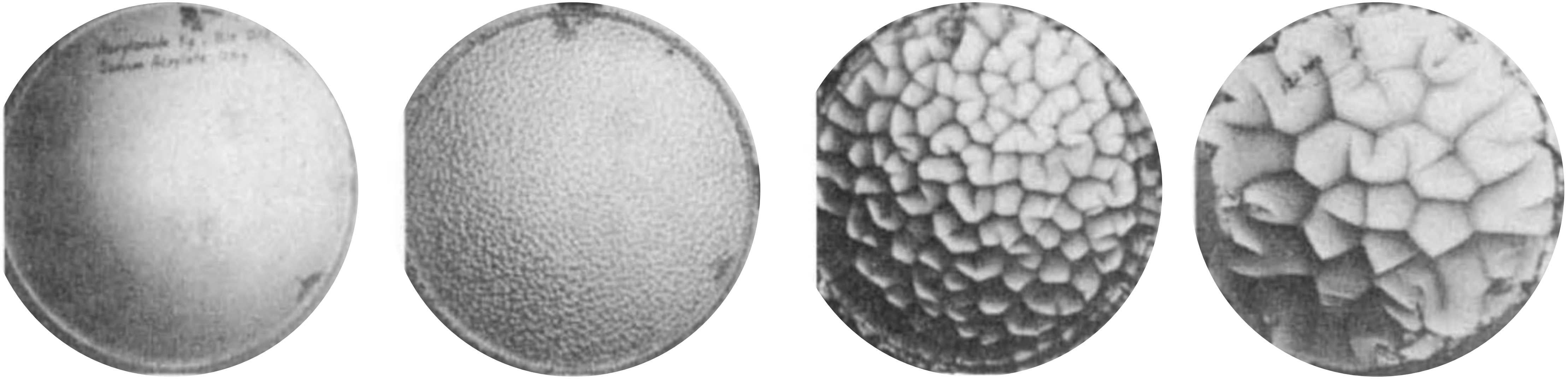

# Buckling and swelling instabilities of super-absorbent gels

**Joseph Webber** *University of Warwick*

BioActive & Non-Newtonian Fluids 2024

*Image from Tanaka et al. (Nature, 1987)*

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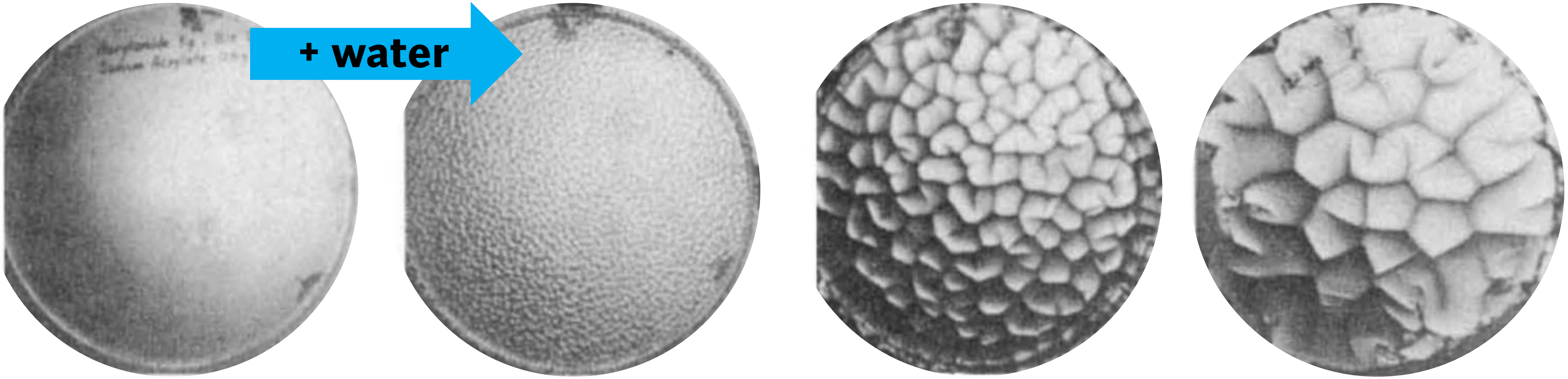
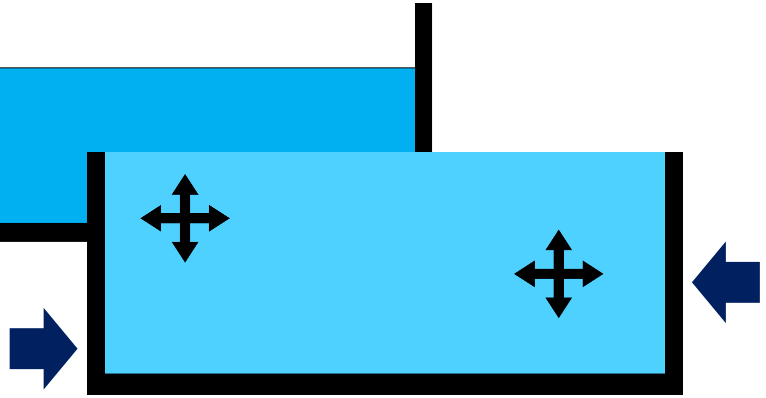
# Introduction



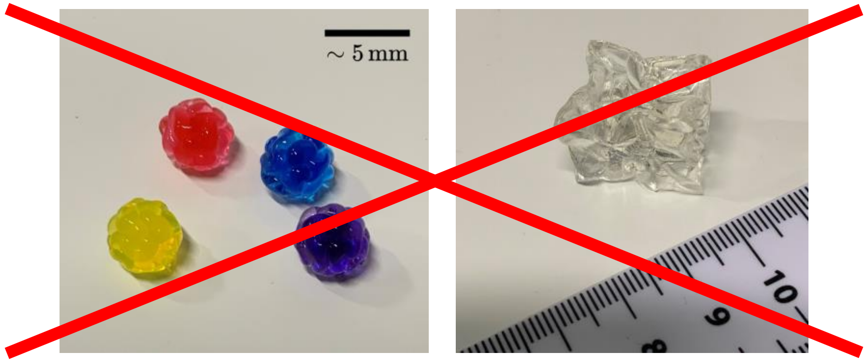

- Wrinkles form when swelling in confined geometries
- Sometimes appear, sometimes coarsen, sometimes disappear after some time, sometimes persist – why?

# Introduction

- We limit our attention to cases where wrinkles arise from mechanical confinement and not differential swelling



Spherical beads just after immersion




Swelling of a cube

- Wavelength of patterns grows like $t^{1/2}$ (at least at early times)

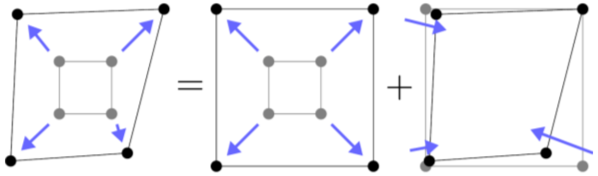
- Seek both the mechanism of wrinkle growth and wavelength selection

# Understanding swelling

**Webber & Worster** *and*
**Webber, Etzold & Worster**
*JFM*, 2023

- Consider all deformation and swelling relative to a 'fully-swollen' base state where $\phi \equiv \phi_0$ then define a Cauchy strain tensor. Linearise around small deviatoric strains.

$$\mathsf{e} = \frac{1}{2}\left[\nabla\boldsymbol{\xi} + \nabla\boldsymbol{\xi}^{\mathrm{T}}\right] = \left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/2}\right]\mathsf{I} + \epsilon$$



Deviatoric strain

Osmotic pressure

Shear modulus

$$\boldsymbol{\sigma} = -\left[p + \Pi(\phi)\right]\mathsf{I} + 2\mu_s(\phi)\epsilon$$

Pore (pervadic) pressure

$$\boldsymbol{u} = -\frac{k(\phi)}{\mu_l}\nabla p \quad \& \quad \nabla\cdot\boldsymbol{\sigma} = \mathbf{0}$$

Darcy coefficient
*(permeability/viscosity)*

$$\frac{D_{\boldsymbol{q}}\phi}{Dt} = \nabla\cdot\left\{\frac{k(\phi)}{\mu_l}\left[\phi\frac{\partial\Pi}{\partial\phi} + \mu_s(\phi)\left(\frac{\phi}{\phi_0}\right)^{1/2}\right]\nabla\phi\right\}$$

# Understanding swelling

**Webber & Worster** *and*
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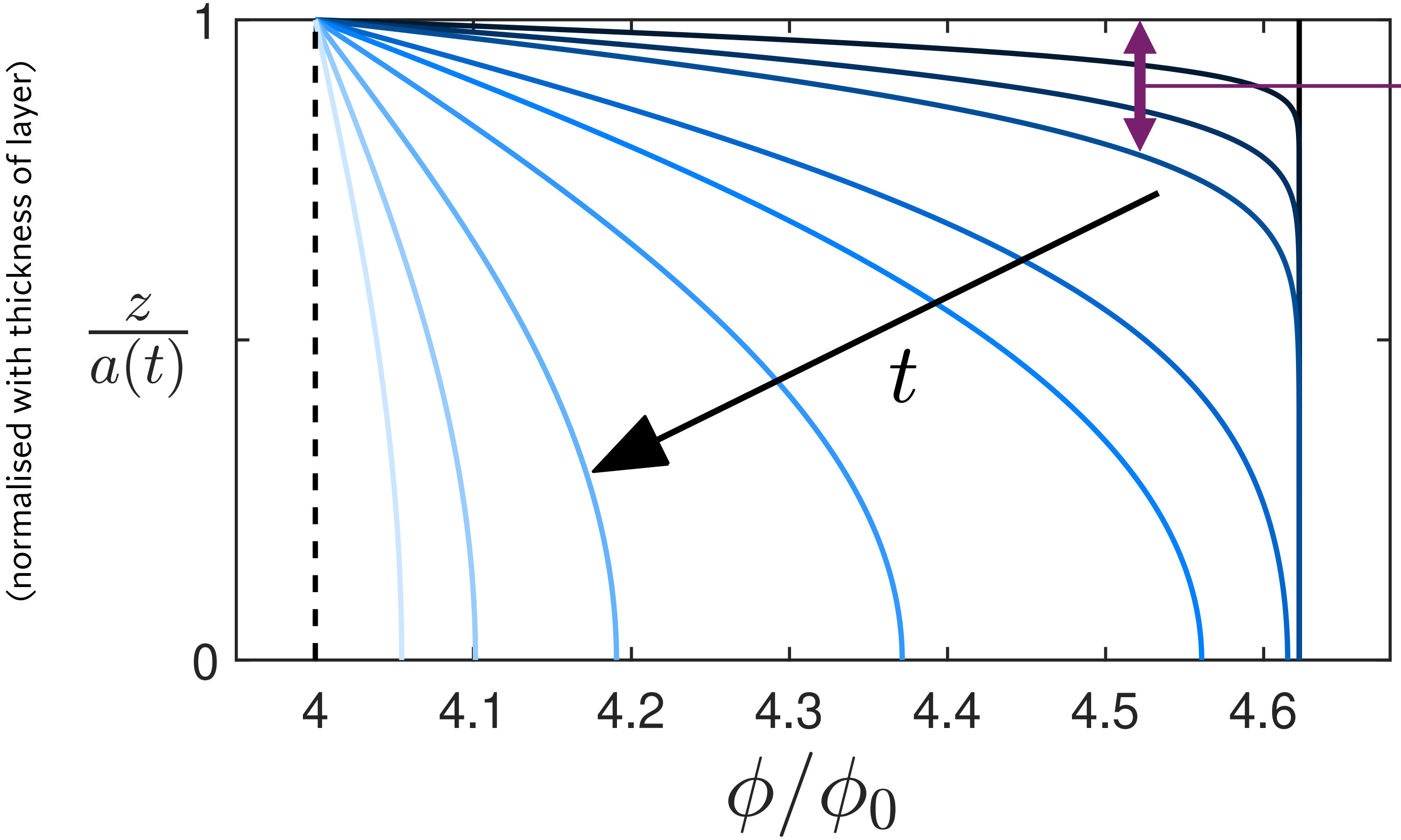
*JFM*, 2023

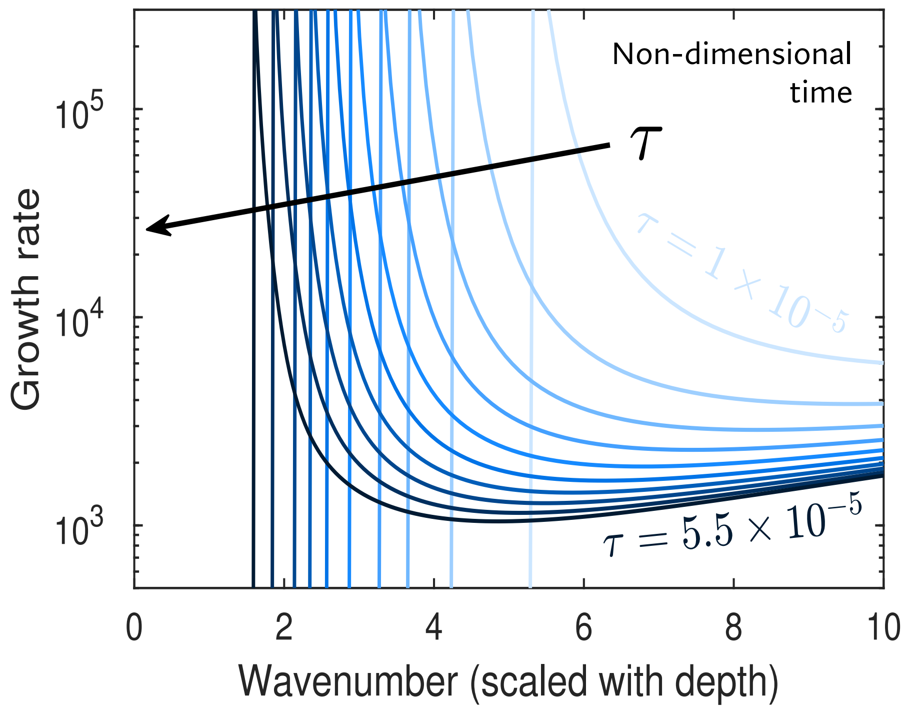


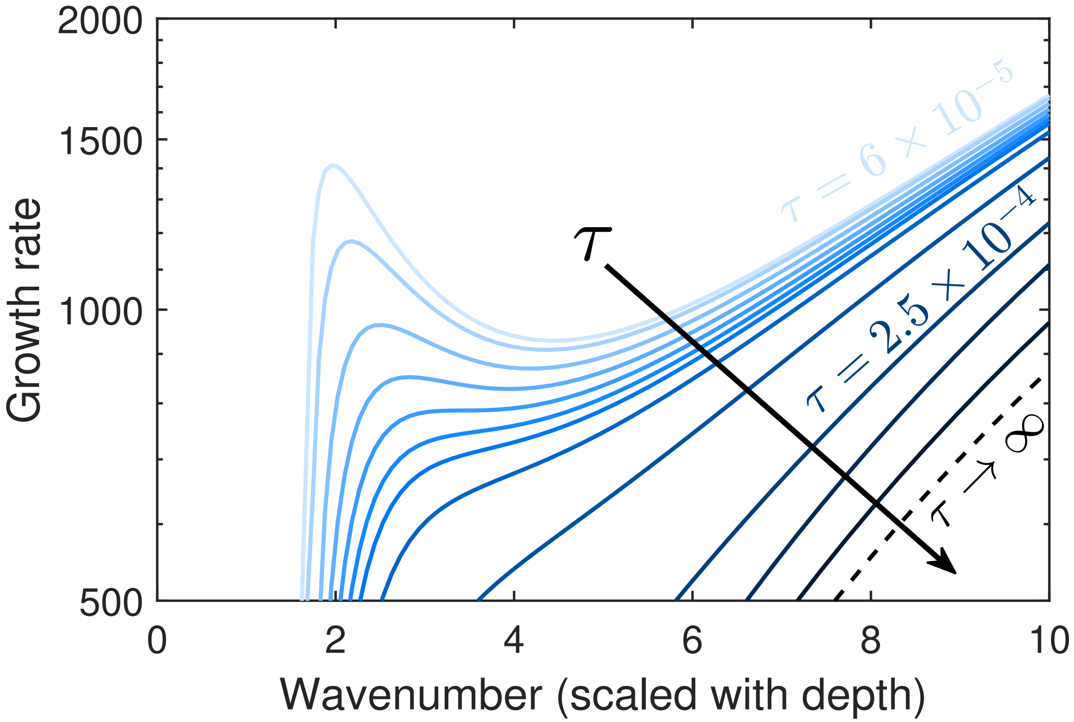

Swollen boundary layer *thickness* $\delta(t)$

Initially, when effect of base is not seen,

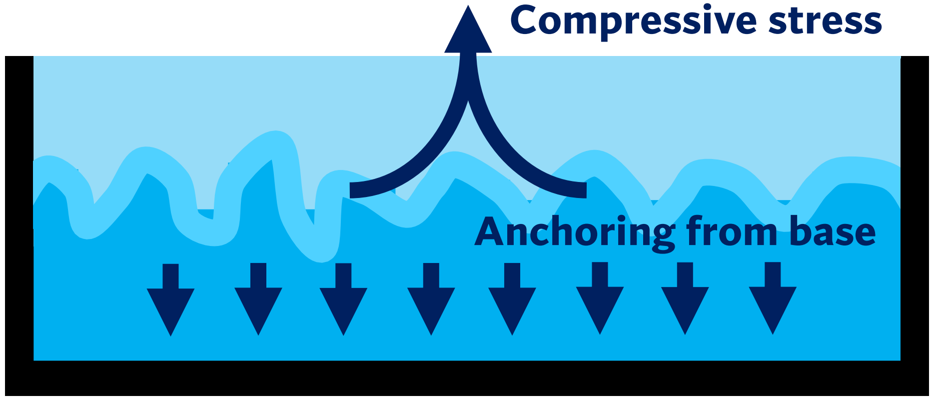
$\delta(t) \sim \sqrt{t}$

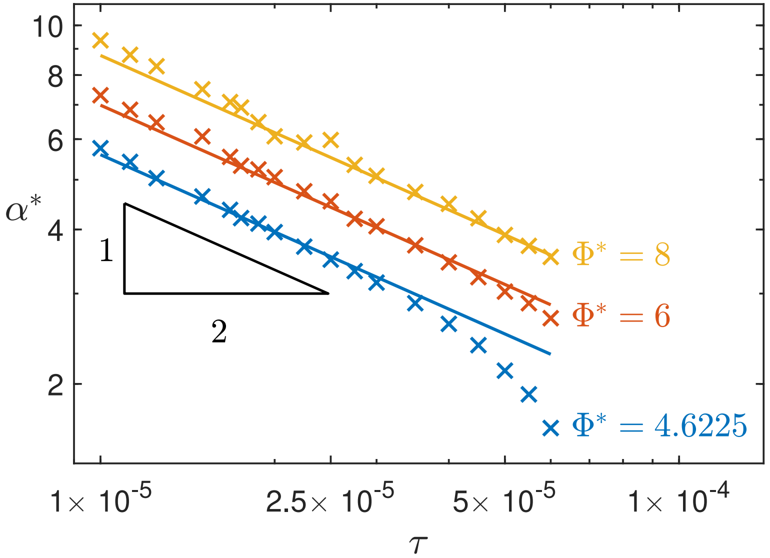
# Instabilities



# Early times – elastic buckling




- Treat the swollen layer as a linear-elastic beam or plate of thickness $\delta(t)$ attached to an elastic base of unswollen gel.
- Solving the linear elastic incompressible problem selects a finite unstable wavenumber where buckles form on the inertial timescale.

$$\alpha^* \approx \frac{1}{2\mathcal{M}\,a(t)}\left(1 + \mathcal{M}{\Phi^*}^{1/2}\right)\frac{\partial \Phi}{\partial z}$$

Shear parameter

Dry polymer fraction, scaled by $\phi_0$

- Note, at early times $\partial\Phi/\partial z \sim 1/\delta(t) \sim 1/\sqrt{t}$

# Early times – elastic buckling

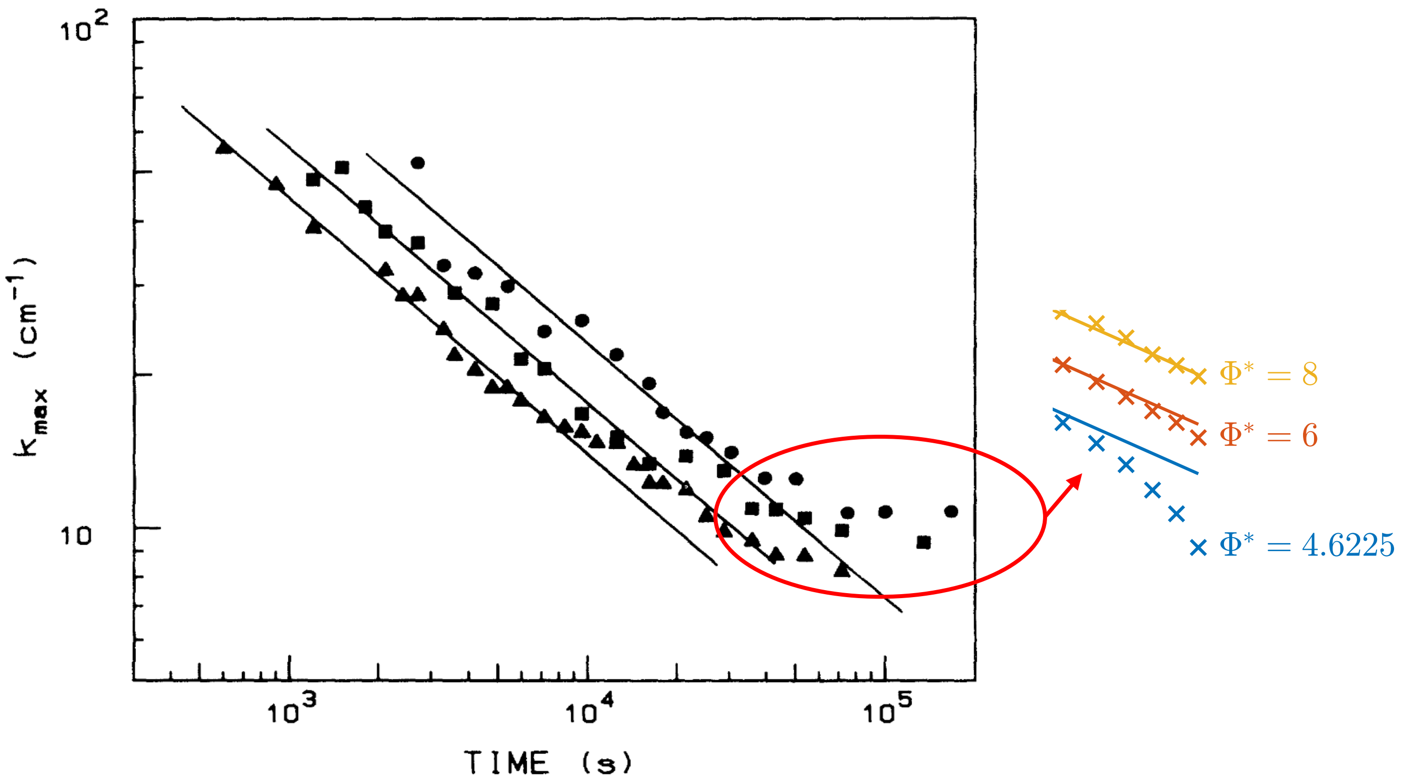
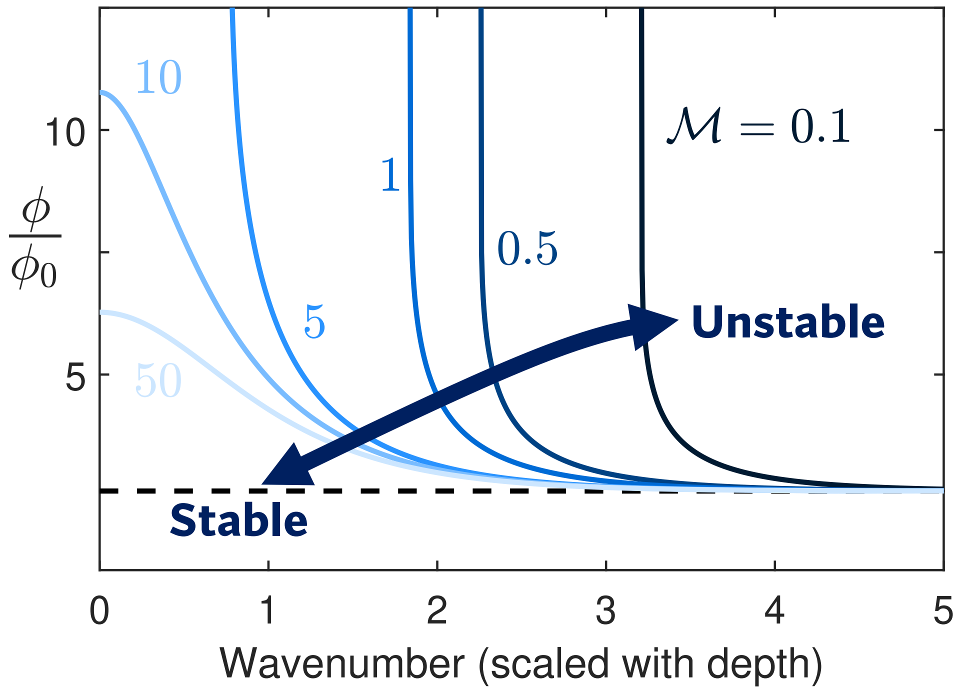


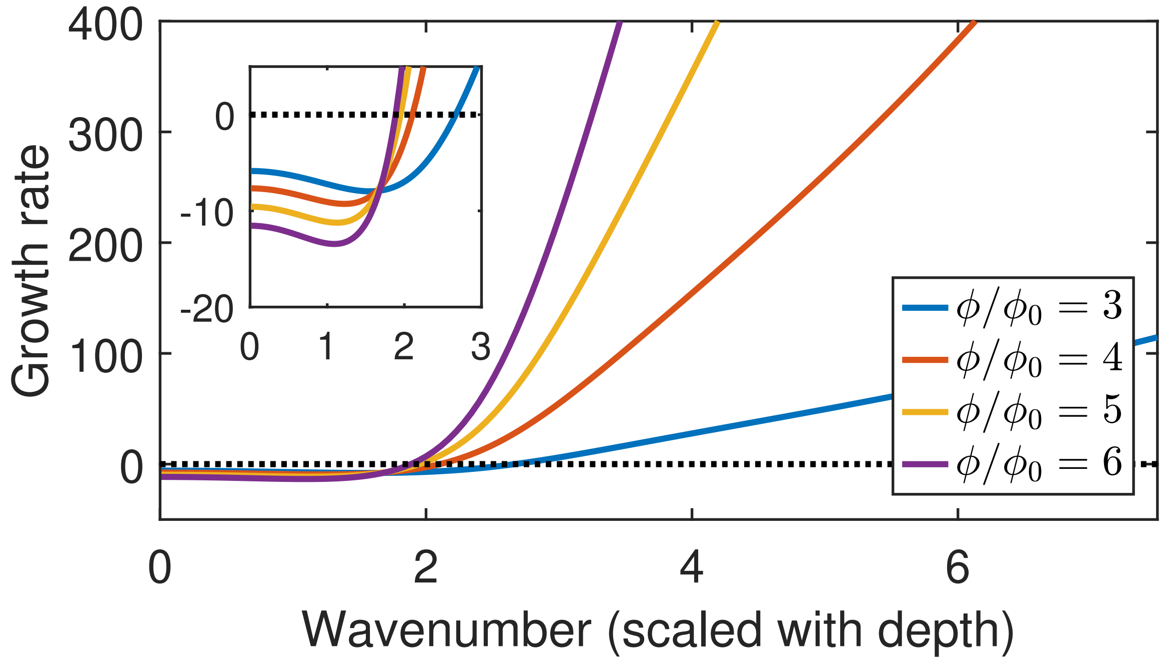

FIG. 4. Temporal change in $k_{max}$ for the gels I-1–I-3. (●) I-1; (■) I-2; (▲) I-3. All the solid lines have a slope of $-\frac{1}{2}$.

Figure from Tanaka *et al.*, Phys Rev Lett **68**:2794-2798, 1987

# Late times – diffusive wrinkling

- At late times, base state approaches a uniform polymer fraction, so stiff base + swollen interface model can no longer apply
- Perturb displacement field and carry out a linear stability analysis on uniform state

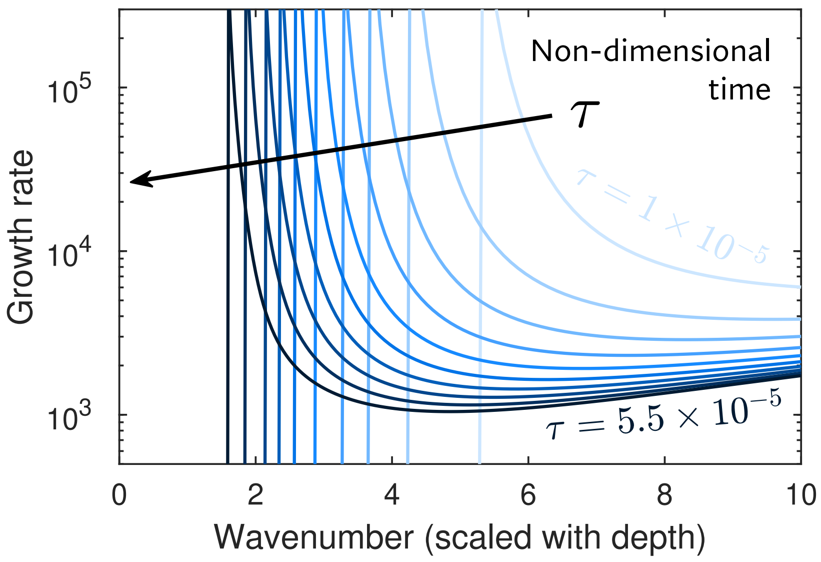


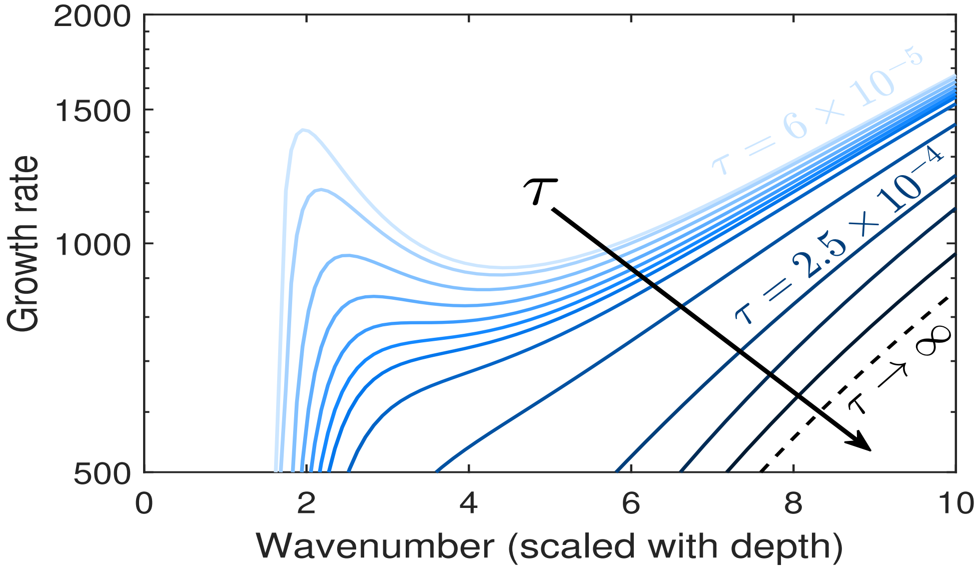



# Healing of instabilities

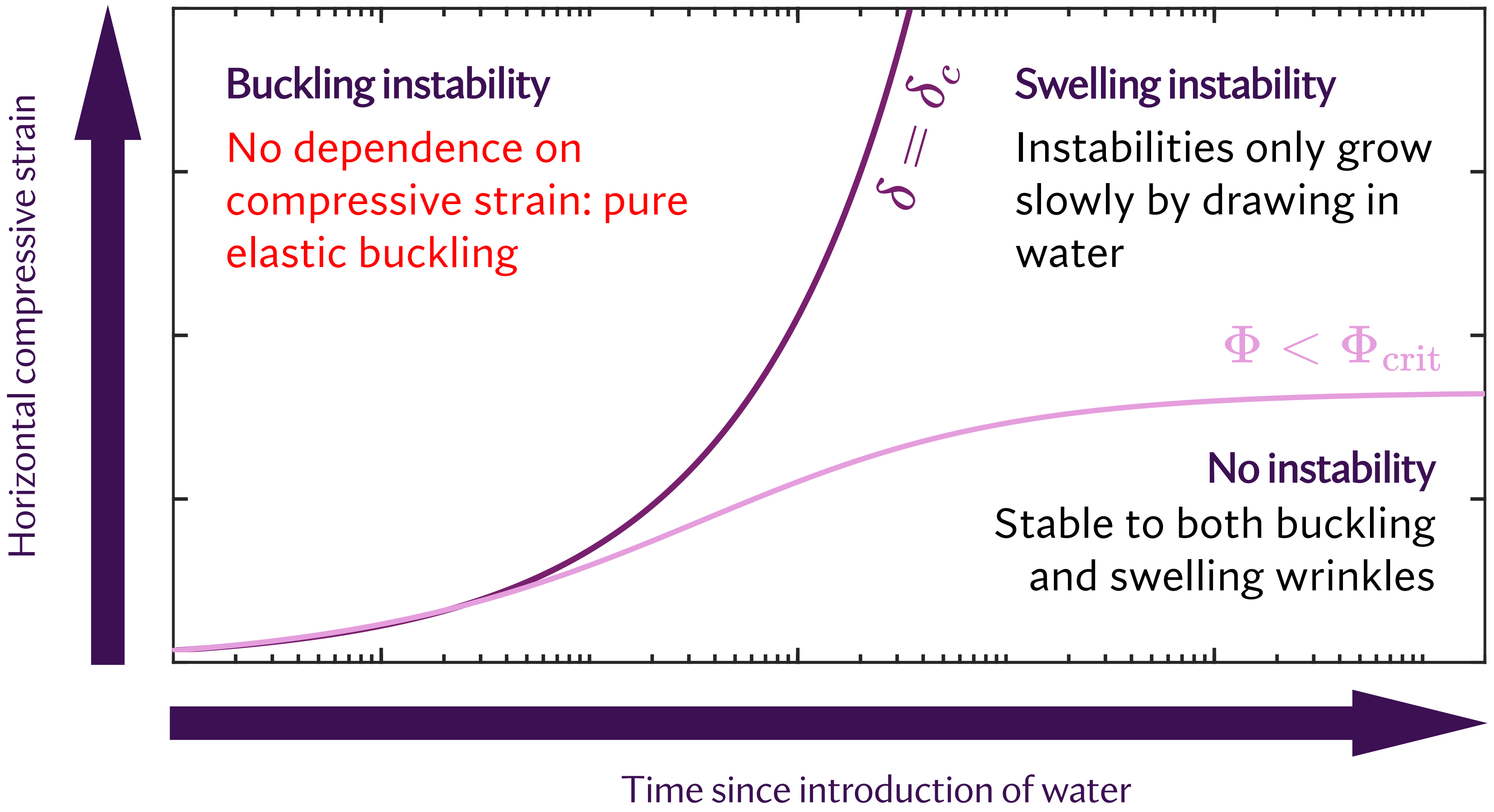
- At intermediate times, perturb a 'frozen-in-time' base state to connect the two regimes of pattern formation

- At early times, see 'peaks' corresponding to the elastic buckling, which smooth out, and result in eventually-stable or unstable situations



# Healing of instabilities



# Conclusions

- Wrinkles form *via* two distinct mechanisms at different times in the swelling process
- At early times, we see elastic buckles that form quickly and the wavelength evolves like the square root of time (*cf* Biot instability of an elastic half-space)
- At late times, the only mechanism is diffusive in nature, and this only occurs if there is sufficient compression, explaining the healing that is sometimes seen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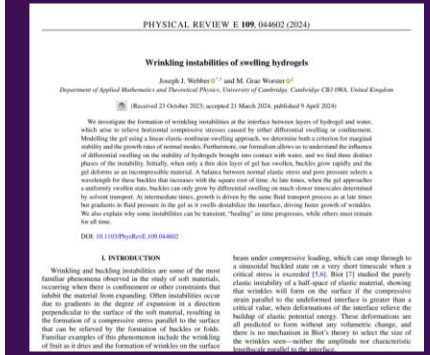
**Wrinkling instabilities of swelling hydrogels**



PHYSICAL REVIEW E 109, 044602 (2024)

**Wrinkling instabilities of swelling hydrogels**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We investigate the formation of wrinkling instabilities at the interface between layers of hydrogel and water, which arise to relieve horizontal compressive stresses caused by either differential swelling or confinement. Modelling the gel using a linear-elastic–nonlinear-swelling approach, we determine both a criterion for marginal stability and the growth rates of normal modes. Furthermore, our formalism allows us to understand the influence of differential swelling on the stability of hydrogels brought into contact with water, and we find three distinct phases of the instability. Initially, when only a thin skin layer of gel has swollen, buckles grow rapidly and the gel deforms as an incompressible material. A balance between normal elastic stress and pore pressure selects a wavelength for these buckles that increases with the square root of time. At late times, when the gel approaches a uniformly swollen state, buckles can only grow by differential swelling on much slower timescales determined by solvent transport. At intermediate times, growth is driven by the same fluid transport process as at late times but gradients in fluid pressure in the gel as it swells destabilize the interface, driving faster growth of wrinkles. We also explain why some instabilities can be transient, "healing" as time progresses, while others must remain for all time.

DOI: 10.1103/PhysRevE.109.044602

I. INTRODUCTION

Wrinkling and buckling instabilities are some of the most familiar phenomena observed in the study of soft materials, occurring when there is confinement or other constraints that inhibit the material from expanding. Often instabilities occur due to gradients in the degree of expansion in a direction perpendicular to the surface of the soft material, resulting in the formation of a compressive stress parallel to the surface that can be relieved by the formation of buckles or folds. Familiar examples of this phenomenon include the wrinkling of fruit as it dries and the formation of wrinkles on the surface

beam under compressive loading, which can snap through to a sinusoidal buckled state on a very short timescale when a critical stress is exceeded [5,6]. Biot [7] studied the purely elastic instability of a half-space of elastic material, showing that wrinkles will form on the surface if the compressive strain parallel to the undeformed interface is greater than a critical value, when deformations of the interface relieve the buildup of elastic potential energy. These deformations are all predicted to form without any volumetric change, and there is no mechanism in Biot's theory to select the size of the wrinkles seen—neither the amplitude nor characteristic lengthscale parallel to the interface.

J. J. Webber and M. G. Worster

*Phys. Rev. E* *109 (2024)*

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