabla p.$$

+ Irrotational:

$$\Delta \phi = 0, \text{ in } D(t),$$
$$\phi_t + \frac{1}{2} |\nabla \phi|^2 + \frac{\sigma \kappa}{\rho} = 0, \text{ on the free surface},$$

- ρ : the density of the fluid, σ : surface tension coefficient,
- κ : the curvature of the free surface,
- $\mathbf{u} = \nabla \phi$, i.e., ϕ is the potential function of the velocity $\mathbf{u}$,
- $D(t)$ is the fluid domain bounded by solid walls and the free boundary.

Contact line B.C. & volume constraint



(1-1) Pinned end B.C. $f|_{x=\pm a} = 0,$

(1-2) Free end B.C. $f_x|_{x=\pm a} = 0,$

(2)Volume conservation $\int_{-a}^{a} f(x,t)dx = 0.$

We consider both cases :

(1-1) with (2) and (1-2) with (2)

Linearized problem

- $\triangle\phi = 0,\ y \leq 0,$
- $\phi_x,\ \phi_y \to 0,\ \text{ as }\ y \to -\infty\ \text{ or }\ |x| \to \infty,$

  with boundary conditions:
- $\phi_y = 0,\ y = 0,\ |x| > a,$
- $\phi_y = f_t,\ y = 0,\ |x| \leq a,$
- $\phi_t = \frac{\sigma}{\rho} f_{xx},\ y = 0,\ |x| \leq a.$

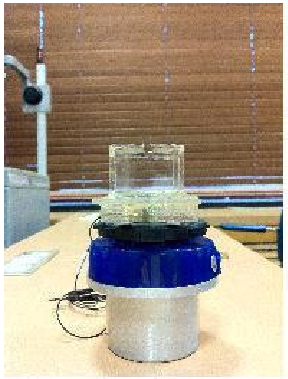
Integro-differential equation

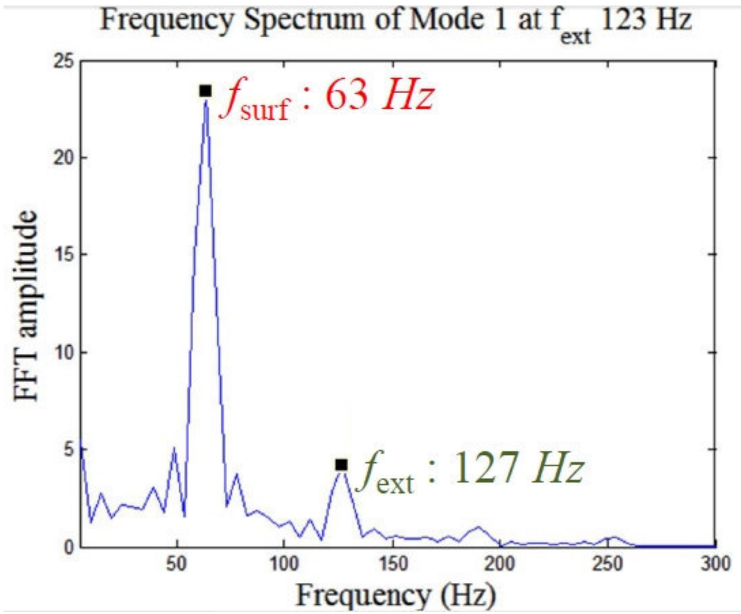
$$\frac{\sigma}{\rho} f_{xxx}(x,t) = -\frac{1}{\pi} \text{P.V.} \int_{-a}^{+a} \frac{f_{tt}(z,t)}{x-z}\, dz, \ |x| \leq a.$$

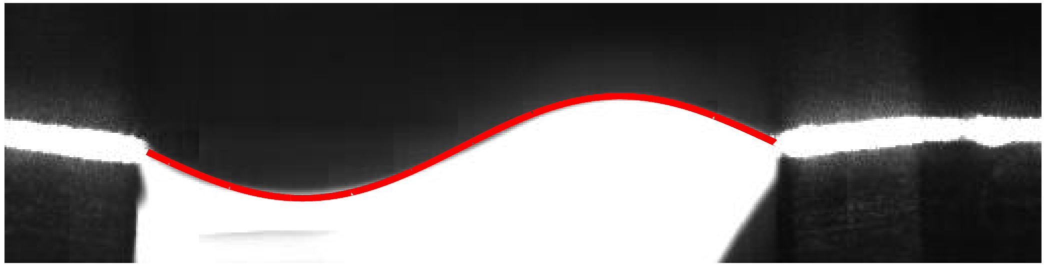
- Re-scale variables: $x \to ax, \ \ t \to \sqrt{\frac{\rho a^3}{\sigma}}\, t, \ \ f \to af$
- Separation method: $f(x,t) = A(t)S(x) = e^{i\sqrt{\lambda}t} S(x)$

$$\lambda \left(\frac{1}{\pi} \text{P.V.} \int_{-1}^{1} \frac{S(z)}{x-z}\, dz \right) = S'''(x)$$

$$\iff \ \ \lambda S(x) = -\frac{1}{\pi} \text{P.V.} \int_{-1}^{1} \sqrt{\frac{1-x^2}{1-z^2}}\, \frac{S'''(z)}{x-z}\, dz.$$



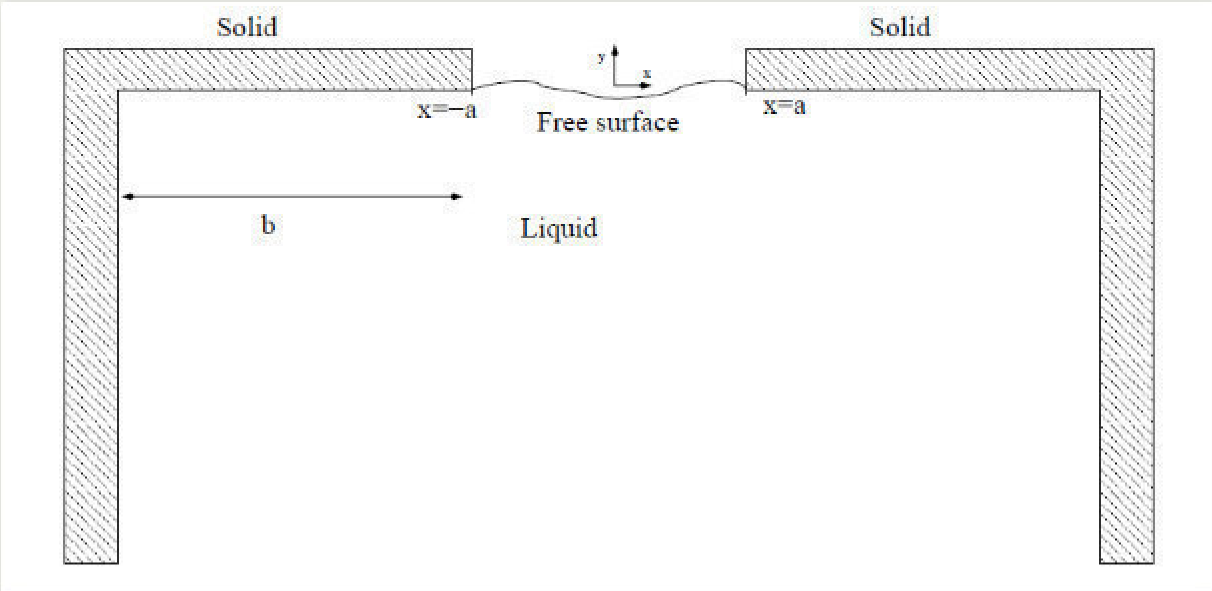




Numeric: 63.67 Hz

Conformal mapping

We can *extend* to the following problem for both cases $b = 0$ and $b >> a$:



$$\left(\frac{\pi}{2(a+b)}\right)\cos\left(\frac{\pi x}{2(a+b)}\right)\frac{1}{\pi}P.V.\int_{-a}^{a}\frac{h_{tt}(x',t)dx'}{\sin\left(\frac{\pi x}{2(a+b)}\right)-\sin\left(\frac{\pi x'}{2(a+b)}\right)} = -\frac{\sigma}{\rho}h_{xxx}$$

$$\Longrightarrow \frac{1}{\pi}P.V.\int_{-a}^{a}\frac{h_{tt}(x',t)}{x-x'}\,dx' = -\frac{\sigma}{\rho}h_{xxx},\ b\to\infty.$$

Capillary oscillations at a circular orifice

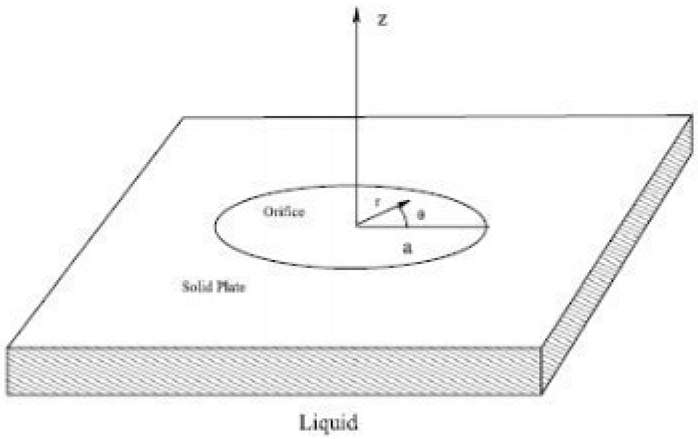


Fig. 1. Sketch of the physical setting of the problem.

Capillary oscillations at a circular orifice,

H.J.Kim, M.A.Fontelos & H.J.Hwang

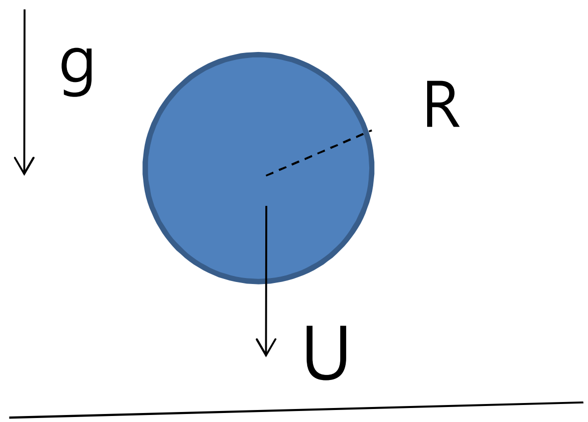
Applied Mathematics Letter (2013)

Integro-differential equations

$$\varphi \equiv \sin(n\theta)\Phi(r,z,t), \;\; h(r,\theta,t) \equiv \sin(n\theta)H(r,t),$$

$$\begin{aligned} h_t &= \varphi_z, & z=0, \quad r<1, \\ \varphi_t - \frac{\sigma}{\rho a^3}\Delta_\perp h &= 0, & z=0, \quad r<1. \end{aligned} \quad \Longrightarrow \quad \begin{aligned} H_t &= \Phi_z, \\ \Phi_t &= \frac{\sigma}{\rho a^3}\left[\frac{1}{r}\frac{d}{dr}\left(r\frac{dH}{dr}\right) - \frac{n^2 H}{r^2}\right] \end{aligned}$$

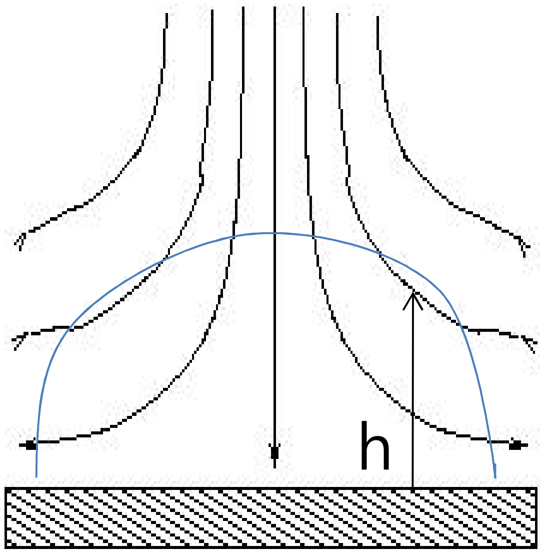
$$\Longrightarrow \frac{\sigma}{\rho a^3}\left[\frac{1}{r}\frac{d}{dr}\left(r\frac{dH(r,t)}{dr}\right) - \frac{n^2 H(r,t)}{r^2}\right] = \int_0^1 r' I_n(r,r') H_{tt}(r',t)\,dr'.$$



γ: surface tension, ν: viscosity ρ: density

$$\mathrm{We} = \frac{2\rho R U^2}{\gamma}, \quad \mathrm{Re} = \frac{2RU}{\nu}, \quad \mathrm{Fr} = \frac{U^2}{2gR},$$

Explicit inviscid solution:



$$v_r = \frac{r}{t}, \quad v_z = -\frac{2z}{t}. \quad p(z,r,t)/\rho = -3z^2/t^2.$$

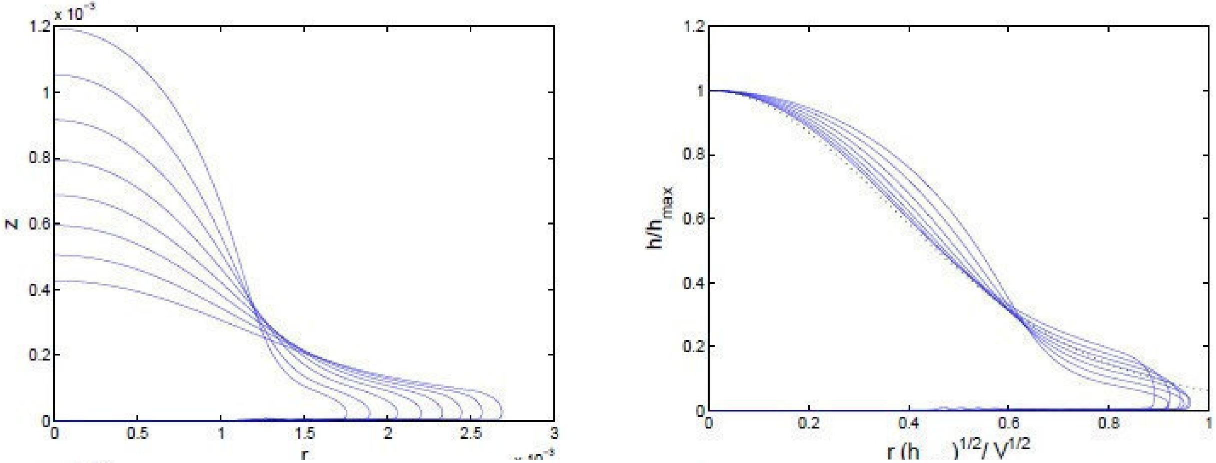
$$\partial_t h + v_r \frac{\partial h}{\partial r} = v_z.$$

This equation has the similarity solution

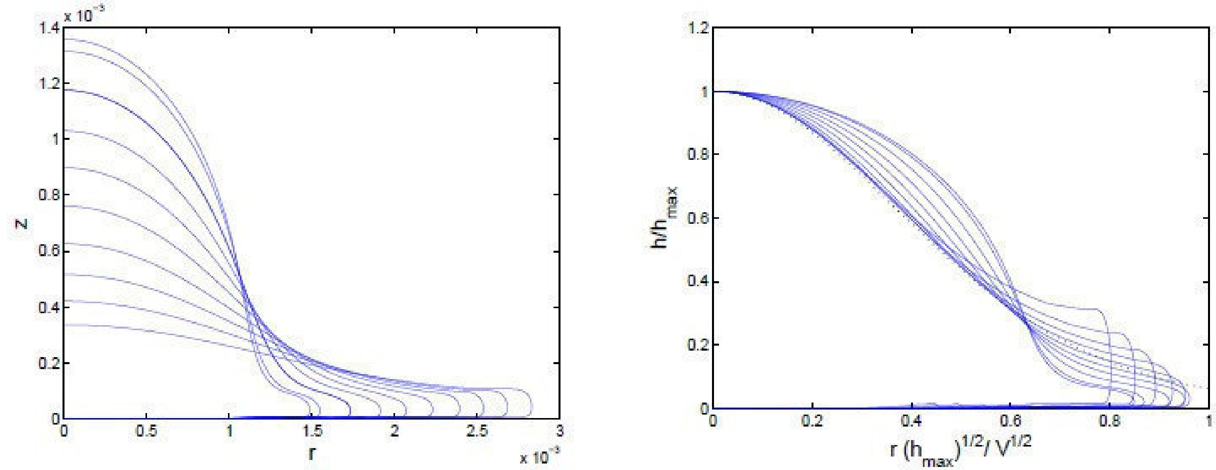
$$h(r,t) = \frac{1}{t^2} H\left(\frac{r}{t}\right), \qquad p = -3h^2(z,t)/t^2.$$

Similarity solution: $H_s(x) = 1/(1 + Cx^2)^6,$

with the constant $C = 0.625.$



$Re = 400$ and $We = 400$



$Re = 400$ and $We = 8000$

Kinetic energy
Into surface energy

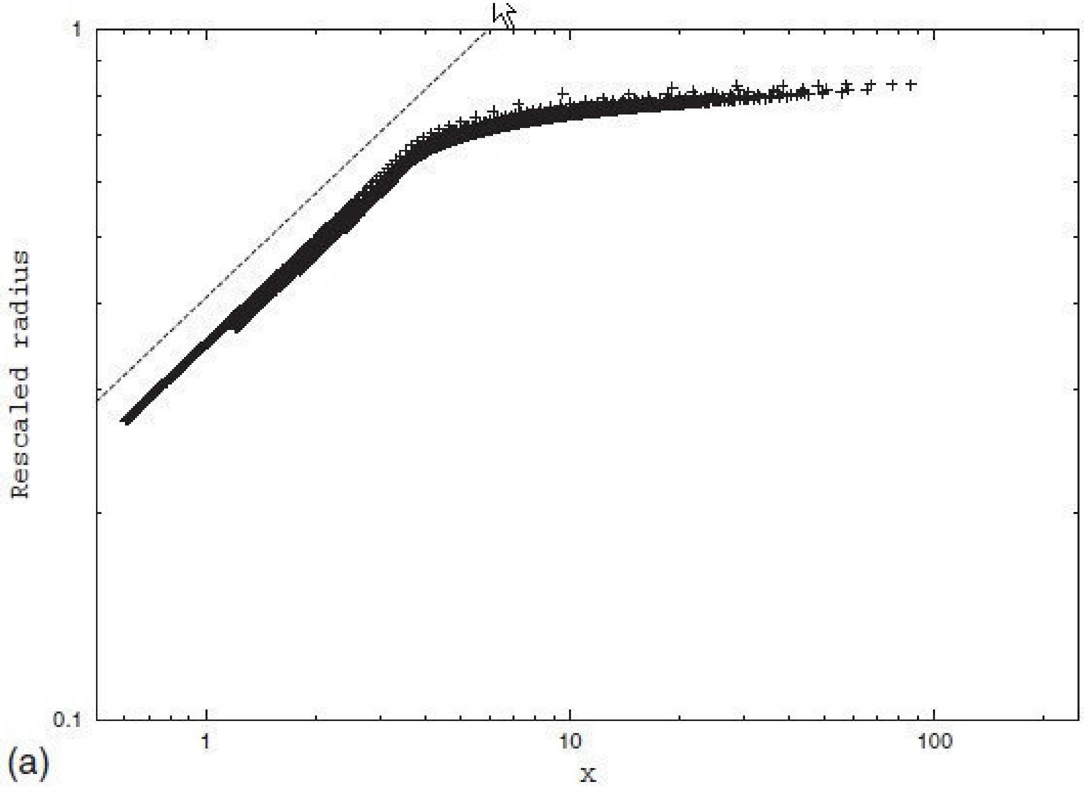
$$\frac{R_m}{R} \sim \left(\frac{\text{We}}{6}\right)^{1/2}.$$

Kinetic energy
Into viscous disip.

$$\frac{R_m}{R} \sim C_v \text{Re}^{1/5},$$

Scaling law for the maximum radius:

$$\frac{R_m}{R} \propto \text{Re}^{1/5} f_c(\text{WeRe}^{-2/5})$$



(a)

$(R_m/R)/\text{Re}^{1/5})$ is plotted (a) as a function of $x=\text{We}/\text{Re}^{2/5}$; the expected behavior for small x, $\sqrt{x/6}$, is indicated in dotted line; (b) as function of $x=\text{We}/\text{Re}^{4/5}$, again showing the expected behavior for small x, $(8x/9)^{1/4}$ (dotted line).