Wrinkling instabilities of swelling hydrogels

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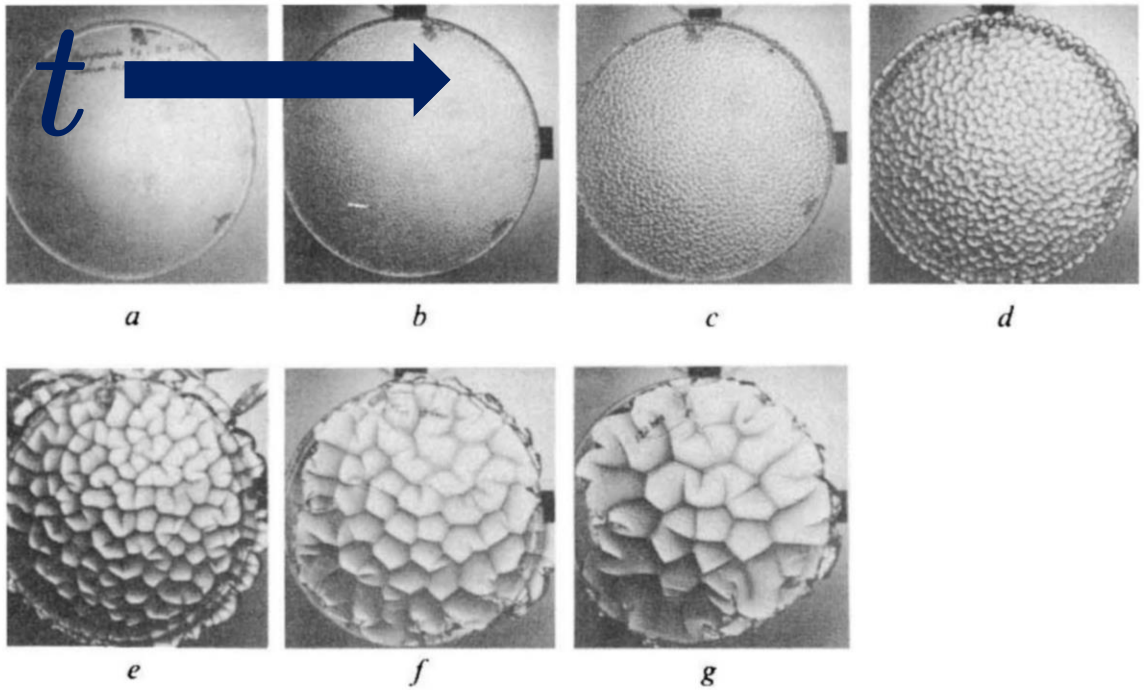
Swelling of confined gels

- When water is introduced to a ‘dry’ gel subject to mechanical confinement, wrinkles can form
- Swelling produces horizontal compressive stresses relieved by buckles



- Some gels form wrinkles, some don’t; what’s the criterion?
- Patterns smooth in time (wavelength grows like $t^{1/2}$)
- In some cases, patterns disappear
- **How do wrinkles grow?**

Figure from Tanaka *et al.*, Nature **325**:796-798, 1987

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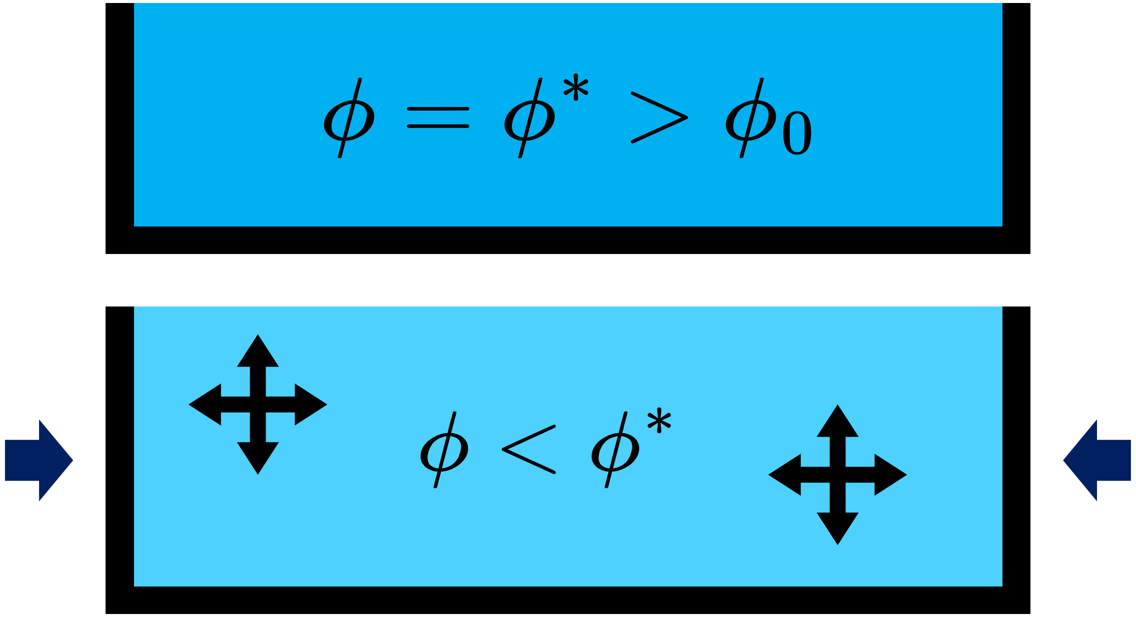
Physical setup

ϕ polymer volume fraction

ϕ_0 equilibrium (free swelling) polymer volume fraction

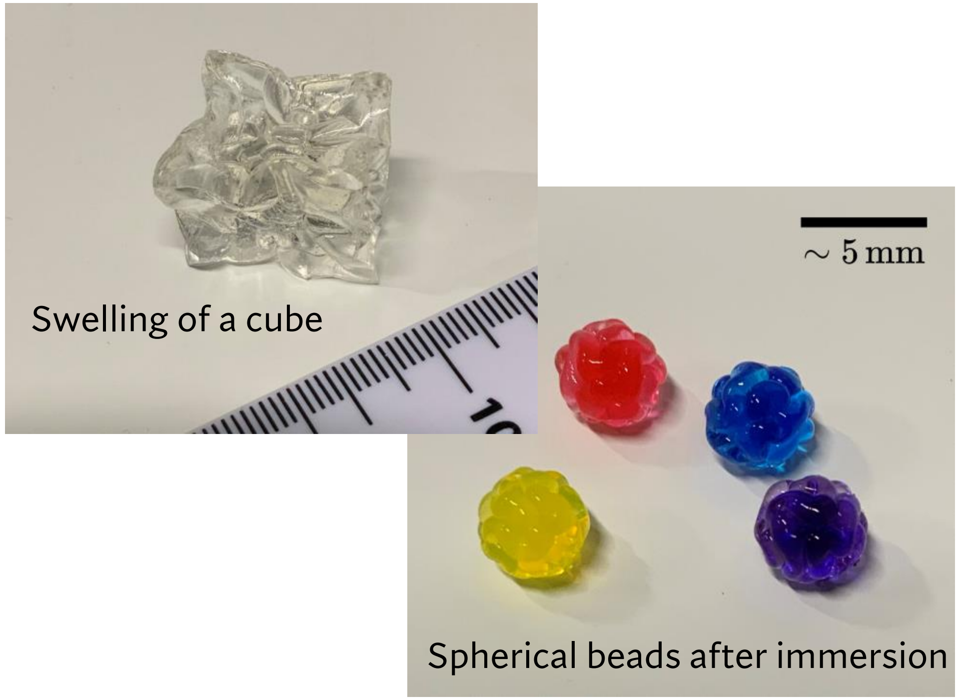
✓ Mechanically-confined swelling



$$\phi = \phi^* > \phi_0$$

$$\phi < \phi^*$$

✗ Confinement from differential swelling



Swelling of a cube

Spherical beads after immersion

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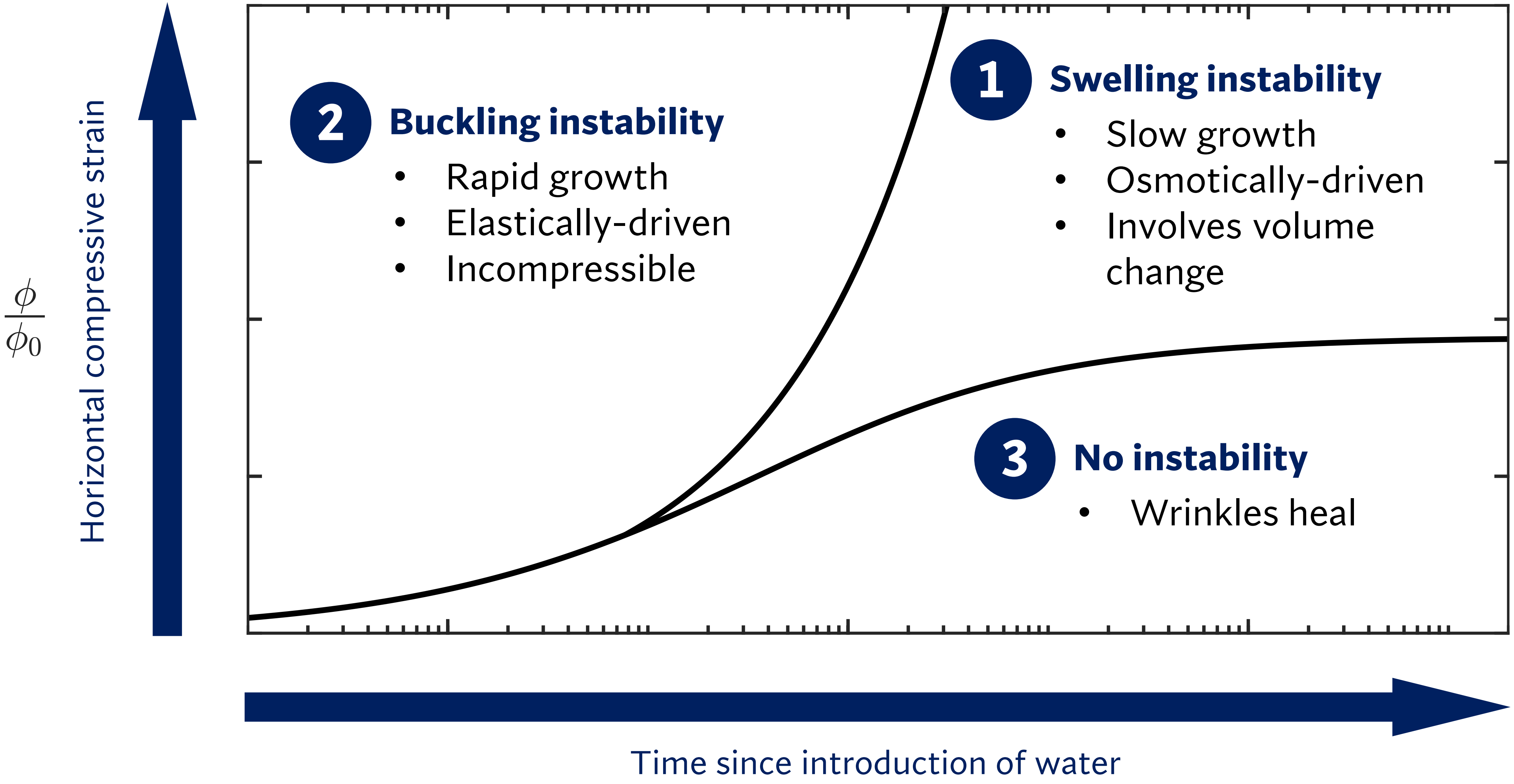


1 Swelling instability

- Slow growth
- Osmotically-driven
- Involves volume change

2 Buckling instability

- Rapid growth
- Elastically-driven
- Incompressible

3 No instability

- Wrinkles heal

Poromechanical modelling

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

Displacement-strain relations

$$\mathbf{e} = \frac{1}{2}\left[\boldsymbol{\nabla}\boldsymbol{\xi} + \boldsymbol{\nabla}\boldsymbol{\xi}^{\mathrm{T}}\right]$$

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

Deviatoric strain tensor

$$\boldsymbol{\nabla}\cdot\boldsymbol{\xi} = 2\left[1 - \left(\frac{\phi}{\phi_0}\right)^{1/2}\right]$$

Constitutive relation

Shear modulus
Assume constant

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

Pervadic (pore) pressure

Osmotic pressure
Assume linear, $\Pi = K(\phi - \phi_0)/\phi_0$

Transport equation

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

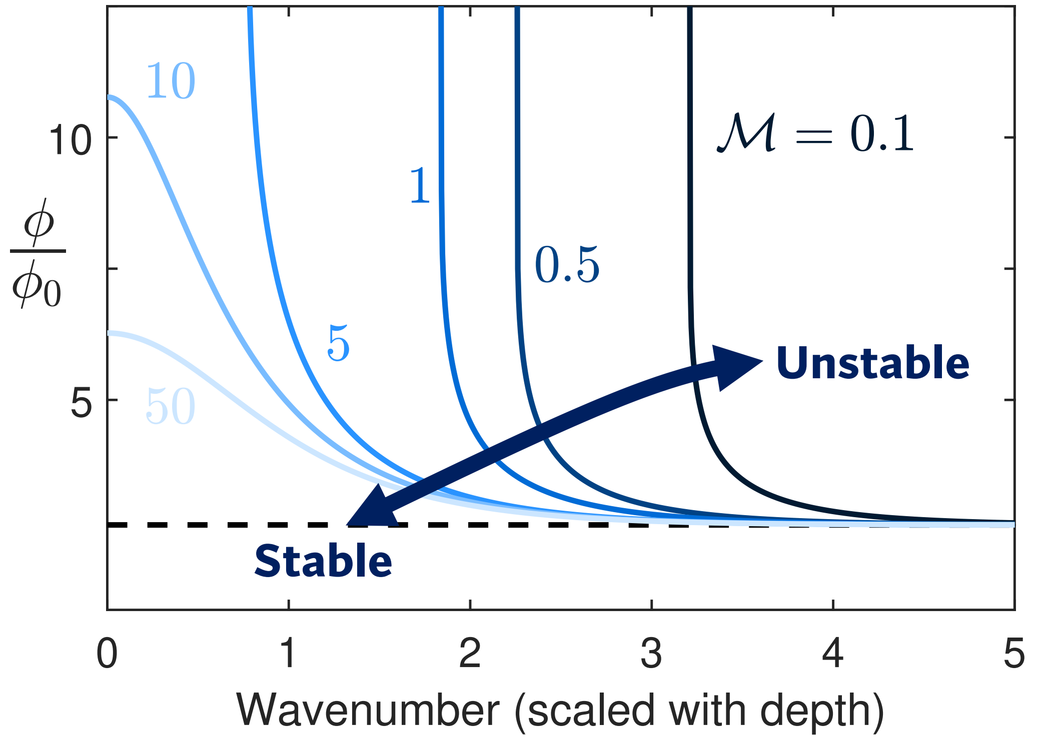
Advect with total flux

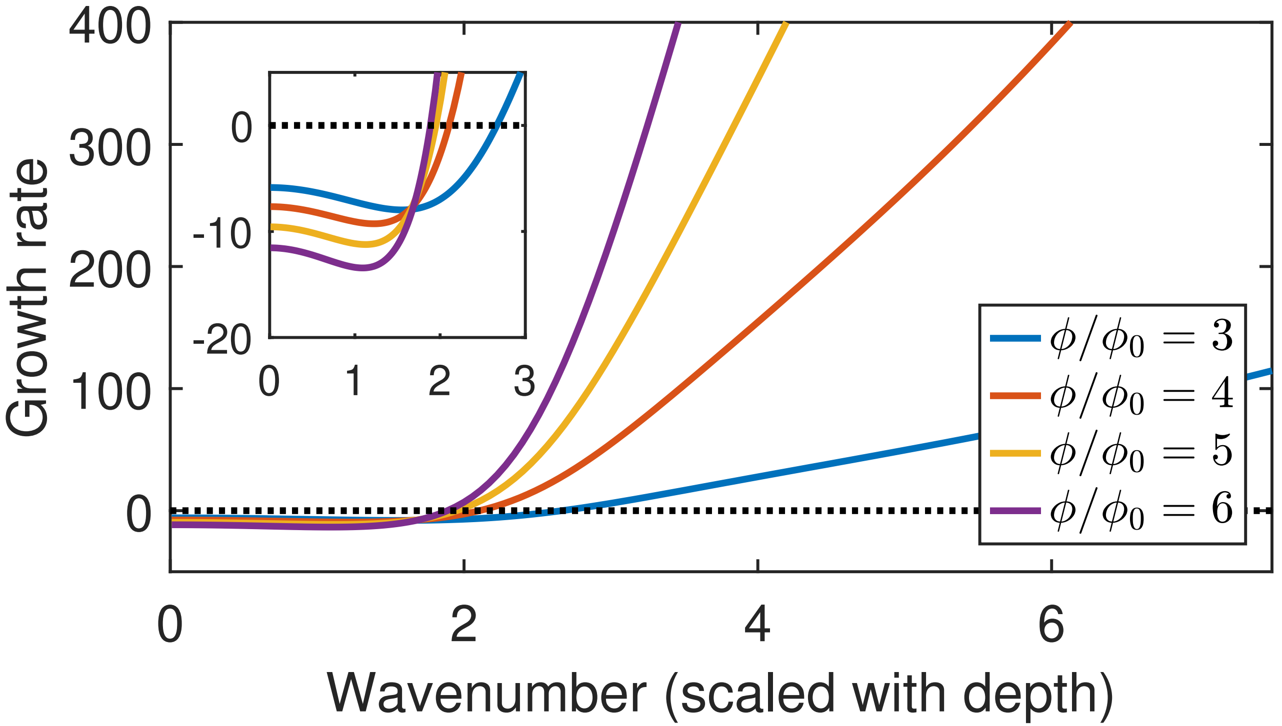
Coefficient from Darcy's law
Permeability over viscosity

- Continuity of normal and tangential stress
- Fixed base
- Continuity of pore pressure

Boundary conditions

Swelling instability





- Anchoring from the base stabilises long wavelength ripples
- Growth is faster with more compressive strain
- Criterion for instability is weaker with a greater relative shear modulus $\mathcal{M} = \mu_s/K$

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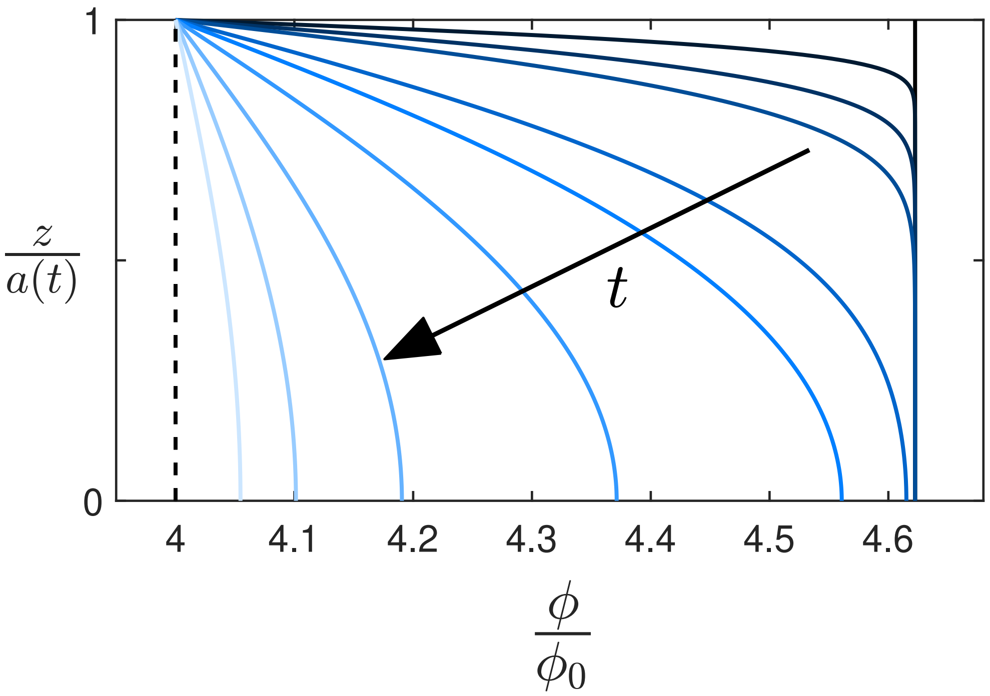
Confined swelling base state



- The interface immediately swells to its equilibrium polymer fraction
- Water diffuses into the bulk to swell the rest of the layer
- Final steady state reached with uniform polymer fraction
- In transient state, there is a swollen boundary layer of thickness δ

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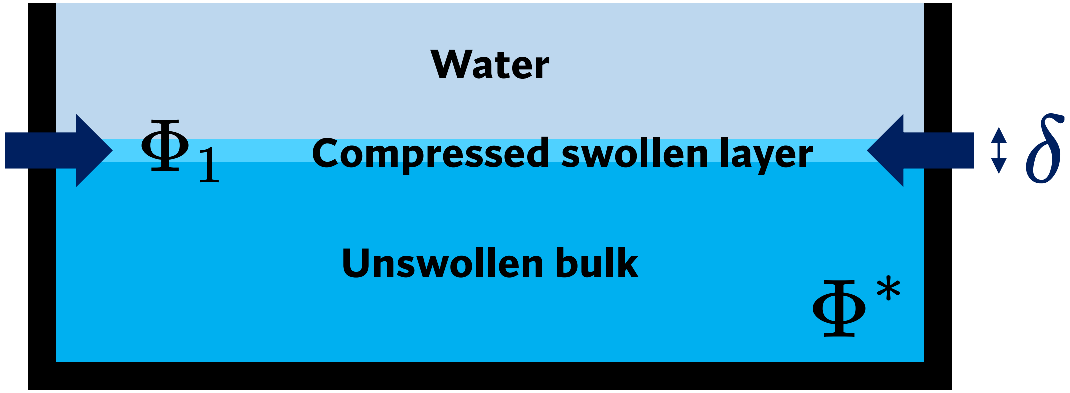
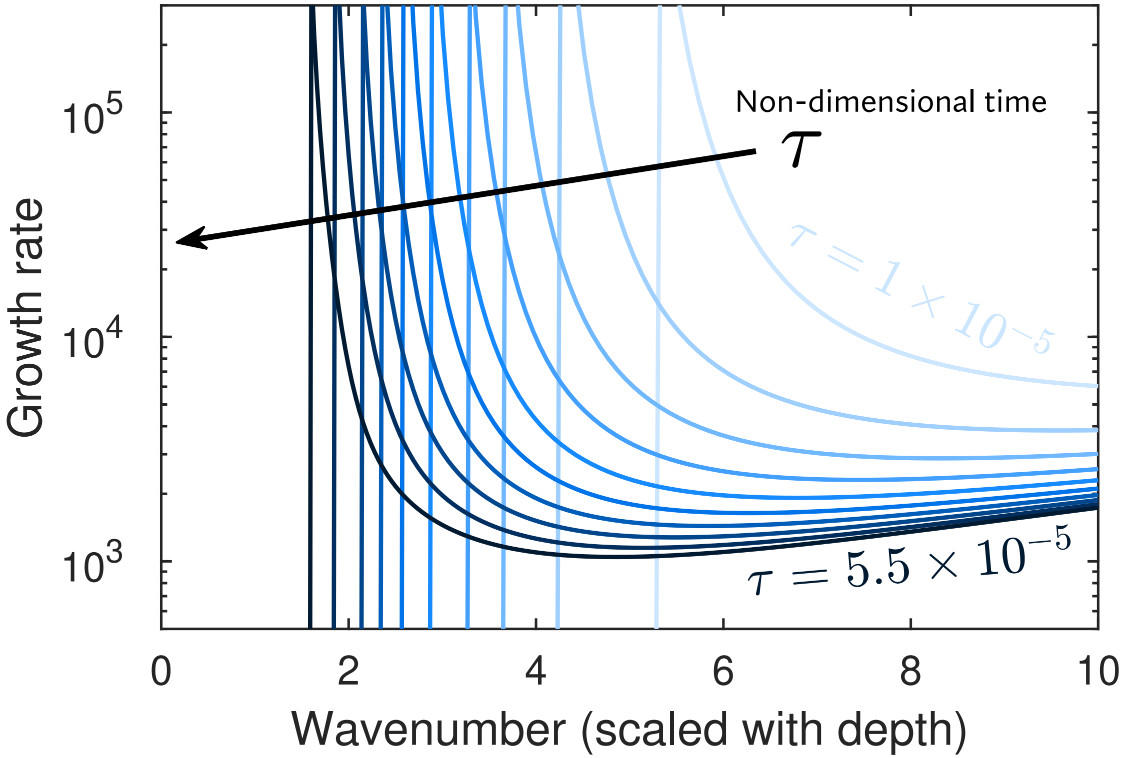
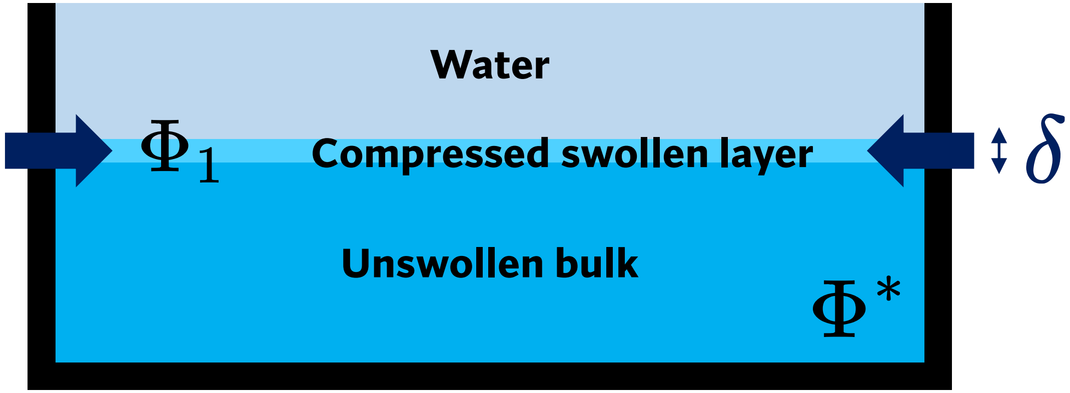
The transient state



- There’s an apparent peak in growth rates at a finite wavenumber
- Fast growth – different mechanism
- Approach late-time limit
- Is this an elastic buckle?



The transient state

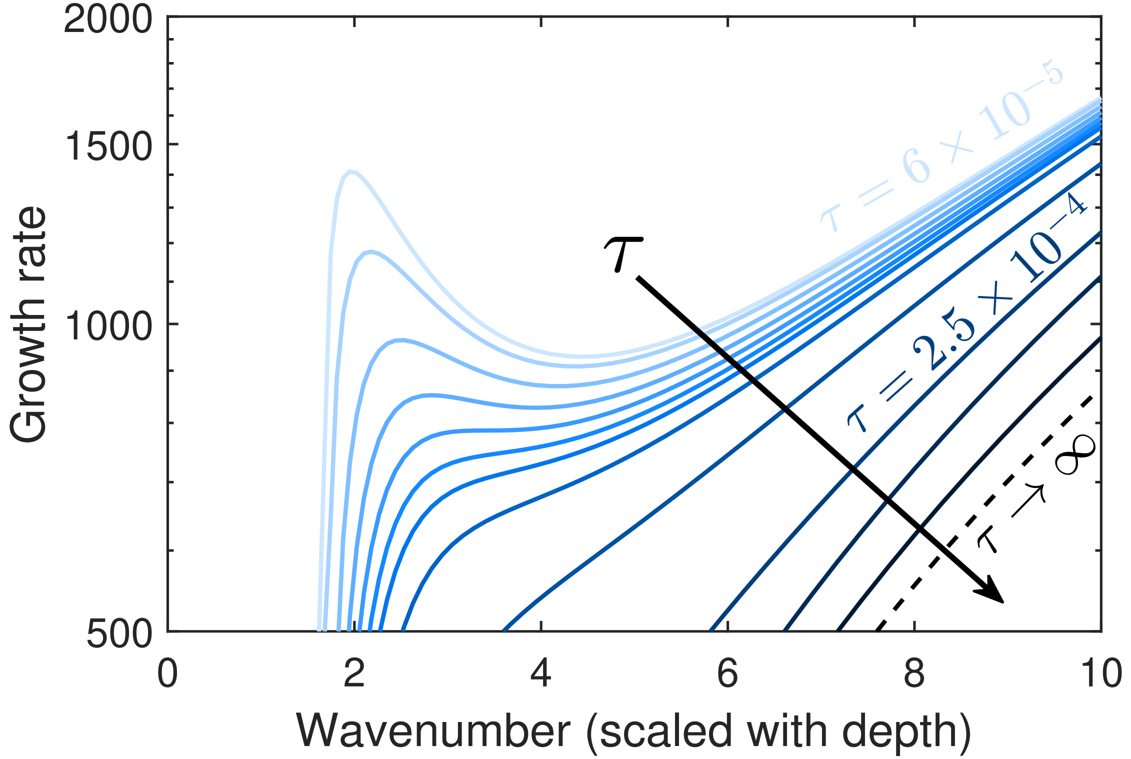


Water

Φ_1 **Compressed swollen layer** δ

Unswollen bulk

Φ^*



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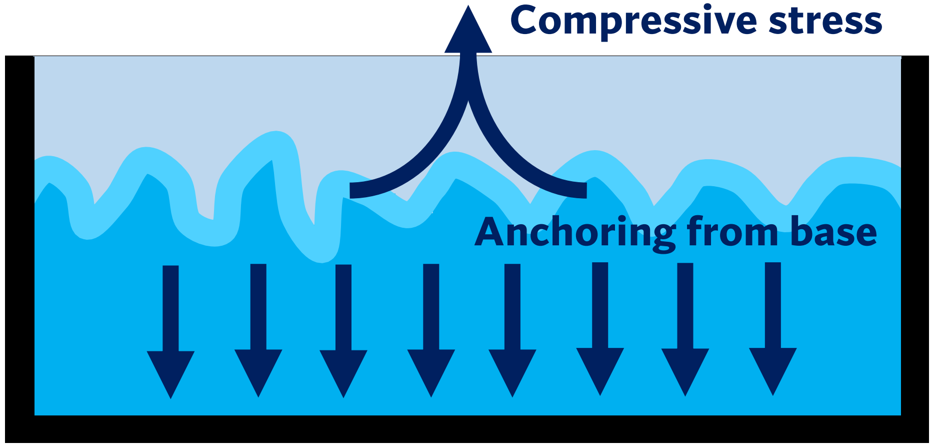
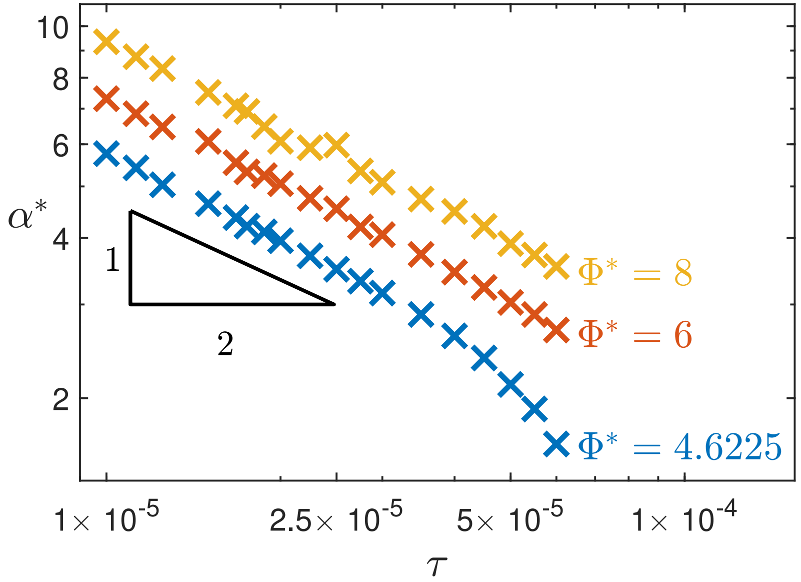
Buckling instability





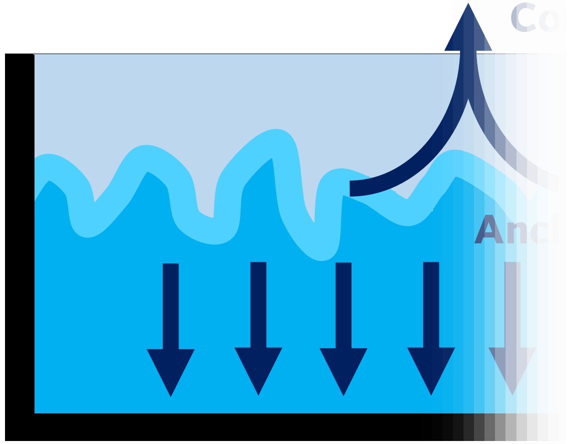
- View the base as an elastic incompressible material, bonded to a thin elastic swollen layer
- Classical plate theory gives a balance between stresses to select a finite wavenumber α^* for wrinkles

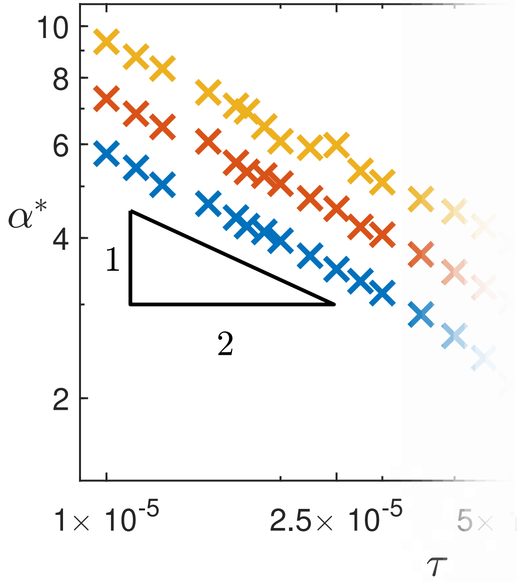
$\Phi = \phi/\phi_0$
$\Phi_y =$ interfacial polymer fraction gradient

$$\frac{E\delta^3}{12\left(1-\nu^2\right)}\alpha^4 - 4\mathcal{M}\left(\Phi^{*1/2} - \Phi_1^{1/2}\right)\delta\alpha^2 = -\left[\left(1+\mathcal{M}\Phi^{*-1/2}\right)\Phi_y + \frac{2\mathcal{M}\alpha}{\tanh\left[\alpha(1-\delta)\right]}\right]\cos\left(\alpha x\right).$$

- The solution here gives the wavenumber seen at early times

Buckling instability





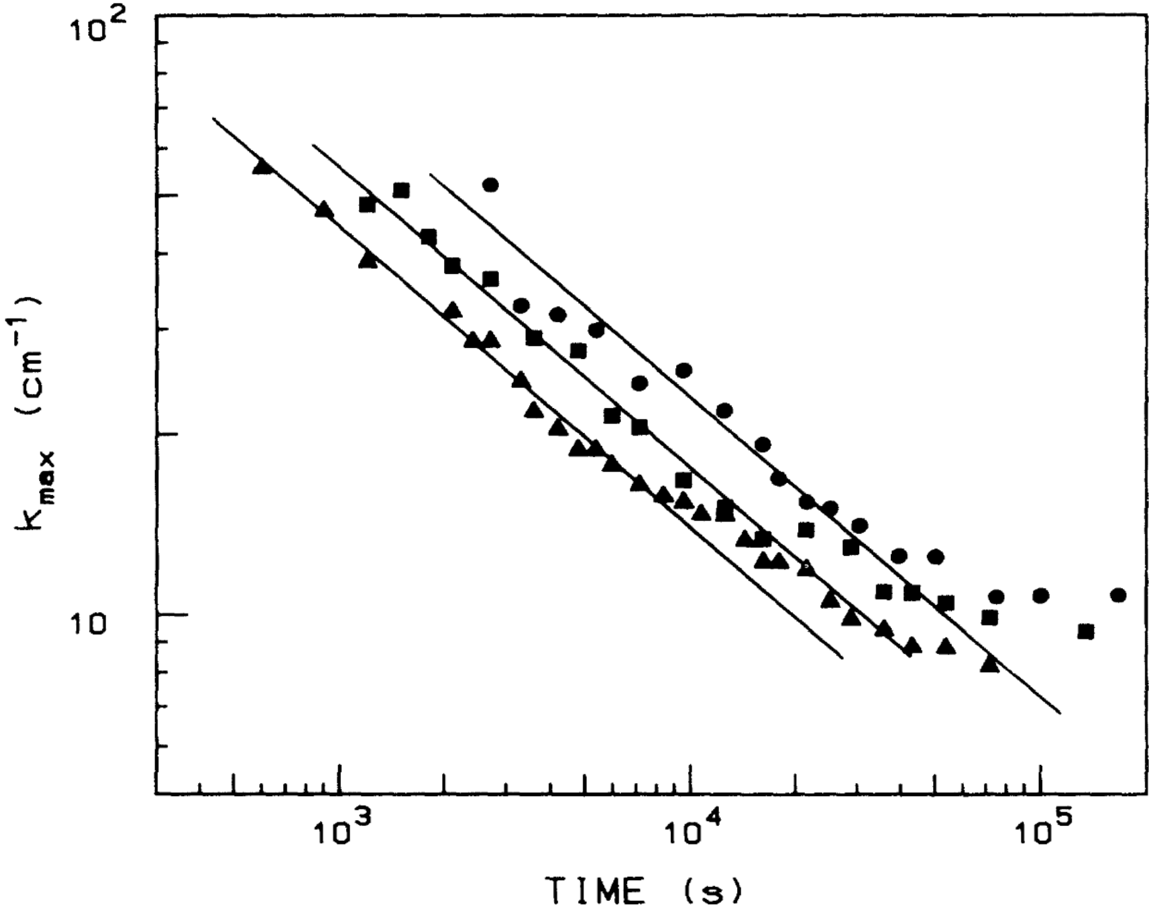


FIG. 4. Temporal change in $k_{\max}$ for the gels I-1–I-3. (●) I-1; (■) I-2; (▲) I-3. All the solid lines have a slope of $-\frac{1}{2}$.

incompressible
elastic swollen

a balance
a finite
es

$\Phi = \phi/\phi_0$
$\Phi_y =$ interfacial polymer fraction gradient

$=$

$$\frac{2\mathcal{M}\alpha}{\tanh\left[\alpha(1-\delta)\right]}\Bigg]\cos\left(\alpha x\right).$$

wavenumber

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

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Healing of instabilities

- There is no longer a solution to the equation for buckling if the thickness of the swollen layer is above a critical value

$$\delta_c = \frac{\Phi^* + \mathcal{M}{\Phi^*}^{1/2}}{2\mathcal{M}}\left(1 - \frac{\Phi_1}{\Phi^*}\right)$$

- Here, the effect of the base is too strong and buckling is suppressed
- Thus, we can no longer have a buckling instability
- If we swell to a low enough polymer fraction, we no longer see a swelling instability either, and the wrinkles on the surface 'heal'

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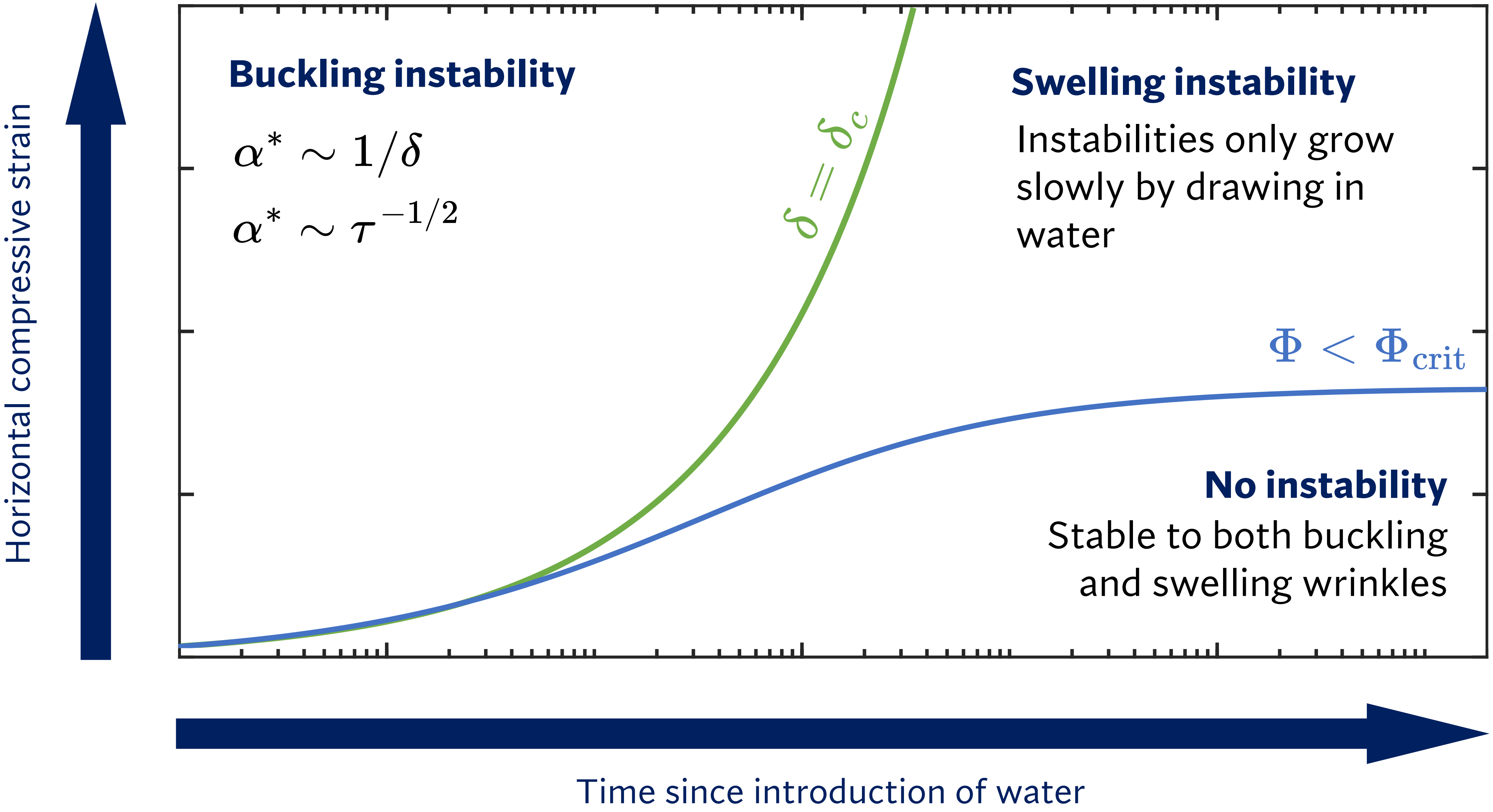

Buckling instability

$$\alpha^* \sim 1/\delta$$

$$\alpha^* \sim \tau^{-1/2}$$

Swelling instability

Instabilities only grow slowly by drawing in water

No instability

Stable to both buckling and swelling wrinkles

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