北京大學
PEKING UNIVERSITY

软物质分子间作用力和表面力研究

Intermolecular and surface forces on elastic solids

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Outline

I. **Background: van der Waals forces**

II. A saddle-node bifurcation problem

III. An elastic slab

IV. Effect of surface tension

V. Effect of approach velocity

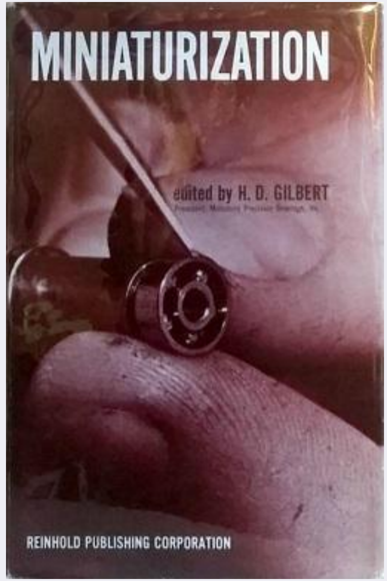
VI. Conclusions

Van der Waals forces

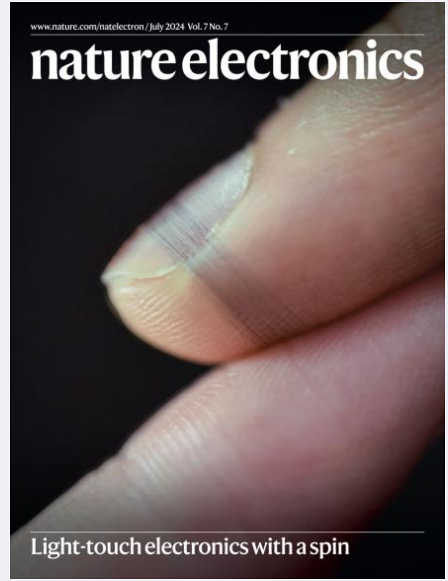
1959

Richard Feynman @ Annual APS meeting
"There's Plenty of Room at the Bottom"

1961



2024



"..As we go down in size, there are a number of interesting problems that arise...There is the problem that <u>materials stick together by the molecular (van der Waals) attractions</u>..."

Theory and experiments before 1950s

Equation of states

van der Waals (1873)
Equation of state for gases and liquids

Jones (1924)
Molecular fields
$\frac{A_m}{r^m} - \frac{B_n}{r^n}$

Molecular systems

Keesom (1915)
Permanent dipoles

Debye (1920)
Permanent-induced dipoles

London (1930)
Dispersion force
$\sim \frac{C}{r^6}$

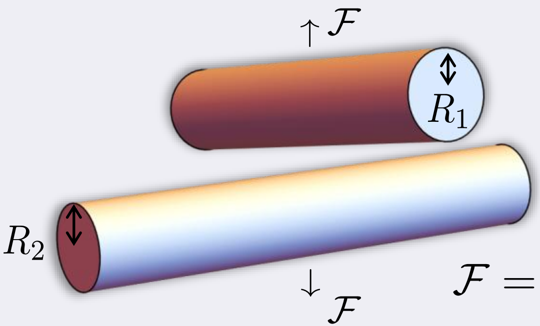
Condensed systems

Hamaker (1937)
Sphere–Body
$\sim \frac{AR_s}{r^2}$

Casimir (1948)
Retarded effect
$\sim \frac{BR_s}{r^3}$

Lifshitz (1955)
Macroscopic theory of van der Waals forces

Tomlinson (1928)

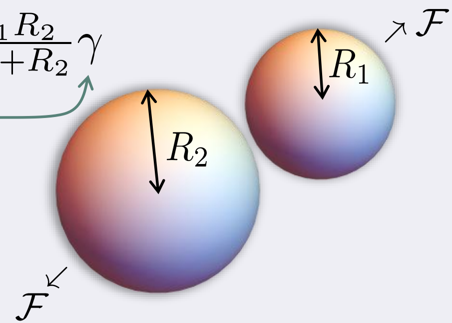


$$\mathcal{F} = 2\pi\sqrt{R_1 R_2}\gamma$$

Bradley (1932)

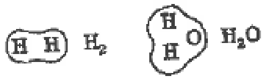
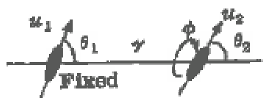
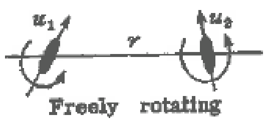
$$\mathcal{F} = 2\pi\frac{R_1 R_2}{R_1 + R_2}\gamma$$

vdW strength (not behavior!)



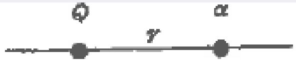
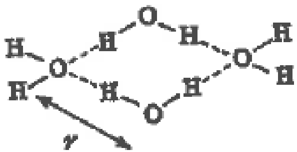
Intermolecular Interactions

Table 2.2 Common Types of Interactions and their Pair-Potentials $w(r)$ between Two Atoms, Ions, or Small Molecules in a Vacuum ($\varepsilon = 1$)[a]

Type of interaction		Interaction energy $w(r)$
Covalent, metallic		Complicated, short range
Charge–charge		$+Q_1Q_2/4\pi\varepsilon_0 r$ (Coulomb energy)
Charge–dipole		$-Qu\cos\theta/4\pi\varepsilon_0 r^2$
		$-Q^2u^2/6(4\pi\varepsilon_0)^2kTr^4$
Dipole–dipole		$-u_1u_2[2\cos\theta_1\cos\theta_2 - \sin\theta_1\sin\theta_2\cos\phi]/4\pi\varepsilon_0 r^3$
		$-u_1^2u_2^2/3(4\pi\varepsilon_0)^2kTr^6$ (Keesom energy)

Israelachvili Intermolecular and Surface Forces, 3rd ed.

Intermolecular Interactions (cont'd)

Charge–non-polar		$-Q^2\alpha/2(4\pi\varepsilon_0)^2 r^4$
Dipole–non-polar		$-u^2\alpha(1+3\cos^2\theta)/2(4\pi\varepsilon_0)^2 r^6$
		$-u^2\alpha/(4\pi\varepsilon_0)^2 r^6$ (Debye energy)
Two non-polar molecules		$-\frac{3}{4}\frac{h\nu\alpha^2}{(4\pi\varepsilon_0)^2 r^6}$ (London dispersion energy)
Hydrogen bond		Complicated, short range, energy roughly proportional to $-1/r^2$

[a] $w(r)$ is the interaction free energy or pair-potential (in J); Q, electric charge (C); u, electric dipole moment (C m); α, electric polarizibility ($C^2\,m^2\,J^{-1}$); r, distance between the centers of the interacting atoms or molecules (m); k, Boltzmann constant (1.381×10^{-23} J K^{-1}); T, absolute temperature (K); h, Planck's constant (6.626×10^{-34} J s); ν, electronic absorption (ionization) frequency (s^{-1}); ε_0, dielectric permittivity of free space ($8.854 \times 10^{-12}\ C^2\,J^{-1}\,m^{-1}$). The force $F(r)$ is obtained by differentiating the energy $w(r)$ with respect to distance r: $F = -dw/dr$. The stabilizing repulsive "Pauli Exclusion" interactions (not shown) usually follow an exponential function $w(r) \propto \exp(-r/r_0)$, but for simplicity they are usually modeled as power laws: $w(r) \propto +1/r^n$ (where n = 9–12).

Israelachvili Intermolecular and Surface Forces, 3rd ed.

Typical Molecular and Related Length Scales

Atom, molecule or bond	Size	Energy
Argon	0.40 nm	
H20	0.28 nm	
Ethanol	0.44 nm	
H-H	0.074 nm	432 kJ/mol
C-C	0.154 nm	346 kJ/mol
C-H	0.109 nm	411 kJ/mol
O-H	0.1 nm	459 kJ/mol
Liquid (water) mean free path	0.25 nm	Room temperature and pressure
Gas (air) mean free path	68 nm	

Mean free path is the average distance traveled by a moving particle between successive collisions. If length scale is greater than ~ 10 mean free paths, continuum mechanics is valid.

Induced Dipole-Induced Dipole Interactions Usually Most Important

Table 6.3 Induction, Orientation, and Dispersion Free Energy Contributions to the Total Van der Waals Energy in a Vacuum for Various Pairs of Molecules at 293 K

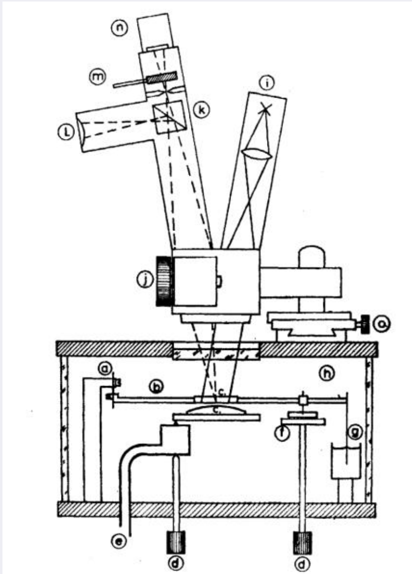
Debye Keesom Dispersion

Similar Molecules				Van der Waals Energy Coefficients C (10^{-79} J m^6)			*Total VDW Energy C_{VDW}*		
Interacting Molecules	**Electronic Polarizability $\frac{\alpha_0}{4\pi\varepsilon_0}$ ($10^{-30} m^3$)**	**Permanent Dipole Moment u (D)[a]**	**Ionization Potential $I = h\nu_I$ (eV)[b]**	**C_{ind} $\frac{2u^2\alpha_0}{(4\pi\varepsilon_0)^2}$**	**C_{orient} $\frac{u^4}{3kT(4\pi\varepsilon_0)^2}$**	**C_{disp} $\frac{3\alpha_0^2 h\nu_I}{4(4\pi\varepsilon_0)^2}$**	**Theoretical Eq. (6.17)**	**From Gas Law Eq. (6.14)**	**Dispersion Energy Contribution to Total (Theoretical) (%)**
Ne–Ne	0.39	0	21.6	0	0	4	4	4	100
CH_4–CH_4	2.60	0	12.6	0	0	102	102	101	100
HCl–HCl	2.63	1.08	12.7	6	11	106	123	157	86
HBr–HBr	3.61	0.78	11.6	4	3	182	189	207	96
HI–HI	5.44	0.38	10.4	2	0.2	370	372	350	99
CH_3Cl–CH_3Cl	4.56	1.87	11.3	32	101	282	415	509	68
NH_3–NH_3	2.26	1.47	10.2	10	38	63	111	162	57
H_2O–H_2O	1.48	1.85	12.6	10	96	33	139	175	24
Dissimilar Molecules				$\frac{u_1^2\alpha_{02} + u_2^2\alpha_{01}}{(4\pi\varepsilon_0)^2}$	$\frac{u_1^2 u_2^2}{3kT(4\pi\varepsilon_0)^2}$	$\frac{3\alpha_{01}\alpha_{02}h\nu_1\nu_2}{2(4\pi\varepsilon_0)^2(\nu_1+\nu_2)}$			
Ne–CH_4				0	0	19	19[c]	—	100
HCl–HI				7	1	197	205	—	96
H_2O–Ne				1	0	11	12	—	92
H_2O–CH_4				9	0	58	67	—	87

[a] 1 D = 3.336 × 10^{-30} Cm.
[b] 1 eV = 1.602 × 10^{-19} J.
[c] This approximate value may be compared with the ab initio calculation by Fowler et al., (1989) that gives 23 × 10^{-79} J m^6.

Rouweler and Overbeek (1971)

Dispersion forces between fused silica objects at distances between 25 and 350 nm



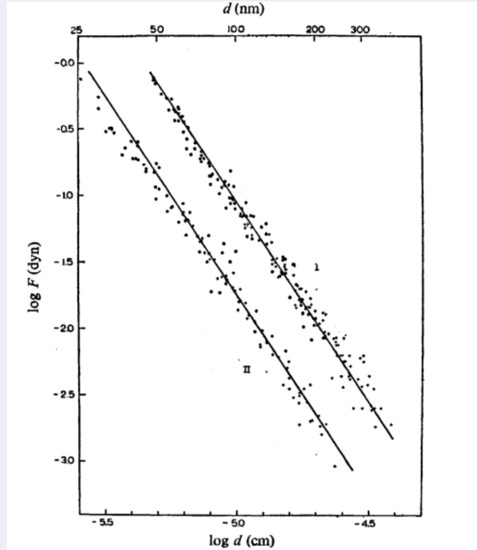
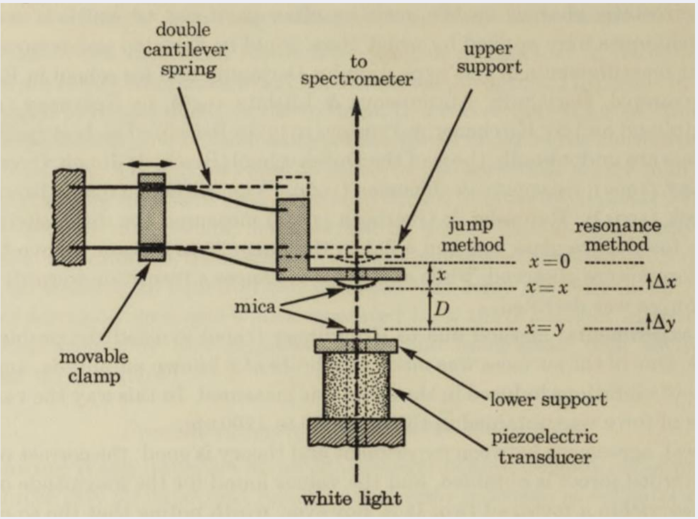
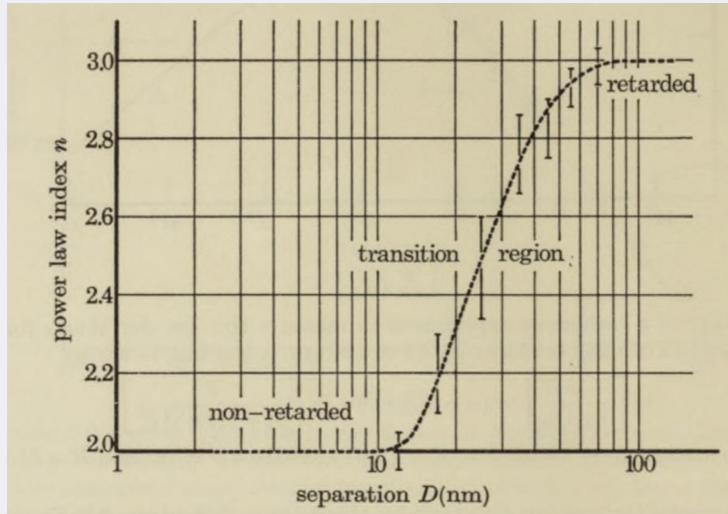


FIG. 2.—Attraction between a flat plate and a plano-convex lens of fused silica. The upper line represents the system with the 413.5 cm radius lens, 6 series of measurements are reported. The lower line represents the system with the 83.75 cm radius lens, here 5 series are reported. Crosses indicate forces measured with a standard deviation in the force of 0.5×10^{-3} dyn; for dots the standard deviation is 12×10^{-3} dyn. The lines are drawn with a slope of -3.00 corresponding to eqn (2) and (3); the standard deviation in $\overline{B}$ amounts 0.04×10^{-19} erg cm.

Israelachvili and Tabor (1972)

The measurement of van der Waals dispersion forces in the range 1.5 to 130 nm





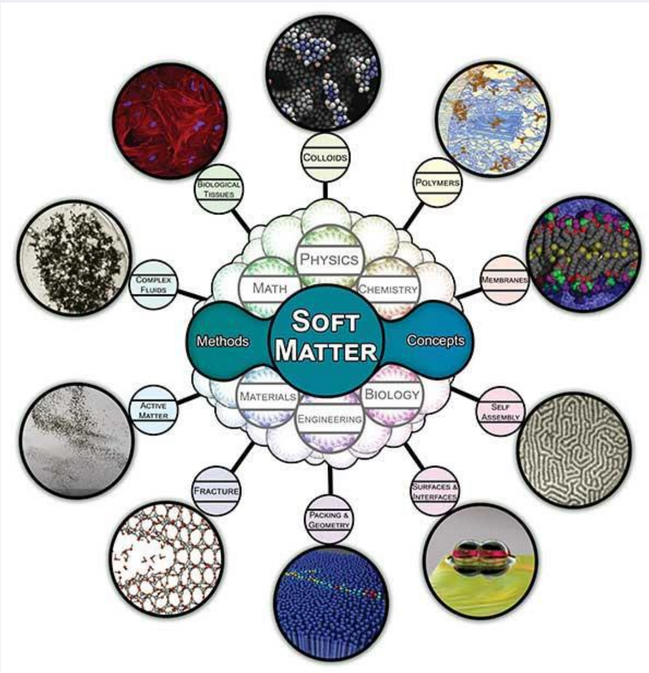
Superfluid – Helium II

Israelachvili *Intermolecular and Surface Forces*, 3rd ed.

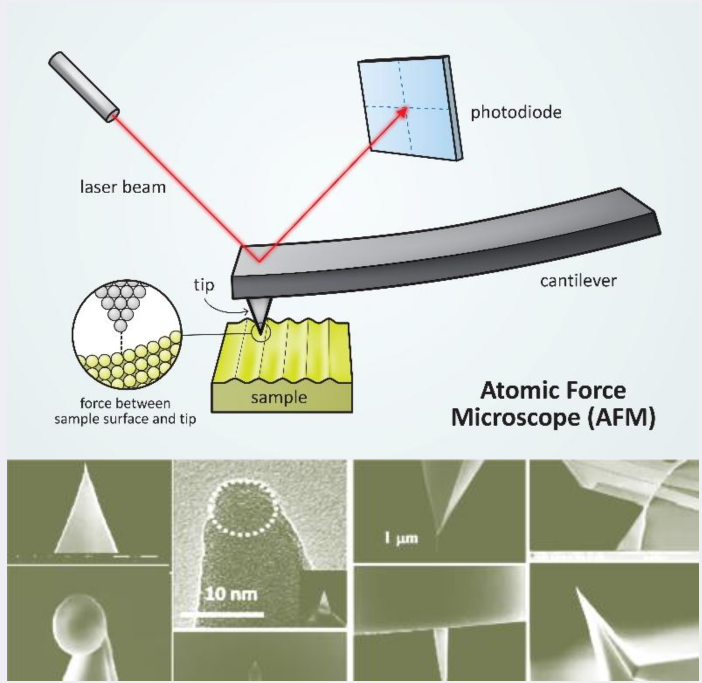
A 1963 film by Alfred Leitner demonstrating the remarkable properties of liquid helium.

Soft matter & atomic force microscopy since 90s

Soft matter is a type of matter that can be deformed or structurally altered by thermal or mechanical stress which is of similar magnitude to thermal fluctuations.

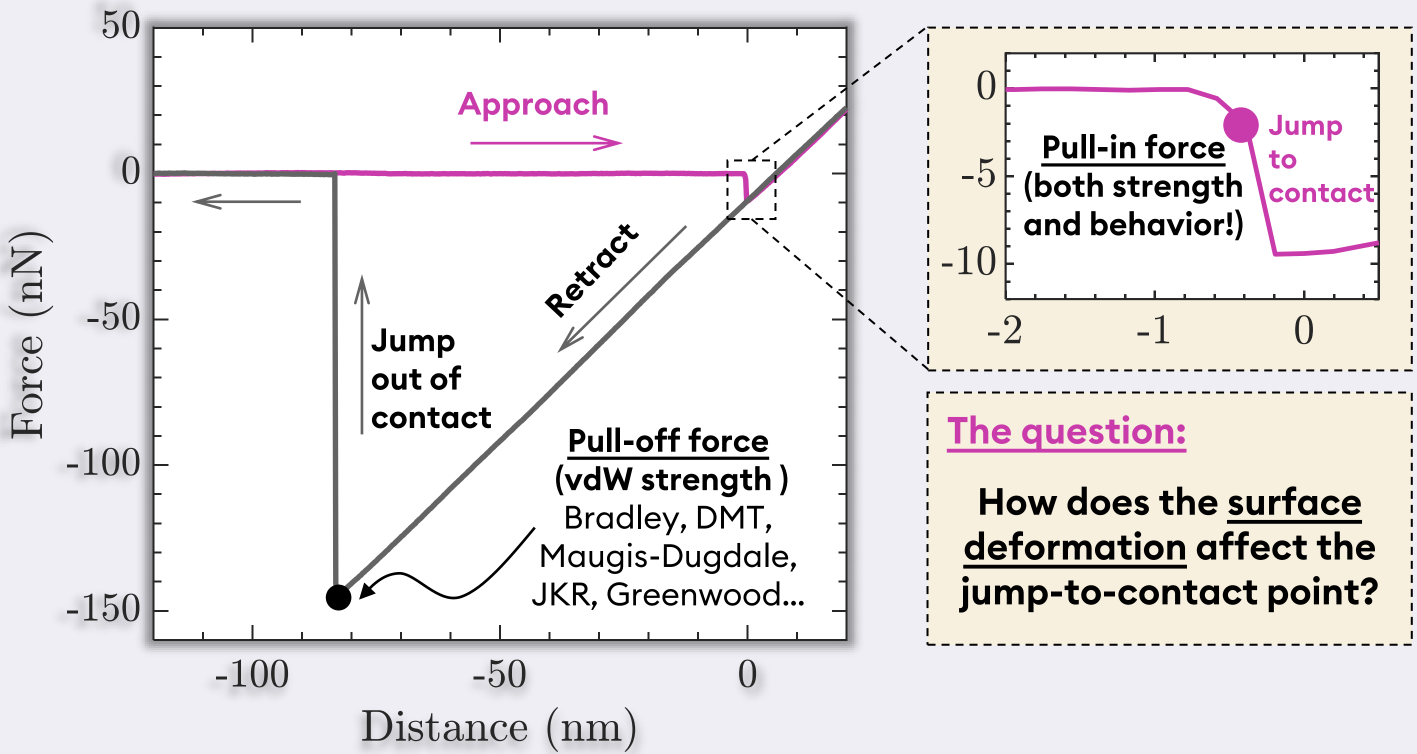


J.L. Silverberg, APS News , May 2015



Gerd Binnig (1986 Nobel Prize for Physics)

Jump-to-contact with soft matter



The question:

How does the surface deformation affect the jump-to-contact point?

Force-distance curve measured from suspended graphene sheets (Yu, Dai, unpublished)

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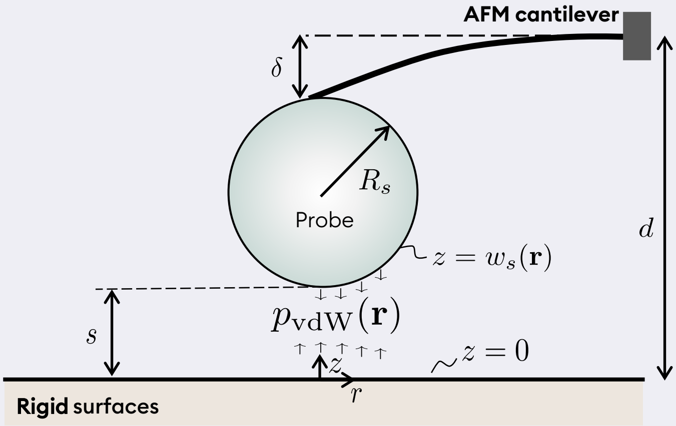
VI. Conclusions

Rigid case: Theoretical settings



- **Kinematics**

$$d = \delta + s + d_{\mathrm{ref}}$$

- **Equilibrium**

$$F = \int_S p_{\mathrm{vdW}}\, \mathrm{d}S \quad \text{(interface)}$$

$$F = k\delta \quad \text{(cantilever)}$$

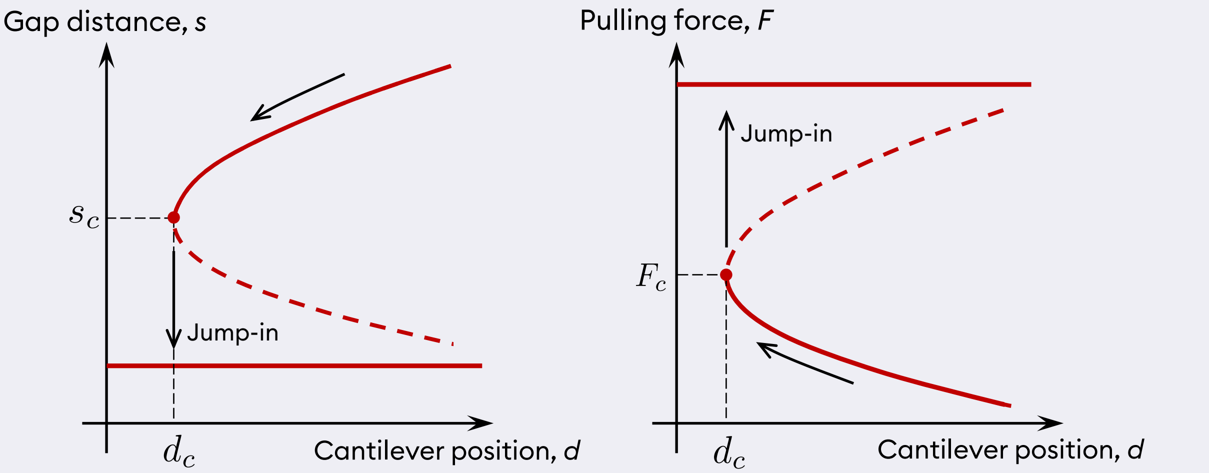
- **"Constitutive" laws**

$$p_{\mathrm{vdW}} = A/w_s^3 \quad \text{(non-retarded vdW)}$$

$$\Longrightarrow \quad F = \pi A R_s / s^2, \quad d = s + \frac{\pi A R_s}{k s^2}$$

Tabor and Winterton (1969)

Rigid case: A saddle-node bifurcation problem

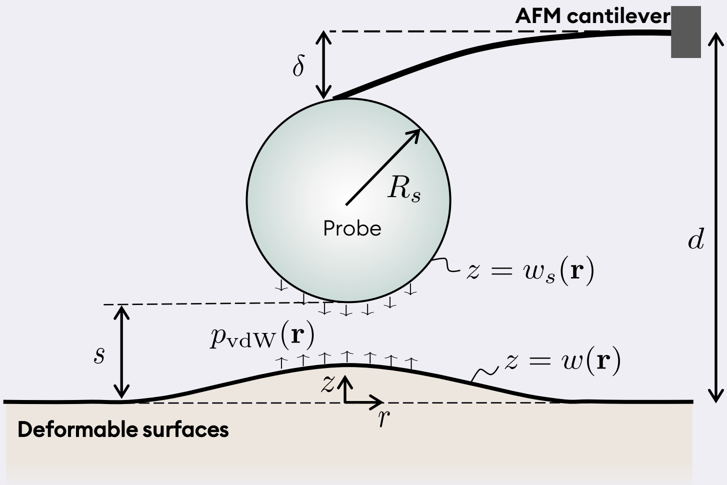


$$d = s + \frac{\pi A R_s}{k s^2}$$

$$F = \pi A R_s / s^2$$

Jump-to-contact, pull-in instability, limit-point instability, snap-through instability...

Deformable case: "Enhanced" vdW interactions



- **Kinematics**

$$d = \delta + s + d_{\mathrm{ref}}$$

- **Equilibrium**

$$F = \int_S p_{\mathrm{vdW}} \, \mathrm{d}S \quad \text{(interface)}$$

$$F = k\delta \quad \text{(cantilever)}$$

- **"Constitutive" laws**

$$p_{\mathrm{vdW}} = A / (w_s - w)^3 \quad \text{(regular vdW)}$$

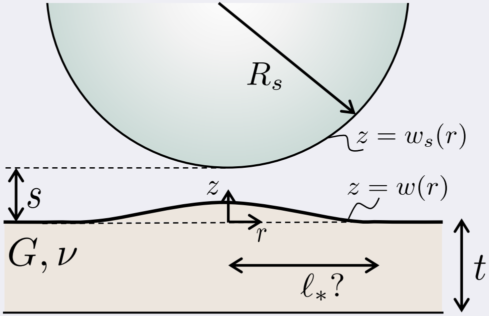
$$w(\boldsymbol{r}) = \int_{\mathbb{R}^2} p_{\mathrm{vdW}}(\boldsymbol{x}) \mathrm{G}(\boldsymbol{r} - \boldsymbol{x}) \, \mathrm{d}^2\boldsymbol{x}$$

(Continuum with linear response, e.g., linear solids, plates, shells, stretched membranes, liquid surfaces…)

For linear mechanical response, the problem remains nonlinear due to vdW interactions!

Outline

Elastic solids with a finite thickness



- **Hankel transform of order 0:**

$$\tilde{f}(\xi) = \mathcal{H}_0[f(r)] = \int_0^\infty f(r) J_0(\xi r) r \, \mathrm{d}r$$

- **Inverse Hankel transform of order 0:**

$$f(r) = \mathcal{H}_0^{-1}[\tilde{f}(\xi)] = \int_0^\infty \tilde{f}(\xi) J_0(\xi r) \xi \, \mathrm{d}\xi$$

$$\tilde{w}(\xi) = \mathcal{K}(\xi) \tilde{p}_{\mathrm{vdW}}(\xi), \quad \mathcal{K}(\xi) = \frac{1-\nu}{G\xi} \times \frac{(3-4\nu)\sinh(2\xi t) + 2\xi t}{(3-4\nu)\cosh(2\xi t) + 2(\xi t)^2 + 5 - 12\nu + 8\nu^2}$$

Hannah (1951), Li et al. (2024), Lu & Dai (nonlinear geometry)

Dimensionless parameters:

$$\mathcal{C} = \mathcal{C}(\nu)?$$

A measure of compressibility

$$\mathcal{T} = t/\ell_*$$

A measure of thickness

$$\epsilon = \epsilon(G, \nu, A, k, R_s)$$

A measure of deformability

Outline

I. Background: van der Waals forces

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- I. **Semi-infinite substrate**
- II. Thin compressible substrate
- III. Thin incompressible substrate

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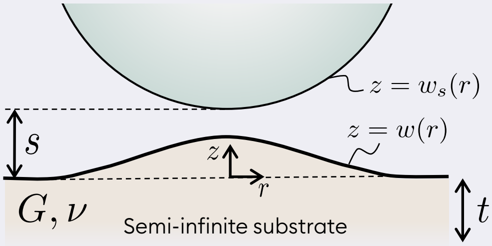
Elastic half-space



- **Characteristic lengths**

$$d = s + \frac{\pi A R_s}{k s^2} \quad \rightarrow \quad s_* = (A R_s / k)^{1/3}$$

$$1/R_s \sim s_*/\ell_*^2 \quad \rightarrow \quad \ell_* = \sqrt{s_* R_s}$$

- **Effective thickness** $\mathcal{T} := \frac{t}{\ell_*} = \frac{k^{1/6} t}{A^{1/6} R_s^{2/3}} \gg 1$

$$\tilde{w}(\xi) = \mathcal{K}(\xi) \tilde{p}_{\mathrm{vdW}}(\xi), \quad \mathcal{K}(\xi) \rightarrow \frac{1-\nu}{G} \times \frac{1}{|\xi|} \quad \text{as} \quad \mathcal{T} \gg 1$$

- **Effective compressibility** $\mathcal{C} := (1 - 2\nu)$

- **Effective deformability** $\epsilon_s := \frac{w_*}{s_*} = \frac{A \ell_*}{G s_*^4} = \frac{k^{7/6}}{G A^{1/6} R_s^{2/3}}$

Perturbation theory

- **The full form of the problem:** $P(\rho) = 1/(W_s - W)^3$

$$W_s(\rho) = S + \tfrac{1}{2}\rho^2 \quad \text{and} \quad W(\rho) = \epsilon_s \int_0^\infty \tilde{\mathcal{K}}_s(X) \left(\int_0^\infty P(\rho) J_0(X\rho)\rho \,\mathrm{d}\rho\right) J_0(X\rho) X \,\mathrm{d}X$$

- **Elastic half-space with limited deformability**

$$W(\rho) = W_0(\rho) + \epsilon_s W_1(\rho) + \epsilon_s^2 W_2(\rho) + O(\epsilon_s^3),$$

$$P(\rho) = P_0(\rho) + \epsilon_s P_1(\rho) + \epsilon_s^2 P_2(\rho) + O(\epsilon_s^3)$$

$$\Downarrow \quad \mathcal{T} \gg 1, \quad \epsilon_s \ll 1$$

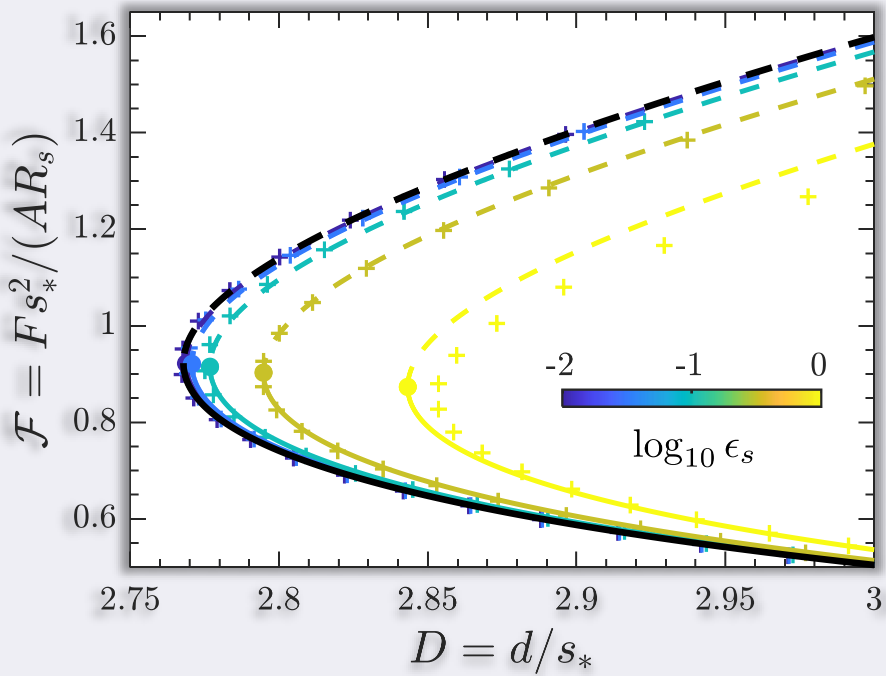
$$\underline{W_0} + \epsilon_s \left\{W_1 - \mathcal{H}^{-1}\left[\tilde{P}_0/|X|\right]\right\} + \epsilon_s^2 \left\{W_2 - \mathcal{H}^{-1}\left[\tilde{P}_1/|X|\right]\right\} + \cdots = 0$$

$$\underline{\left(P_0 - \frac{1}{W_s^3}\right)} + \epsilon_s \left(P_1 - \frac{3W_1}{W_s^4}\right) + \epsilon_s^2 \left(P_2 - \frac{6W_1^2}{W_s^5} - \frac{3W_2}{W_s^4}\right) + \cdots = 0$$

Zeroth order:
rigid case

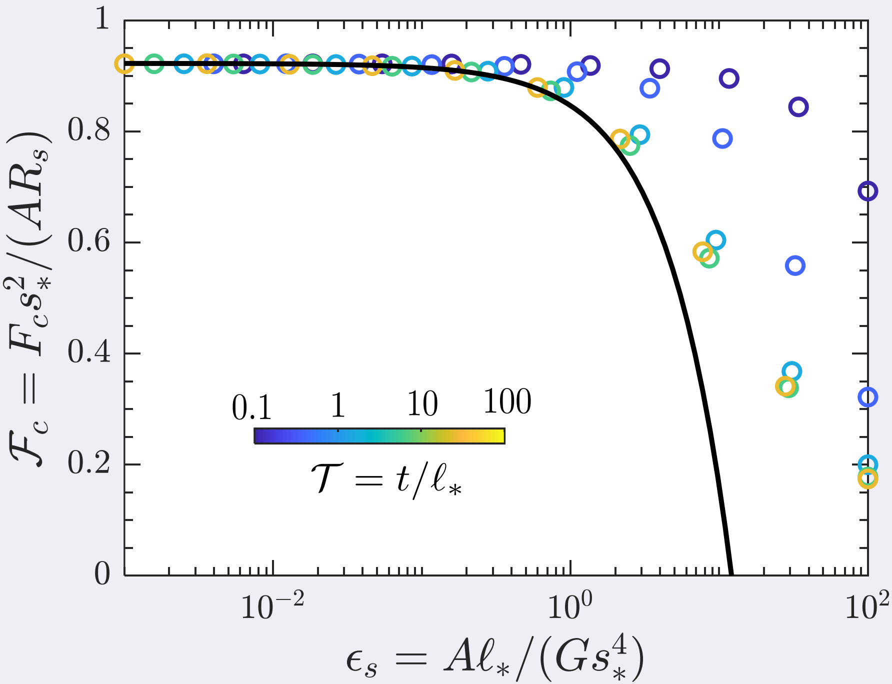
Premature saddle-node bifurcation structure

$$D = S + \frac{\pi}{S^2} + \frac{2835\,\pi^3}{16384\sqrt{2}}\frac{1-\nu}{S^{11/2}}\epsilon_s + O(\epsilon_s^2) \quad \text{and} \quad \mathcal{F} = \frac{\pi}{S^2} + \frac{2835\,\pi^3}{16384\sqrt{2}}\frac{1-\nu}{S^{11/2}}\epsilon_s + O(\epsilon_s^2)$$



Surface deformation makes the jump-to-contact occur earlier:
at a larger cantilever position, with a greater gap, and thus at smaller pulling forces.

Jump-to-contact point



$$D_c = \left(\frac{27\pi}{4}\right)^{1/3} + \frac{2835\,\pi^{7/6}}{65536\times 2^{1/3}}(1-\nu)\epsilon_s, \quad \mathcal{F}_c = \left(\frac{\pi}{4}\right)^{1/3} - \frac{4725\,\pi^{7/6}}{131072\times 2^{1/3}}(1-\nu)\epsilon_s.$$

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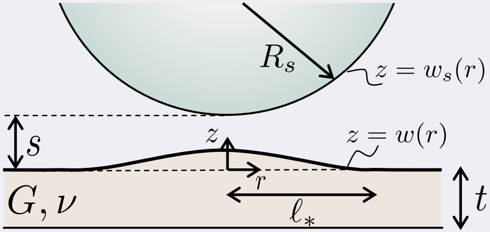
Thin compressible substrates



- **Characteristic lengths**

$$d = s + \frac{\pi A R_s}{k s^2} \quad \rightarrow \quad s_* = (A R_s / k)^{1/3}$$

$$1/R_s \sim s_*/\ell_*^2 \quad \rightarrow \quad \ell_* = \sqrt{s_* R_s}$$

- **Effective thickness** $\mathcal{T} := \frac{t}{\ell_*} = \frac{k^{1/6} t}{A^{1/6} R_s^{2/3}}$

$$\tilde{w}(\xi) = \mathcal{K}(\xi)\tilde{p}_{\mathrm{vdW}}(\xi), \quad \mathcal{K}(\xi) \rightarrow \frac{1}{G} \times \left[\frac{(1-2\nu)}{2(1-\nu)} t + \frac{\nu(4\nu-1)}{6(1-\nu)^2} \xi^2 t^3\right] \quad \text{as} \quad \mathcal{T} \ll 1$$

- **Effective compressibility** $\mathcal{C} := (1 - 2\nu)\frac{\ell_*^2}{t^2} = (1 - 2\nu)\mathcal{T}^{-2}$

- **Effective deformability** $\epsilon_c := \frac{w_*}{s_*} = \frac{(1-2\nu) t A}{2(1-\nu) G s_*^4} = \frac{(1-2\nu) t k^{4/3}}{2(1-\nu) G A^{1/3} R_s^{4/3}}$

Thin compressible substrates: $\mathcal{T} \ll 1, \quad \mathcal{C} \gg 1, \quad \epsilon_c$ arbitrary

Perturbation theory

- The full form of the problem $P(\rho) = 1/(W_s - W)^3$

$$W_s(\rho) = S + \tfrac{1}{2}\rho^2$$

$$W(\rho) = \epsilon_c \int_0^\infty \tilde{\mathcal{K}}_c(X) \left(\int_0^\infty P(\rho) J_0(X\rho)\rho\,\mathrm{d}\rho\right) J_0(X\rho) X\,\mathrm{d}X$$

- Thin compressible substrate with limited deformability

$$W(\rho) = W_0(\rho) + \epsilon_c W_1(\rho) + \epsilon_c^2 W_2(\rho) + O(\epsilon_c^3),$$

$$P(\rho) = P_0(\rho) + \epsilon_c P_1(\rho) + \epsilon_c^2 P_2(\rho) + O(\epsilon_c^3)$$

$$\Downarrow \quad \mathcal{T} \ll 1, \quad \mathcal{C} \gg 1, \quad \epsilon_c \ll 1$$

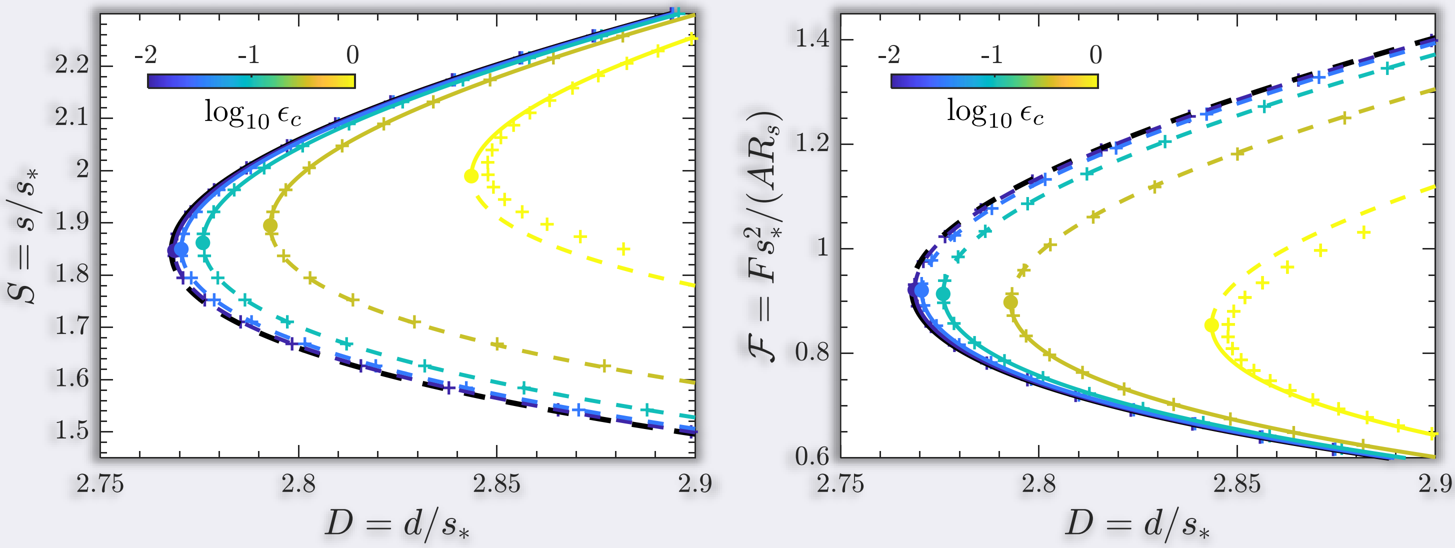
Zeroth order: rigid case

$$W_0 + \epsilon_c(W_1 - P_0) + \epsilon_c^2(W_2 - P_1) + \cdots = 0$$

$$\left(P_0 - \frac{1}{W_s^3}\right) + \epsilon_c\left(P_1 - \frac{3W_1}{W_s^4}\right) + \epsilon_c^2\left(P_2 - \frac{6W_1^2}{W_s^5} - \frac{3W_2}{W_s^4}\right) + \cdots = 0$$

Premature saddle-node bifurcation structure

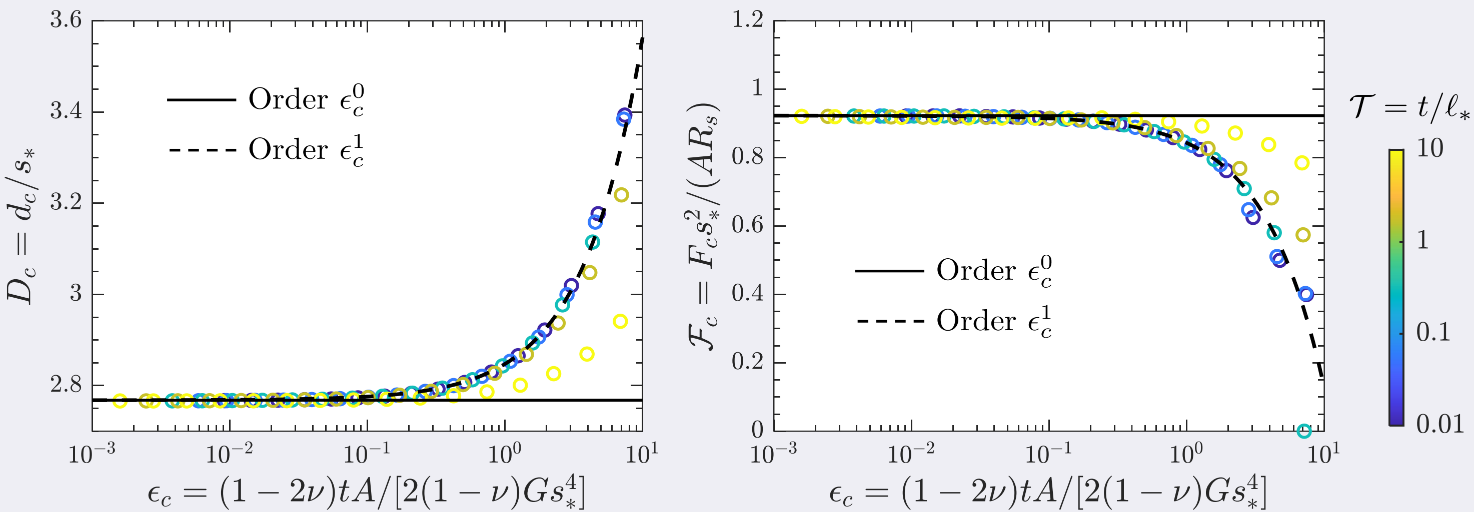
$$D = \left(S + \frac{\pi}{S^2}\right) + \frac{\pi}{S^6}\epsilon_c + \frac{3\pi}{S^{10}}\epsilon_c^2 + O(\epsilon_c^3), \quad \mathcal{F} = \frac{\pi}{S^2} + \frac{\pi}{S^6}\epsilon_c + \frac{3\pi}{S^{10}}\epsilon_c^2 + O(\epsilon_c^3)$$



Surface deformation makes the jump-to-contact occur earlier:
at a larger cantilever position, with a greater gap, and thus at smaller pulling forces.

The jump-to-contact point

$$S_c = (2\pi)^{1/3} + \frac{1}{2\pi}\epsilon_c, \quad D_c = \left(\frac{27\pi}{4}\right)^{1/3} + \frac{1}{4\pi}\epsilon_c, \quad \text{and} \quad \mathcal{F}_c = \left(\frac{\pi}{4}\right)^{1/3} - \frac{1}{4\pi}\epsilon_c$$



Perturbation solutions work surprisingly well even at moderate thickness and deformability

(high-order terms vanish – checked up to the 11th order)

Outline

I. Background: van der Waals forces

II. A saddle-node bifurcation problem

III. An elastic slab

   I. Semi-infinite substrate

   II. Thin compressible substrate

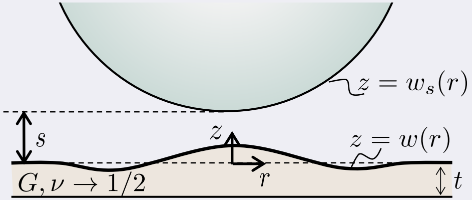
   III. Thin incompressible substrate

IV. Effect of surface tension

V. Effect of approach velocity

VI. Conclusions

Thin incompressible substrates



Thin incompressible substrate

- **Effective thickness** $\mathcal{T} := \frac{t}{\ell_*} = \frac{k^{1/6} t}{A^{1/6} R_s^{2/3}} \ll 1$

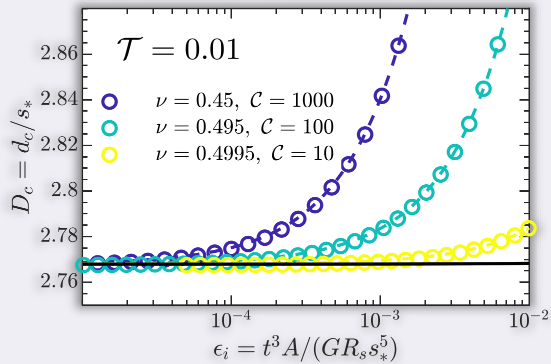
- **Effective compressibility** $\mathcal{C} := (1 - 2\nu)\frac{\ell_*^2}{t^2} = (1 - 2\nu)/\mathcal{T}^2 \to 0$

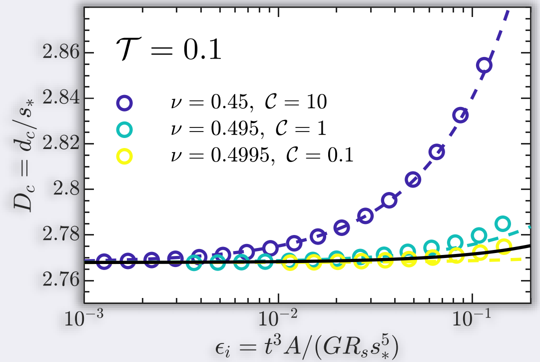
- **Effective deformability** $\epsilon_i := \frac{w_*}{s_*} = \frac{t^3 A}{G R_s s_*^5} = \frac{t^3 k^{5/3}}{G A^{2/3} R_s^{8/3}}$

2

Thin systems are easily compressible...

- Effective thickness $\mathcal{T} := \frac{t}{\ell_*} = \frac{k^{1/6}t}{A^{1/6}R_s^{2/3}} \ll 1$
- Effective compressibility $\mathcal{C} := (1-2\nu)\frac{\ell_*^2}{t^2} = (1-2\nu)\mathcal{T}^{-2} \to 0$





Compressibility is governed not only by its Poisson's ratio but also by its slenderness.
(conceptually similar to the effective Reynolds number in fluid mechanics)

Outline

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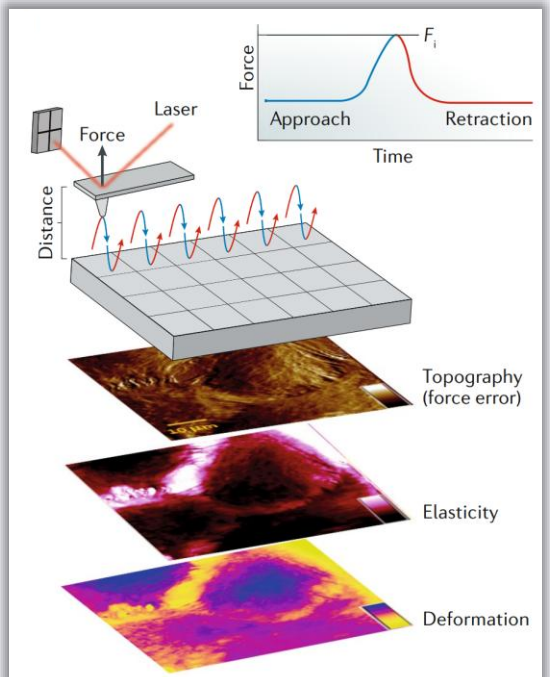
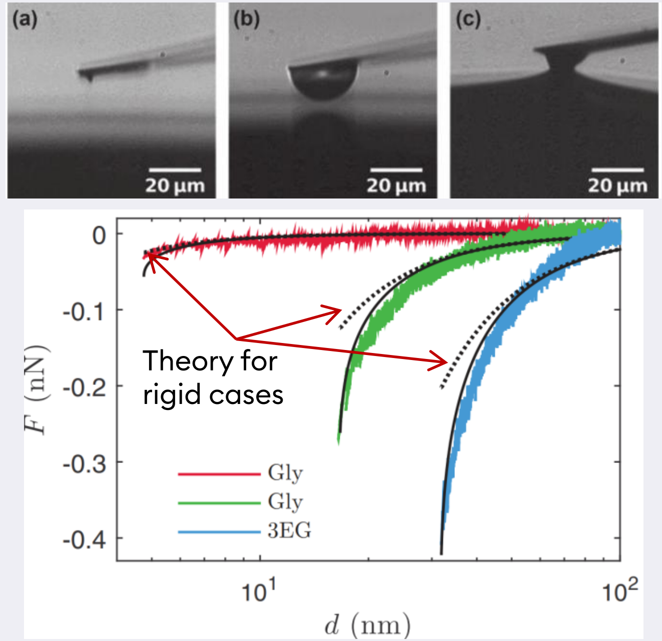
Atomic force microscopy-based mechanobiology

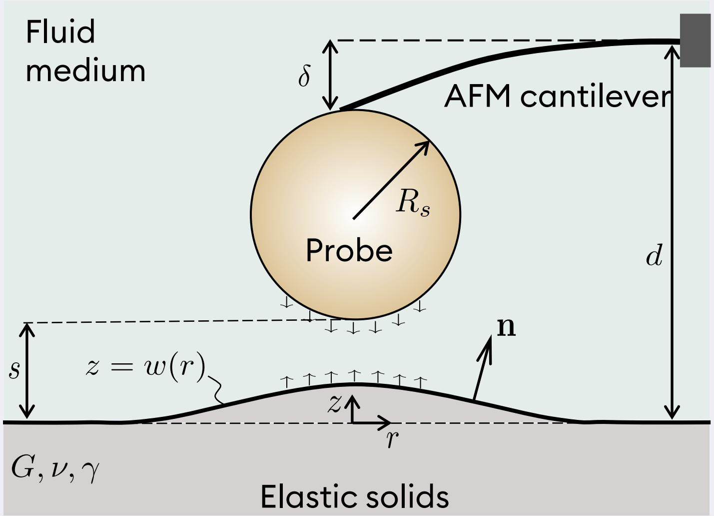


Mortagne et al. Phys. Rev. E (2017)



"AFM has emerged as a key platform enabling the simultaneous morphological and mechanical characterization of living biological systems."

Krieg et al. Nat. Rev. Phys. (2019)



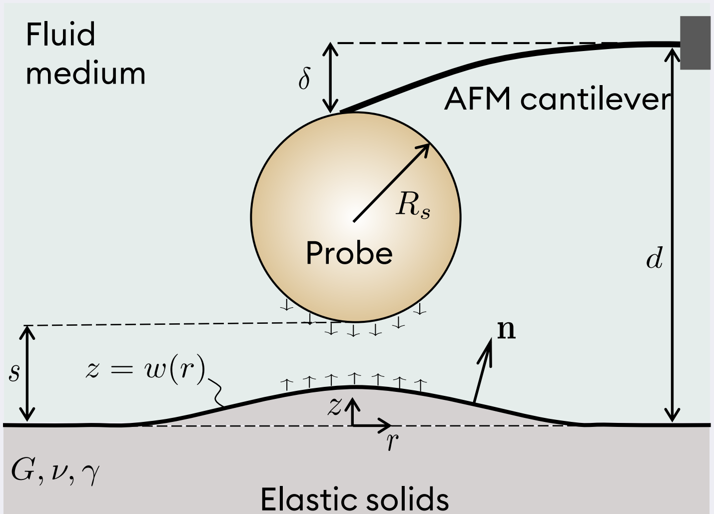
vdW interactions

Surface tension

$$\sigma_{zz}(r, z=0) = p(r) + \gamma \nabla \cdot \boldsymbol{n}$$

- **Elastic substrates** (Greenwood 1997; Feng 2000; Wu 2010; Ciavarella et al. 2017; Yu & Dai 2024)
- **Lquid surfaces** (Nan et al. 2005; Ledesma-Alonso et al. 2012; Quinn et al. 2013; Mortagne et al. 2017; Chireux et al. 2018, Beaty & JR Lister 2023)

Effective deformability of an elastocapillary surface



- Characteristic lengths (rigid)

$$s_* = (AR_s/k)^{1/3}$$

$$\ell_* = \sqrt{s_* R_s}$$

- Effect of surface tension

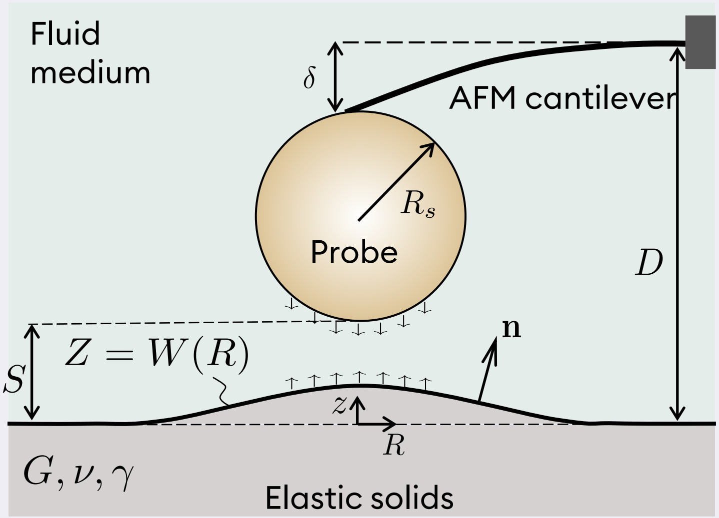
$$\Gamma := \frac{(1-\nu)\gamma}{G\ell_*}$$

$$\epsilon := \frac{A\ell_*^2}{G\ell_* s_*^4/(1-\nu) + \gamma s_*^4} = \frac{(1-\nu)AR_s^{1/2}}{(1+\Gamma)Gs_*^{7/2}}$$

Comparing the typical deformation of the substrate with surface tension induced by the typical vdW force to the characteristic sphere-substrate gap

Perturbation theory for small deformability

$$W(R) = W_0(R) + \epsilon W_1(R) + O(\epsilon^2), \quad P(R) = P_0(R) + \epsilon P_1(R) + O(\epsilon^2)$$



- Zeroth-order solution:

$$W_0 = 0$$

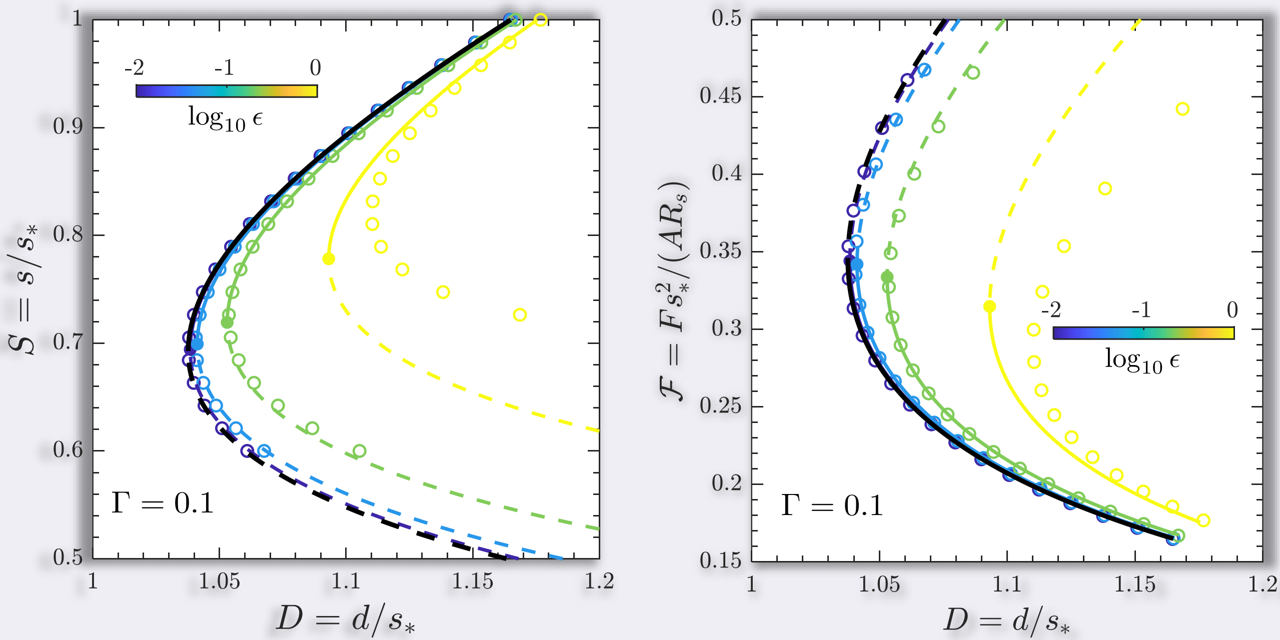
$$P_0 = \frac{4}{3\pi(R^2+2S+\alpha S^2)^3}$$

- First-order solution:

$$W_1 = \mathcal{H}_0^{-1}\left[\frac{\mathrm{K}_2\left(\sqrt{S(2+\alpha S)\Xi}\right)}{6\pi S(2+\alpha S)}\frac{(1+\Gamma)\Xi}{1+\Gamma\Xi}\right]$$

$$P_1 = \frac{8(1+\alpha S)}{\pi(R^2+2S+\alpha S^2)^4}W_1$$

Small surface tension $\left(\Gamma = \frac{(1-\nu)\gamma}{G\ell_*} \ll 1\right)$



$$W_1 = \frac{1+\Gamma}{6\pi S^{3/2}(2+\alpha S)^{3/2}W_s^2}\left[2(2W_s - R^2)\,\mathcal{E}\left(\frac{-R^2}{2S+\alpha S^2}\right) - W_s\,\mathcal{K}\left(\frac{-R^2}{2S+\alpha S^2}\right)\right] - \frac{4\Gamma}{3\pi W_s^3}$$

Large surface tension $\left(\Gamma = \frac{(1-\nu)\gamma}{G\ell_*} \gg 1\right)$

$$\Xi\widetilde{W}_1(\Xi) + \Gamma\,\Xi^2\widetilde{W}_1(\Xi) = (1+\Gamma)\,\widetilde{P}_0(\Xi)$$

This inverse Hankel transform is unbounded!?

- The "inner" solution $(\Gamma \gg 1)$

$$\frac{1}{R}\frac{\mathrm{d}}{\mathrm{d}R}\left(R\frac{\mathrm{d}W_1^{(i)}}{\mathrm{d}R}\right) + \frac{4}{3\pi(R^2+2S+\alpha S^2)^3} = 0 \qquad \Longrightarrow \qquad W_1^{(i)}(R) \propto -\ln R \quad \text{as} \quad R \to \infty$$

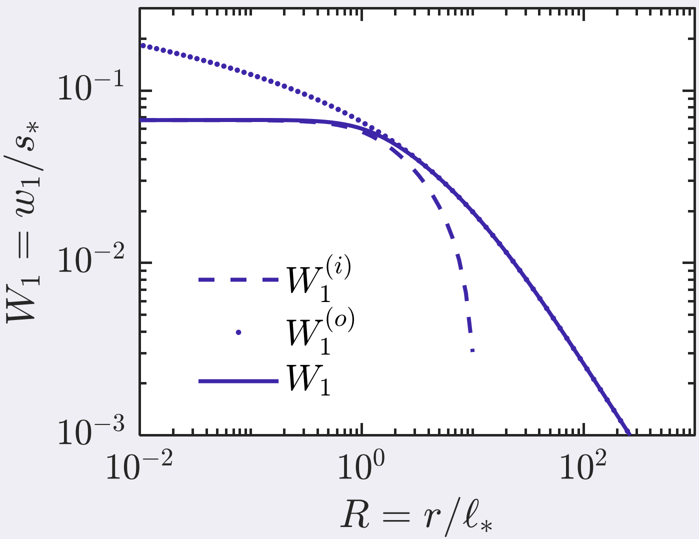
- The "outer" solution $(\hat{R} = R/\Gamma \quad \text{and} \quad \hat{\Xi} = \Gamma\Xi)$

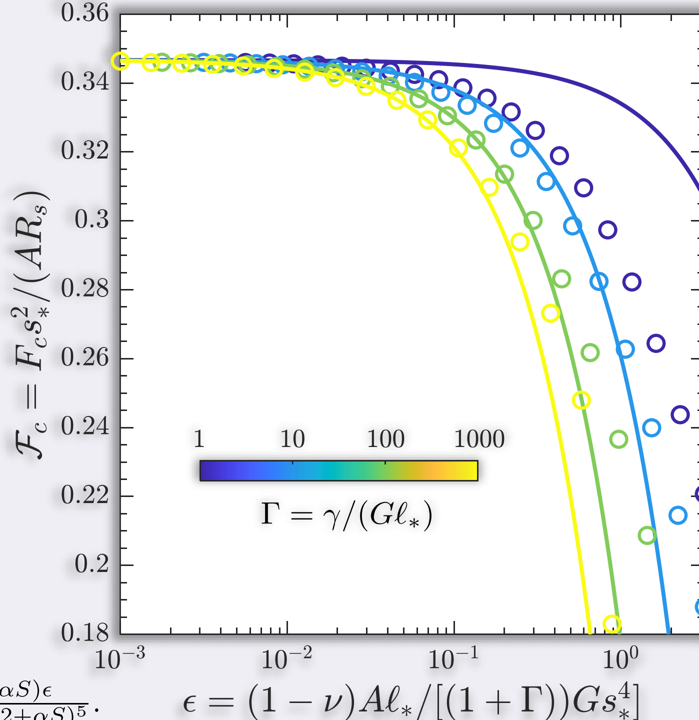
$$\hat{\Xi}\widetilde{W}_1^{(o)}(\hat{\Xi}) + \hat{\Xi}^2\widetilde{W}_1^{(o)}(\hat{\Xi}) = \frac{\hat{\Xi}^2\mathrm{K}_2\left(\sqrt{S(2+\alpha S)}\hat{\Xi}/\Gamma\right)}{6\pi S(2+\alpha S)} \qquad \Longrightarrow \qquad W_1^{(o)}(R) \propto -\ln R \quad \text{as} \quad R \to 0$$

$$\lim_{R\to\infty} W_1^{(i)} = \lim_{\hat{R}\to 0} W_1^{(o)}$$

Far-field elastic forces are required to smooth surface deformations convergently!

Jump-to-contact on a liquid-like surface





$$\mathcal{F} = \frac{2}{3S^2(2+\alpha S)^2} + \left[12\log\frac{4\Gamma^2}{2S+\alpha S^2} + 5 - 24\gamma_E + \frac{9\pi\sqrt{2S+\alpha S^2}}{2\Gamma}\right]\frac{(1+\alpha S)\epsilon}{27\pi S^5(2+\alpha S)^5}.$$

First-order correction

Outline

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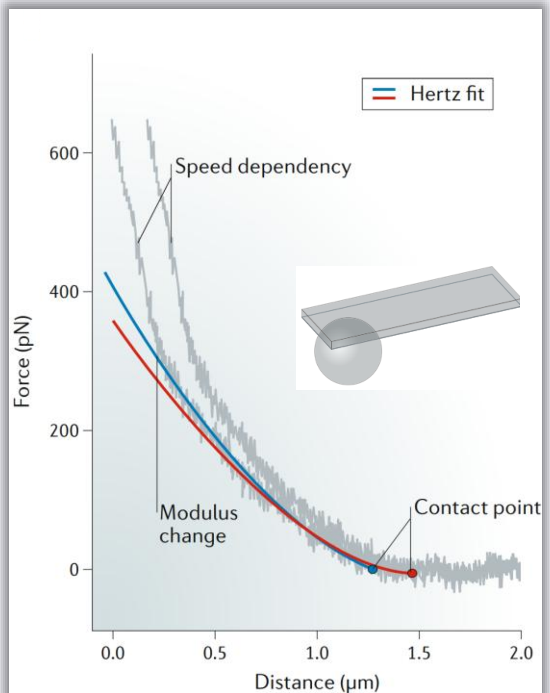
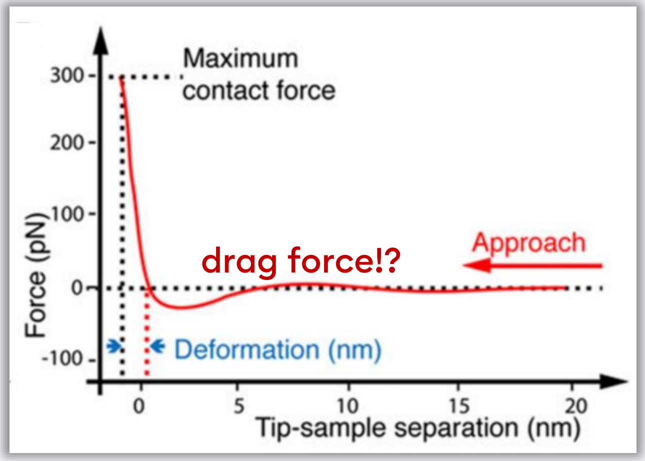
Speed-dependent behavior in indentation



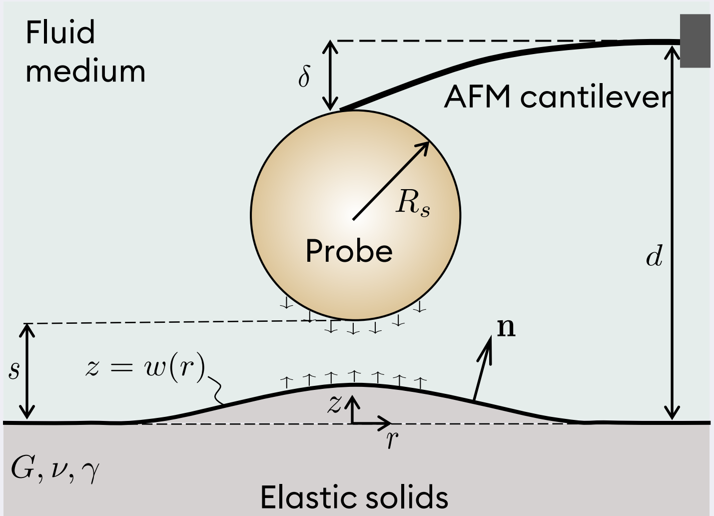
Krieg et al. Nat. Rev. Phys. (2019)



Wegmann et al. PNAS (2012)

"On the other hand, viscous contributions can be reduced if measurements take place on long timescales. However, there is a limit on the lowest achievable indentation speed, as biological systems can quickly remodel and respond to mechanical cues..."

A elasto-capillary hydrodynamic problem



Comparing the hydrodynamic pressure with the vdw pressure:

$$\lambda = \frac{36\pi\mu v_* \ell_*^2}{A}$$

- **The sphere-surface gap**

$$h(r,t) = s(t) + \frac{r^2}{2R_s} - w(r,t)$$

- **The intermolecular interaction**

$$p(r,t) = \frac{A}{6\pi\, h^3(r,t)}$$

- **The hydrodynamic pressure**

$$\frac{\partial h}{\partial t} = \nabla \cdot \left[\frac{h^3(r,t)}{12\mu}\nabla q(r,t)\right]$$

- **The elastic response**

$$\sigma_{zz}(r, z=0, t) = p(r,t) - q(r,t) + \gamma \frac{\partial^2 w(r,t)}{\partial r^2}$$

Perturbation solution

$$V(R,T) = V_0(R,T) + \epsilon V_1(R,T) + O(\epsilon^2), \quad \epsilon := \frac{A\ell_*^2}{G\ell_* s_*^4/(1-\nu)+\gamma s_*^4}$$

- Zeroth-order solution:

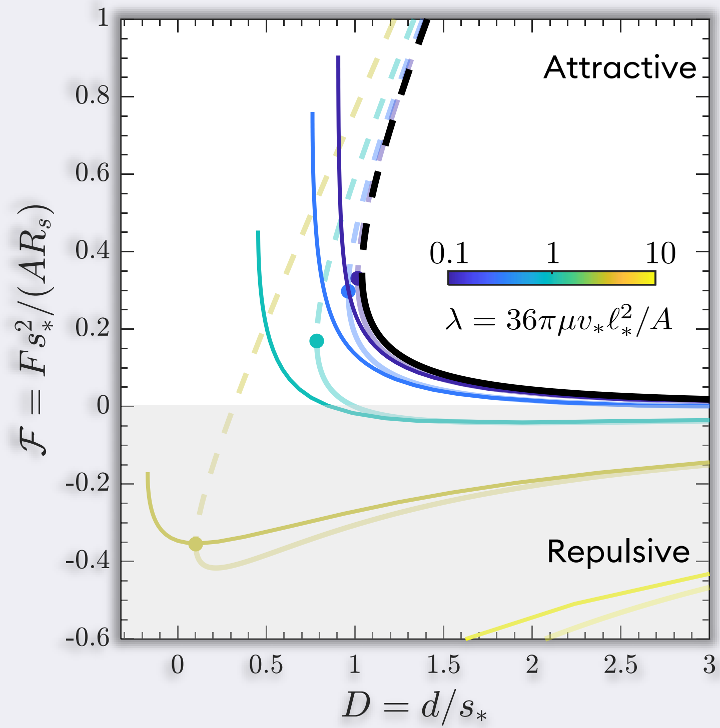
$$D(T) = S + \frac{1}{6S^2} + \lambda S\dot{S}$$

$$\mathcal{F}(T) = \frac{1+\lambda S\dot{S}}{6S^2}$$

vdW force + Reynolds force

Opaque: constant cantilever speed

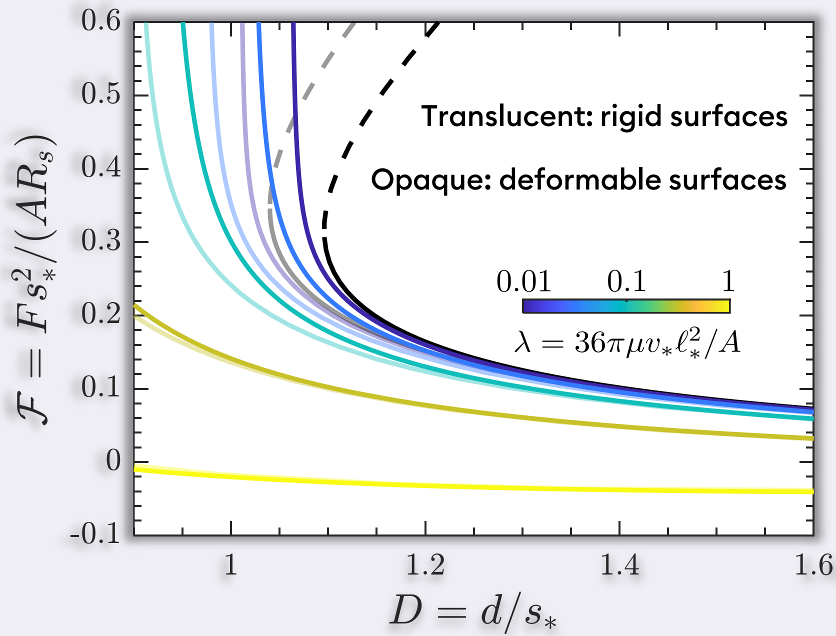
Translucent: constant sphere speed



Perturbation solution

- First-order solution (small surface tension):

$$\mathcal{F}(T) \approx \frac{1}{6S^2}+\frac{\lambda\dot{S}}{6S}+\frac{1}{S^{11/2}}\left(1.0677+1.9377\lambda S\dot{S}+0.6553\lambda^2 S^2\dot{S}^2-0.2892\lambda^2 S^3\ddot{S}\right)\epsilon.$$



$$\lambda = \frac{36\pi\mu v_*\ell_*^2}{A}$$

Typical value: O(1)

A good deal of complexities:
elastic, vdW, viscous forces

Z. Dai*. Jump of an AFM probe towards an elastic substrate in a liquid environment. *Journal of Fluid Mechanics* (2025)

Outline

Conclusions

In situ approaching and detaching from a multilayer graphene sheet

- Pull-off has been well studied, while pull-in can reveal more details but is not well understood.
- We showed that surface deformation can cause the pull-in phenomenon to occur prematurely.
- The concept can be readily extended to other dynamic, nonlinear systems.

$$w(\boldsymbol{r}) = \int_{\mathbb{R}^2} p_{\mathrm{vdW}}(\boldsymbol{x}) \mathrm{G}(\boldsymbol{r} - \boldsymbol{x}) \, \mathrm{d}^2\boldsymbol{x}$$

Parabolic, hyperbolic, elliptic PEDs for statics and dynamics of continuum.

Thanks!