THE UNIVERSITY OF BRITISH COLUMBIA

Presenter Name: Sanket Biswas, MASc Candidate

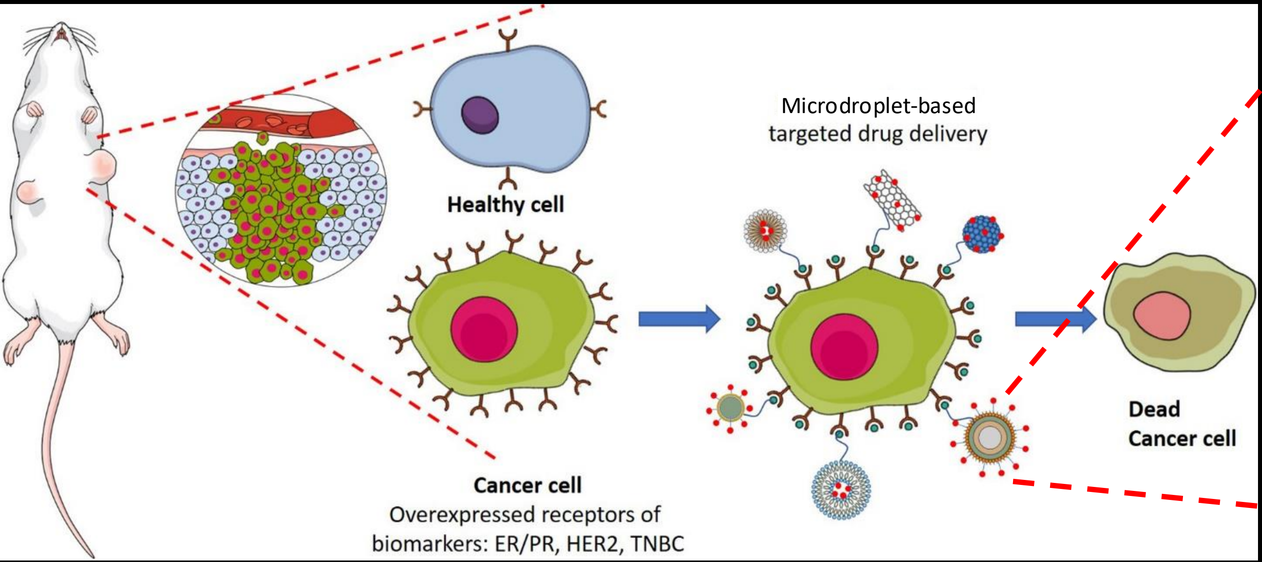
Presentation Title: Rheology of Viscoelastic Microdroplets under Oscillatory Compression

Supervisors: Dr. John M. Frostad and Dr. Gwynn J. Elfring

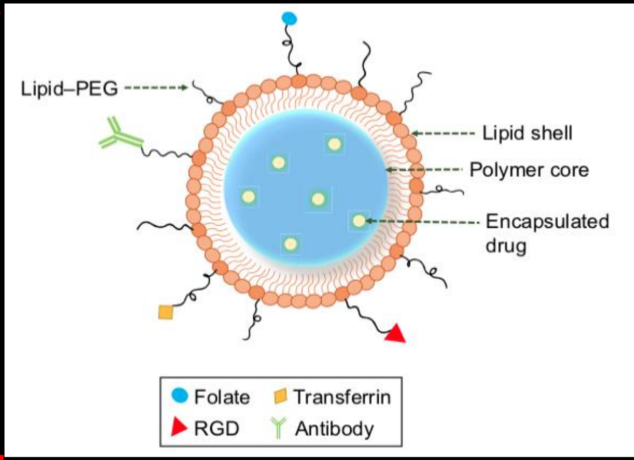
https://ubc.ca1.qualtrics.com/jfe/form/SV_eQZWgRy6emms9kG

Rheology of Micro-Drug Carriers for Targeted Therapy

- 35M+ new cancer cases predicted globally in 2050 (59% rise from 2025)
- Standard treatments (radio-/chemotherapy) cause severe side effects
- *Alternative*: Targeted therapy using viscoelastic micro-drug carriers
- *Challenge*: Precise transport of these microdroplets to cancer cells
- *Solution*: Develop methods to precisely characterize their complex rheology



Liyanage et al., *Biochimica et Biophysica Acta* (2019)



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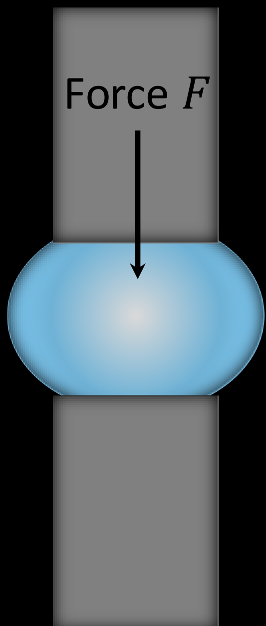
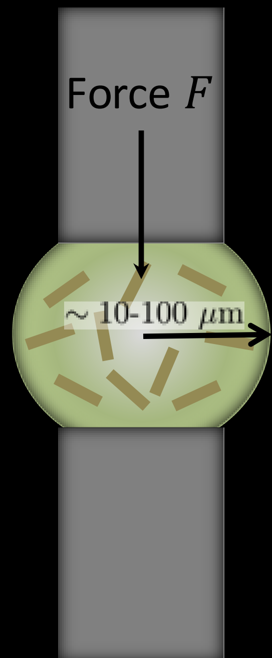
What are Viscoelastic Microdroplets?

Droplets of radius 10-100 μm that exhibit both fluid-like and solid-like behavior.



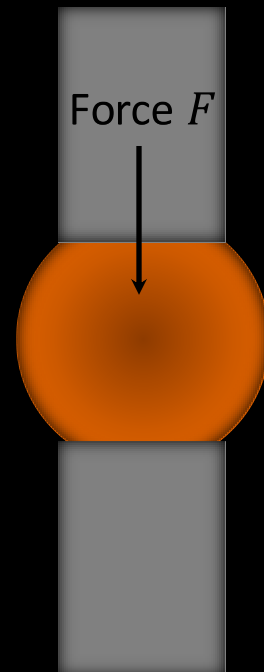
Viscous microdroplet

Bulk viscous stresses
+
Interfacial stresses



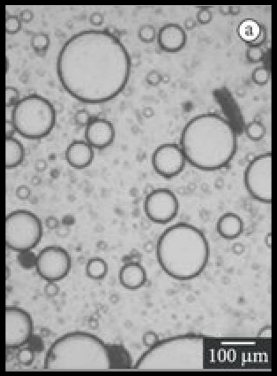
Viscoelastic microdroplet

Bulk viscous stresses
+
Bulk elastic stresses
+
Interfacial stresses

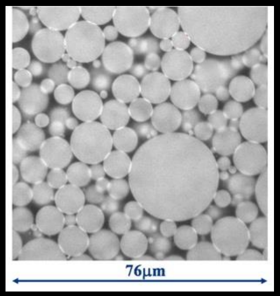


Elastic microparticle

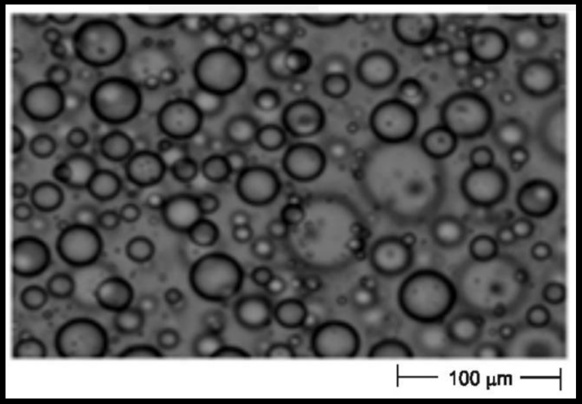
Bulk elastic stresses
+
Interfacial stresses



Salad dressing



Mayonnaise



Conditioner

www.researchgate.net

Response of Viscoelastic Materials under Small Strain

- If a small strain is applied on a viscoelastic material, it can be characterized by its *linear viscoelastic properties*.
- For *small amplitude oscillatory strains*, it is characterized by:
  - Storage modulus G' (elasticity)
  - Loss modulus G'' (viscosity)

Shear modulus G_0

Viscosity μ_0

Maxwell model

$$\boldsymbol{\tau} + \lambda \frac{\partial \boldsymbol{\tau}}{\partial t} = 2\mu_0 \mathbf{E}$$

Relaxation time

$$\lambda = \frac{\mu_0}{G_0}$$

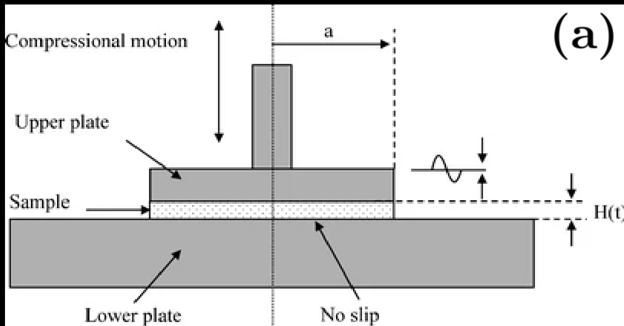
Purely elastic spring

$$\underbrace{\frac{\partial \boldsymbol{\tau}}{\partial t} = 2G_0 \mathbf{E}_e}_{\text{Hooke's law of elasticity}}$$

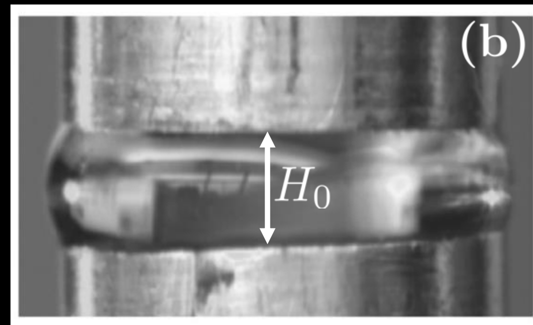
Purely viscous damper

$$\underbrace{\boldsymbol{\tau} = 2\mu_0 \mathbf{E}_v}_{\text{Newton's law of viscosity}}$$

Compress to Characterize Rheology



Bell et al., *Rheologica Acta* (2006)



Barakat et al., *PoF* (2021)

Capillary number

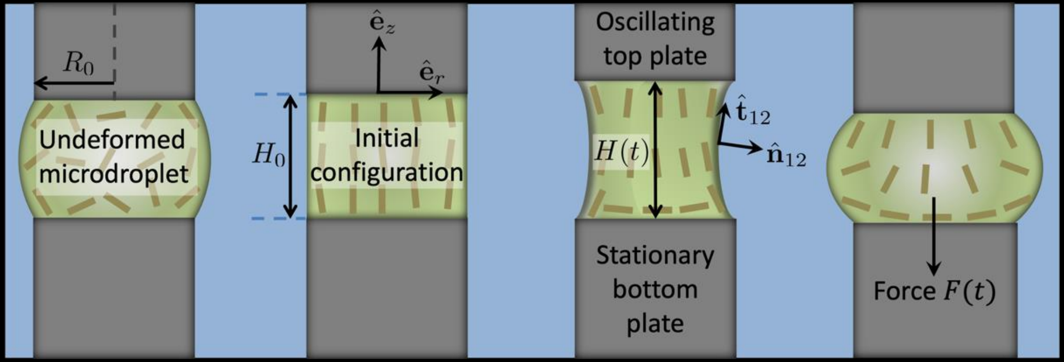
$$\mathrm{Ca} = \frac{\text{viscous forces}}{\text{interfacial forces}} = \frac{G'' H_0}{\gamma}$$

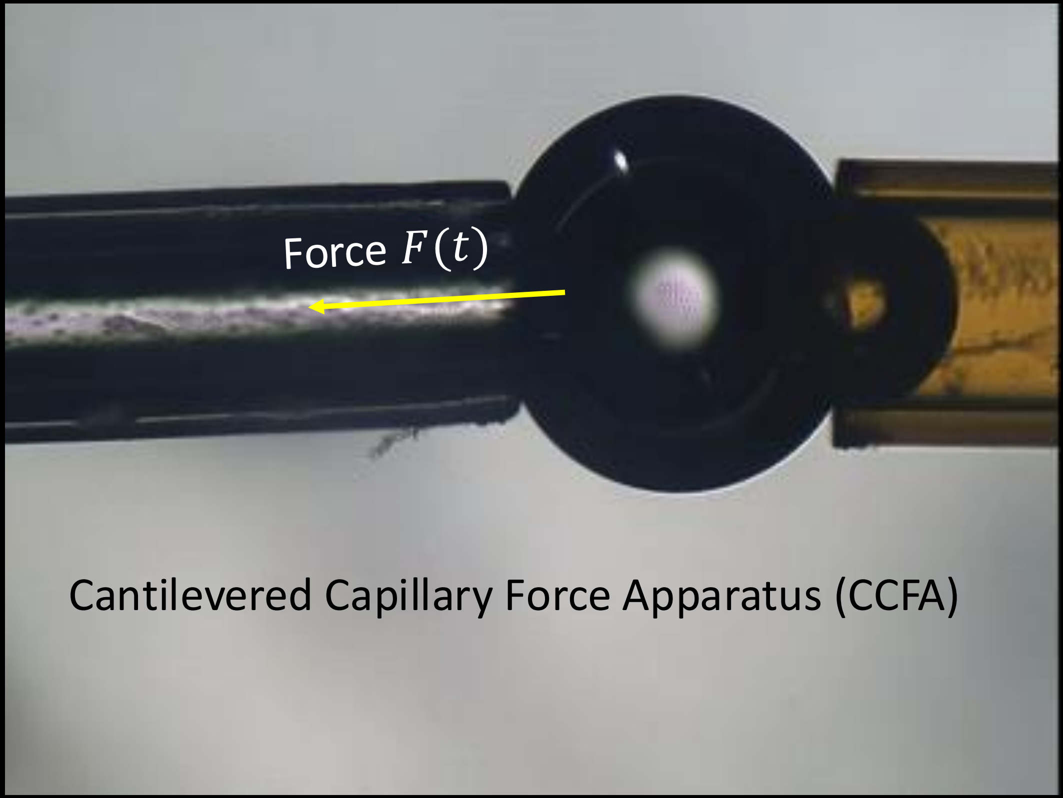
Elasto-Capillary number

$$\mathrm{Ec} = \frac{\text{elastic forces}}{\text{interfacial forces}} = \frac{G' H_0}{\gamma}$$

Author(s)	**Material**	**Type of compression**	**Rheology measured**	Ca	Ec
Stefan et al. (1874)	Thin viscous film	Constant velocity	μ	$\mathrm{Ca} \gg 1$	---
Phan-Thien (1980)	Thin viscoelastic film	Oscillatory; between plates	---	$\mathrm{Ca} \gg 1$	$\mathrm{Ec} \gg 1$
Bell et al. (2006)			G', G''		
Wingstrand et al. (2016)					
Zheng et al. (2023)		Constant velocity; between spheres and plates	---		
Zheng et al. (2024)					
Mederos et al. (2024)		Constant force			
Barakat et al. (2021)	mm-sized viscous droplet	Oscillatory	γ, μ	$\mathrm{Ca} \lesssim 1$	---
No prior literature	Viscoelastic microdroplet	Oscillatory	γ, G', G''	$\mathrm{Ca} \lesssim 1$	$\mathrm{Ec} \lesssim 1$

Force Response and Rheology of Viscoelastic Microdroplets Under Oscillatory Compression





1) Derive the force response of a viscoelastic microdroplet under oscillatory compression.

2) Develop a methodology to decouple its rheology from the measured force response (using the CCFA).

3) Assess the accuracy of this framework across different capillary numbers, elasto-capillary numbers, and microdroplet types.

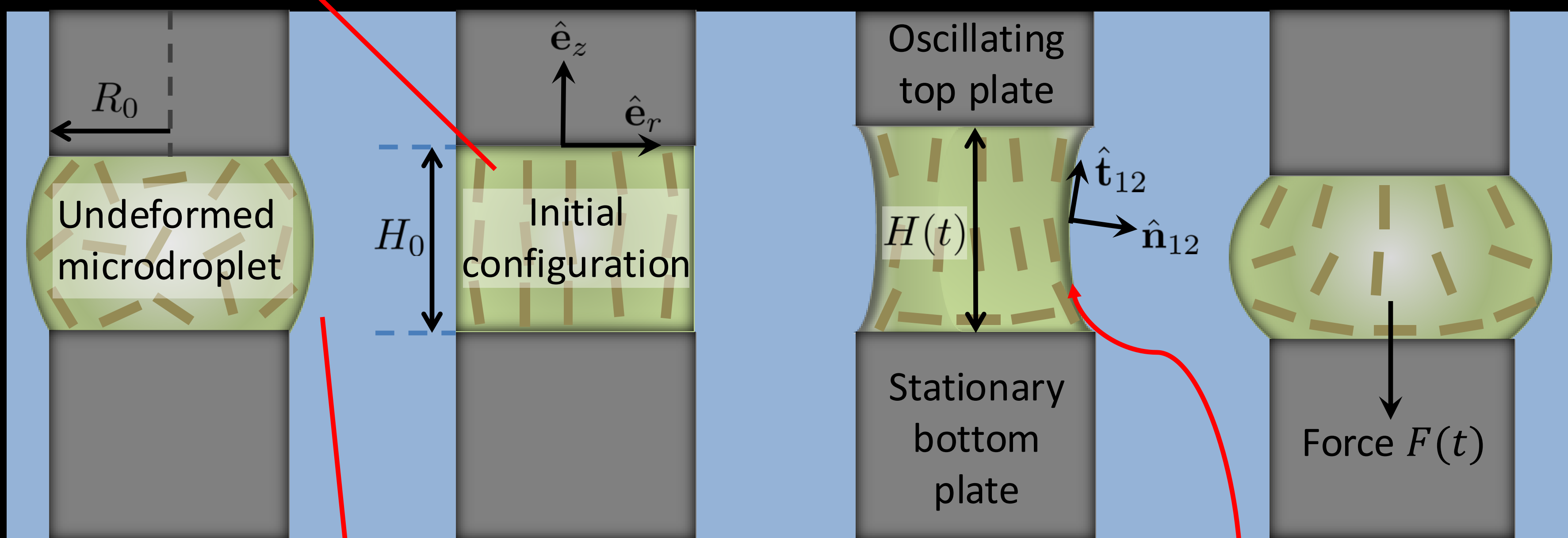
Analytical Model

Lubrication (thin gap) flow in viscoelastic microdroplet

$$\text{Momentum conservation:} \quad \frac{\partial p_d}{\partial r} = \int_0^\infty \left\{ G(\tau) \frac{\partial^2 u_z}{\partial z^2} \right\} \mathrm{d}\tau \qquad (1)$$

$f\left(G', G''\right)$

$$\text{Mass conservation:} \quad \boldsymbol{\nabla} \cdot \mathbf{u} = \mathbf{0} \qquad (2)$$



Stokes (negligible inertia) flow in ambient Newtonian medium

$$\text{Momentum conservation:} \quad \rho_a \mathbf{g} + \boldsymbol{\nabla} \cdot \boldsymbol{\sigma}_a = \mathbf{0} \qquad (3)$$

$$\text{Mass conservation:} \quad \boldsymbol{\nabla} \cdot \mathbf{u}_a = \mathbf{0} \qquad (4)$$

Stress balance at the interface

$$\hat{\mathbf{n}}_{12} \cdot \left(\boldsymbol{\sigma}_a - \boldsymbol{\sigma}\right) = \gamma \hat{\mathbf{n}}_{12} \left(\boldsymbol{\nabla} \cdot \hat{\mathbf{n}}_{12}\right) \qquad (5)$$

Force Response

$$F(t) = \pi R_0 \gamma - \pi R_0^2 H_0 \rho g + \left[\frac{6\pi a R_0^3 \gamma}{H_0^2} + \frac{3\pi a R_0^4 G'}{2H_0^2}\right]\sin(\omega t) + \frac{3\pi a R_0^4 G''}{2H_0^2}\cos(\omega t)$$

Line interfacial tension
+
Laplace pressure

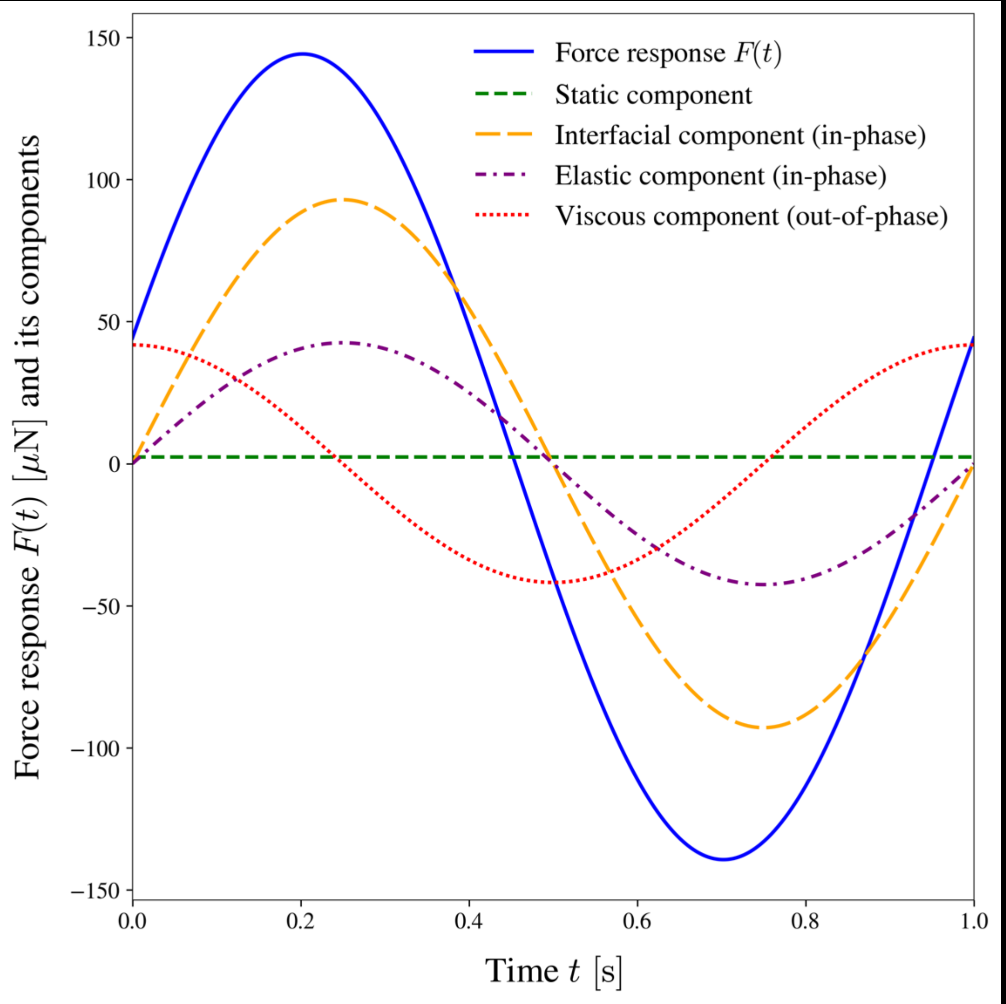
Hydrostatic pressure

Interfacial stresses

Bulk elastic stresses

Bulk viscous stresses

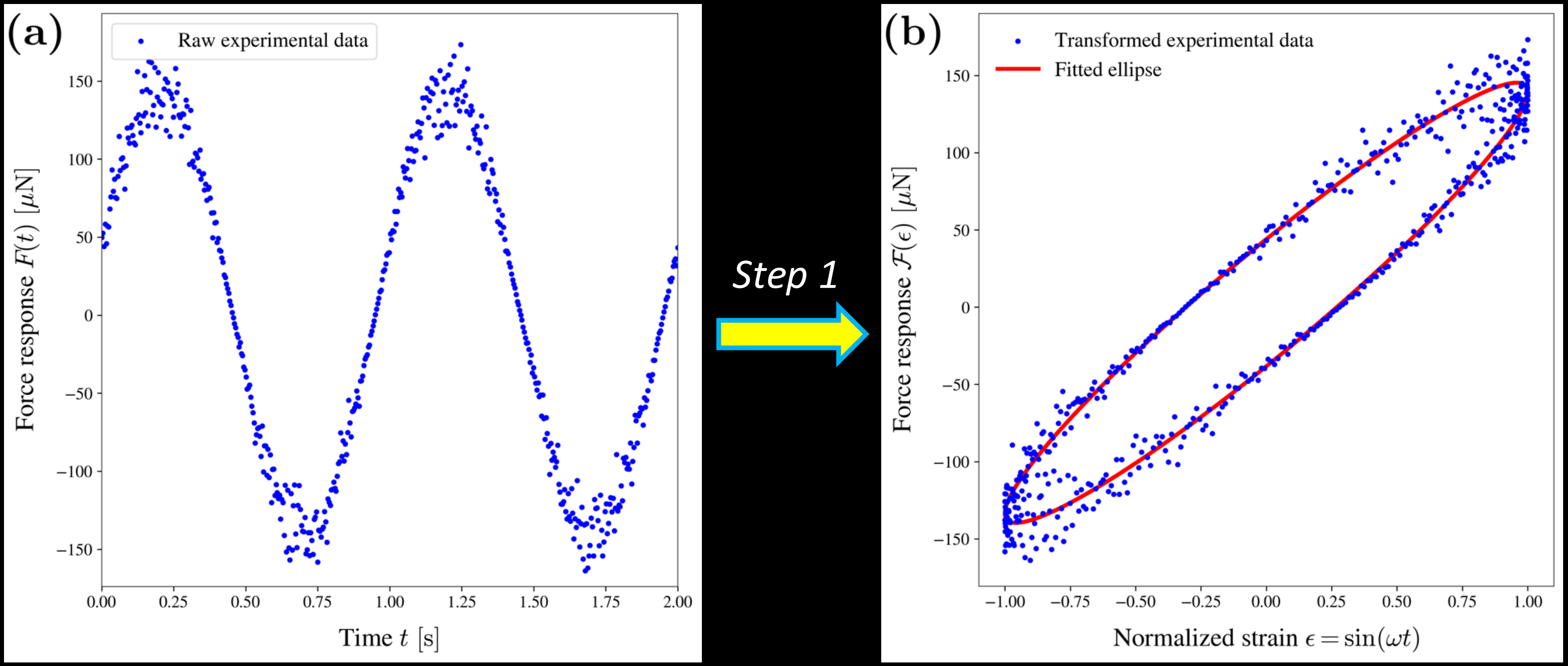
Static component



Applied strain

$$\frac{H(t) - H_0}{H_0} = a\sin(\omega t)$$

Methodology to Decouple Rheological Properties



Step 2: To the transformed force response data, fit an ellipse of the form:

$$B_1\epsilon^2 + B_2\epsilon\mathcal{F} + \mathcal{F}^2 + B_4\epsilon + B_5\mathcal{F} + B_6 = 0 \qquad (1)$$

Step 3: Using the fitting coefficients B_1, B_2, and B_5, determine:

Interfacial tension

$$\gamma = \rho g H_0 R_0 - \frac{B_5}{2\pi R_0}$$

Storage modulus

$$G' = -\frac{1}{R_0}\left(\frac{B_2 H_0^2}{3\pi a R_0^3} + 4\gamma\right)$$

Loss modulus

$$G'' = \frac{2H_0^2}{3\pi a R_0^4}\sqrt{\frac{B_5^2}{4} - B_6}$$

Test Cases

	Droplet	γ [N/m]	G' [Pa]	G'' [Pa]	Ca	Ec
Ca/Ec ~ 1	Droplet 1	0.022	25.120	24.960	3.12×10^{-3}	3.14×10^{-3}
	Droplet 2	0.035	2914.545	2863.636	2.25×10^{-1}	2.29×10^{-1}
	Droplet 3	0.021	7789.091	8018.182	1.05×10^{0}	1.02×10^{0}
Ca/Ec ~ 10	Droplet 4	0.045	53.018	1017.818	6.22×10^{-2}	3.24×10^{-3}
	Droplet 5	0.031	152.182	5906.909	5.24×10^{-1}	1.35×10^{-2}
	Droplet 6	0.025	1224.545	9545.455	1.05×10^{0}	1.35×10^{-1}
Ca/Ec ~ 0.1	Droplet 7	0.045	205.691	38.553	2.36×10^{-3}	1.26×10^{-2}
	Droplet 8	0.027	1441.309	114.578	1.17×10^{-2}	1.47×10^{-1}
	Droplet 9	0.019	7116.364	704.727	1.02×10^{-1}	1.03×10^{0}

Sample viscoelastic microdroplets of radius $R_d = 10\ \mu m$

- Experimental force response for all droplets is modeled as:

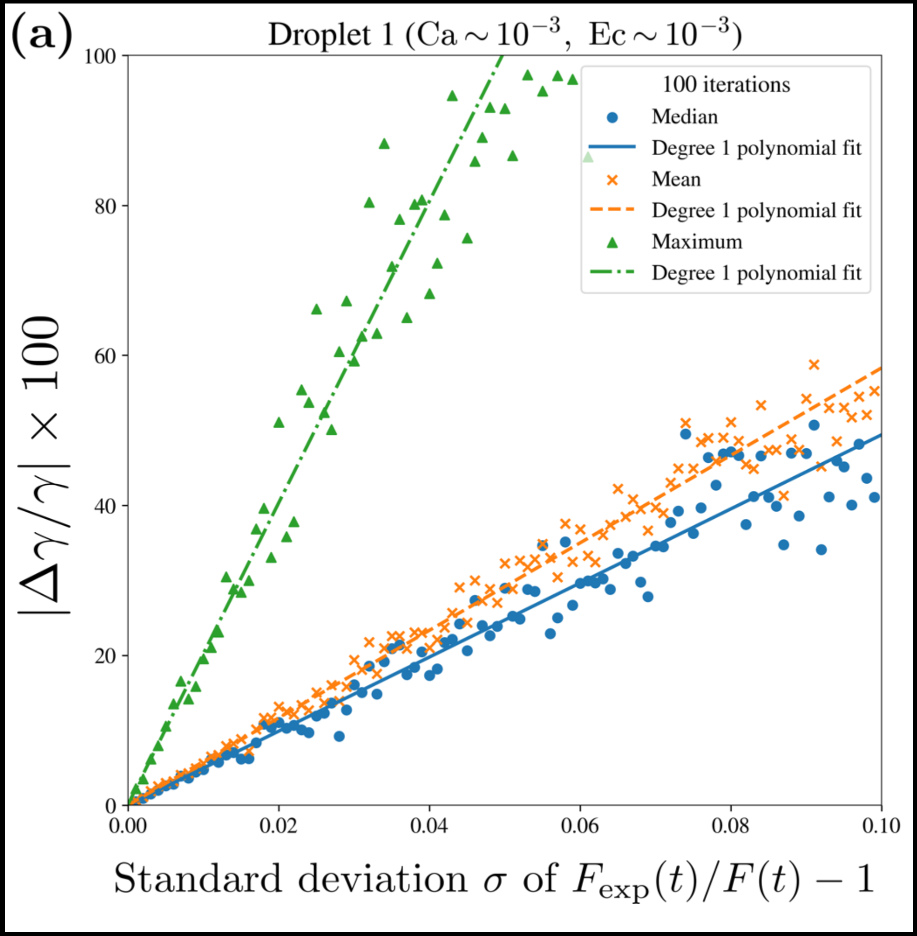
$$F_{\exp}(t) = F(t)\,[1 + \delta(t)]$$

Theoretical force response

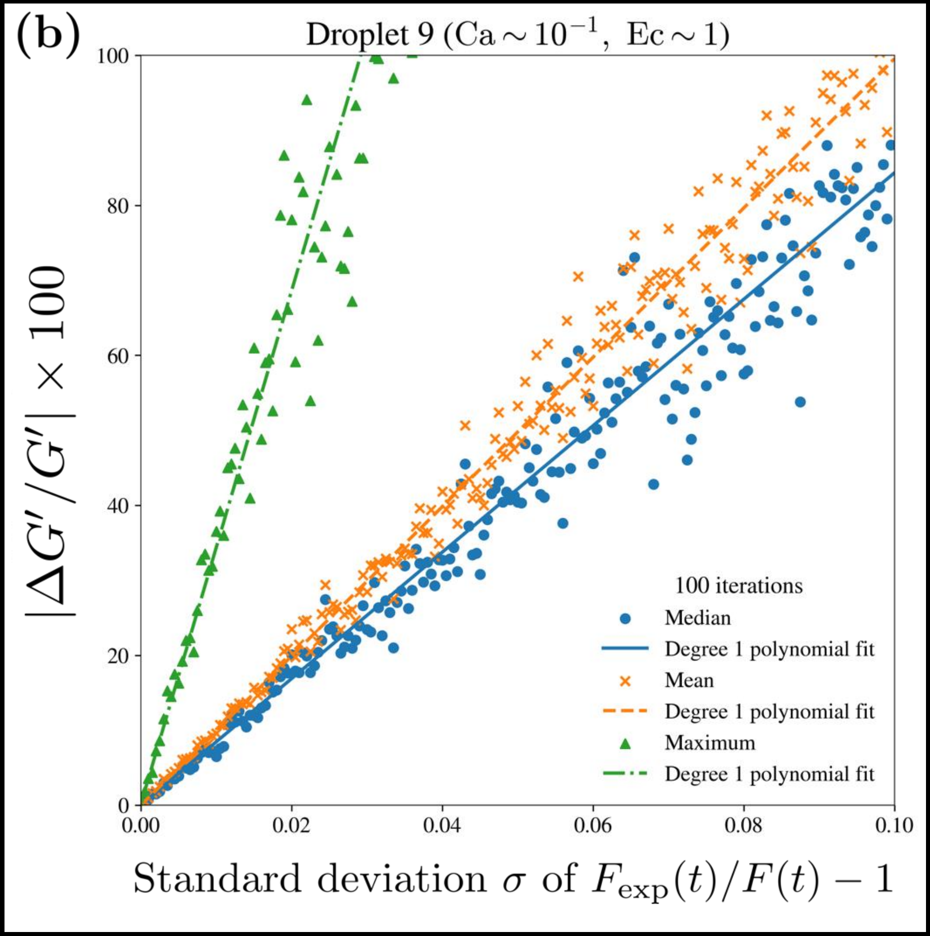
Gaussian white noise with mean 0 and standard deviation $\sigma \ll 1$

Validation

% relative error in γ vs. noise in force data



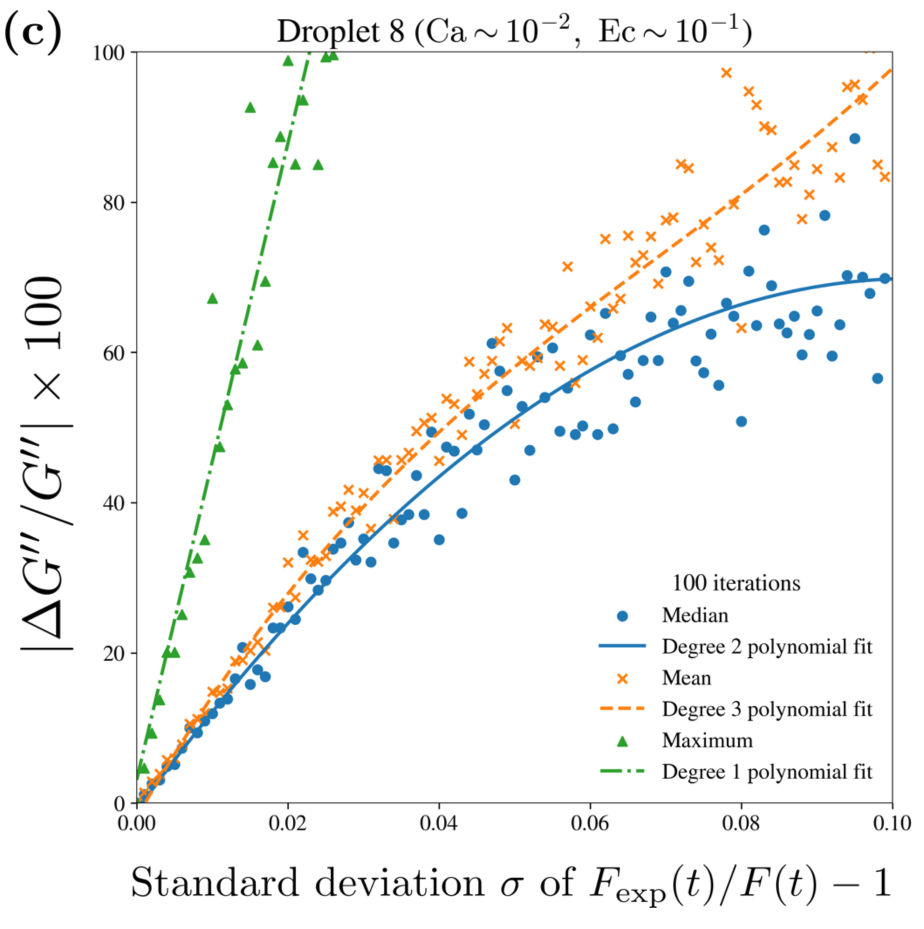
% relative error in G' vs. noise in force data



Thumb rule: At any standard deviation $\sigma = \sigma_0$, $F_{\text{exp}}(t)$ lies within $3\sigma_0$ of $F(t)$

Validation

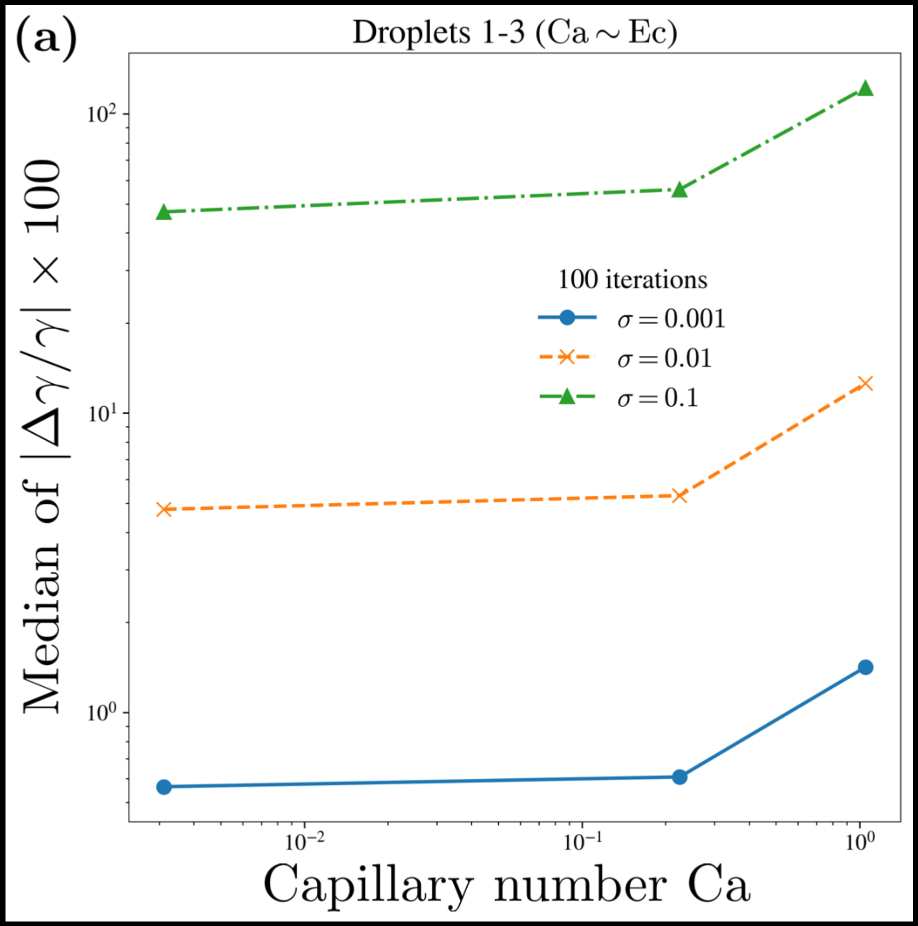
% relative error in G'' vs. noise in force data



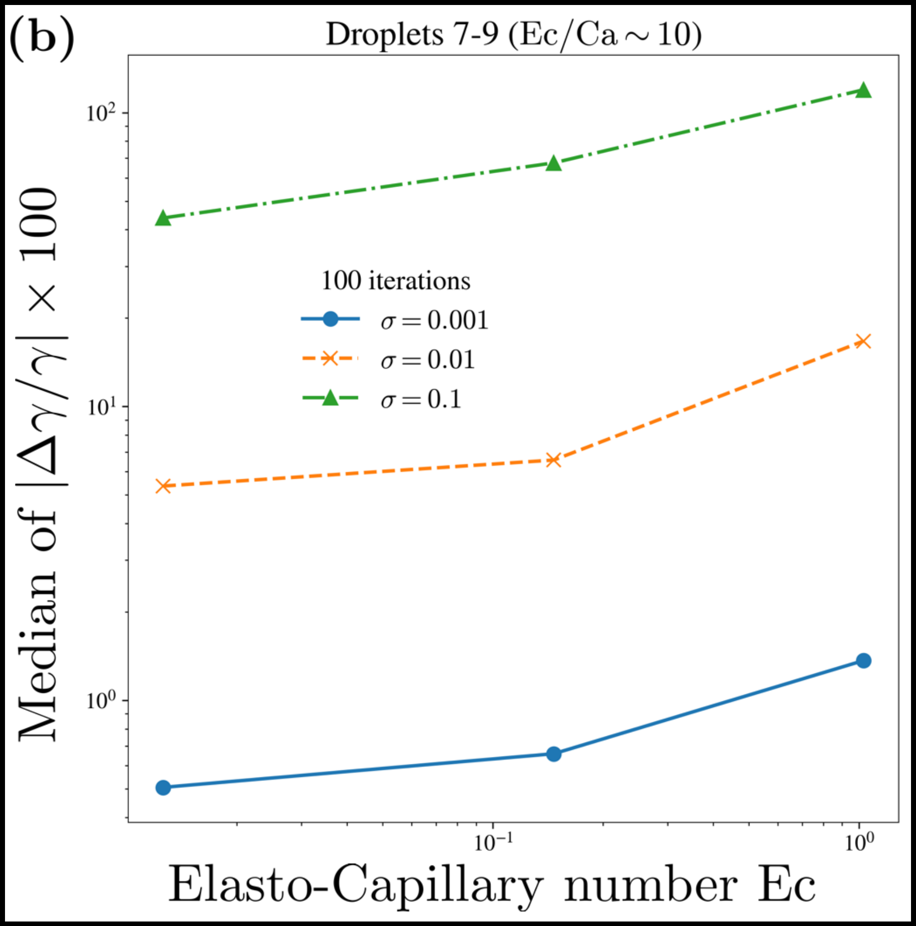
Thumb rule: At any standard deviation $\sigma = \sigma_0$, $F_{\exp}(t)$ lies within $3\sigma_0$ of $F(t)$

Interfacial Tension is Determined More Accurately for Microdroplets with Lower Viscosity and Elasticity

Median % relative error in γ vs. Ca (log-log)



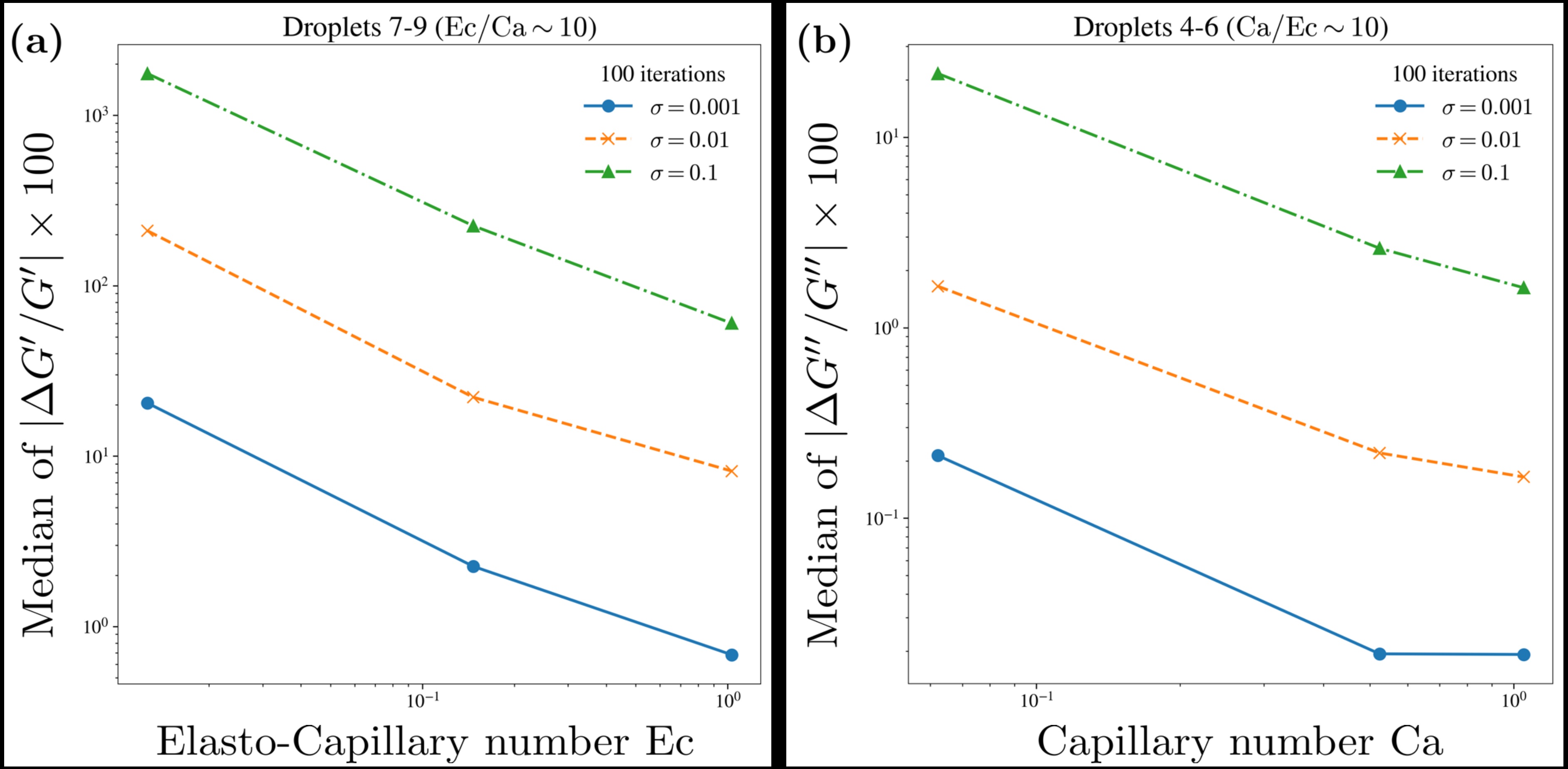
Median % relative error in γ vs. Ec (log-log)



Storage and Loss Moduli is Determined More Accurately for Microdroplets with Higher Elasticity and Viscosity, respectively

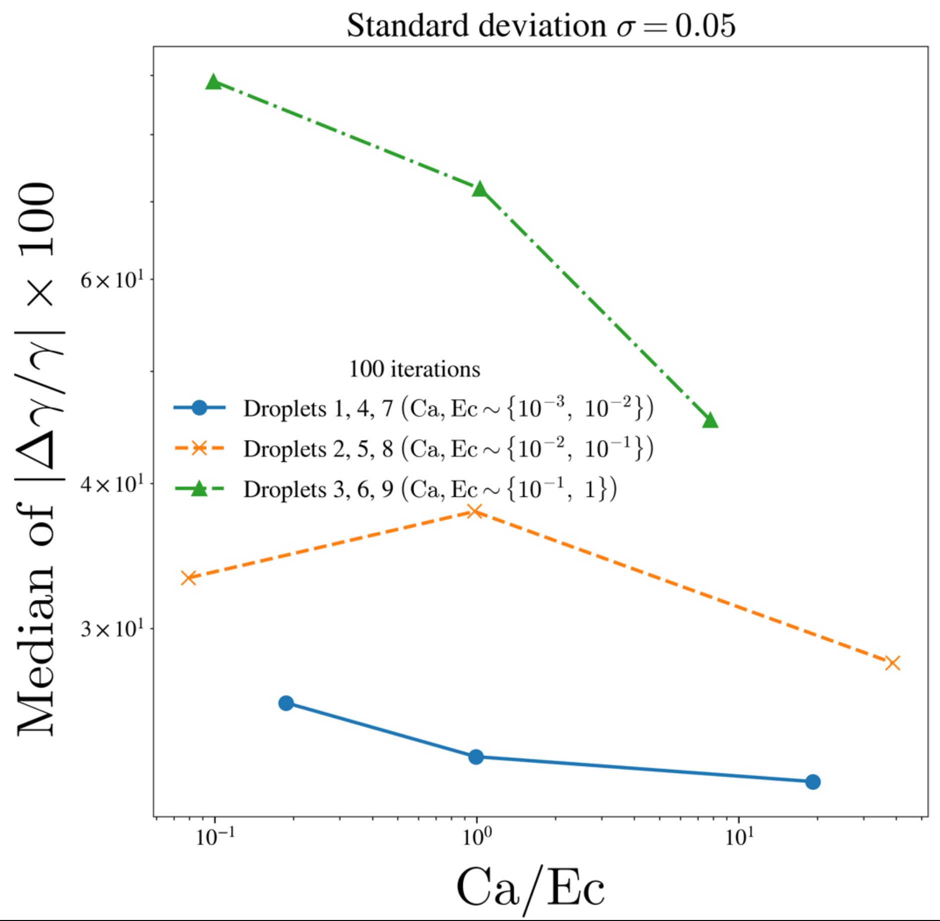
Median % relative error in G' vs. Ec (log-log)

Median % relative error in G'' vs. Ca (log-log)



Accuracy in Determining Interfacial Tension is Highest for Fluid-Like Droplets

Median % relative error in γ vs. Ca/Ec (log-log)



Thumb rule

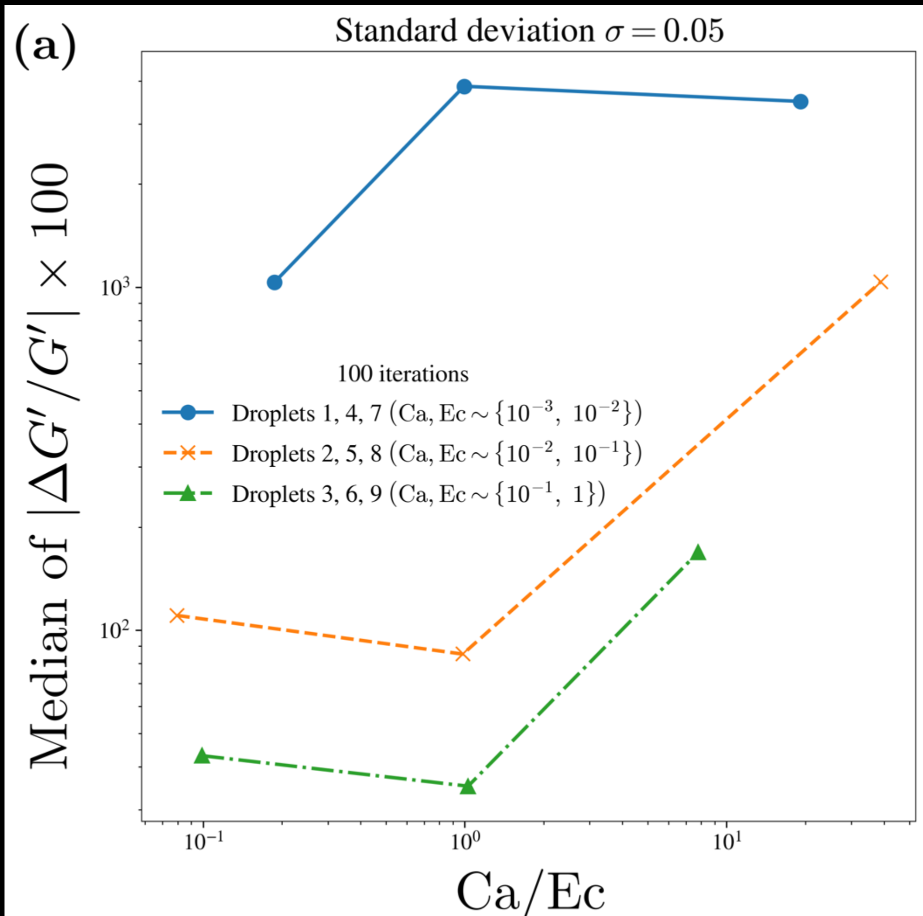
(a) $\mathrm{Ca/Ec} \sim 10^{-1}$: Solid-like

(b) $\mathrm{Ca/Ec} \sim 10^{0}$: Equally fluid-like and solid-like

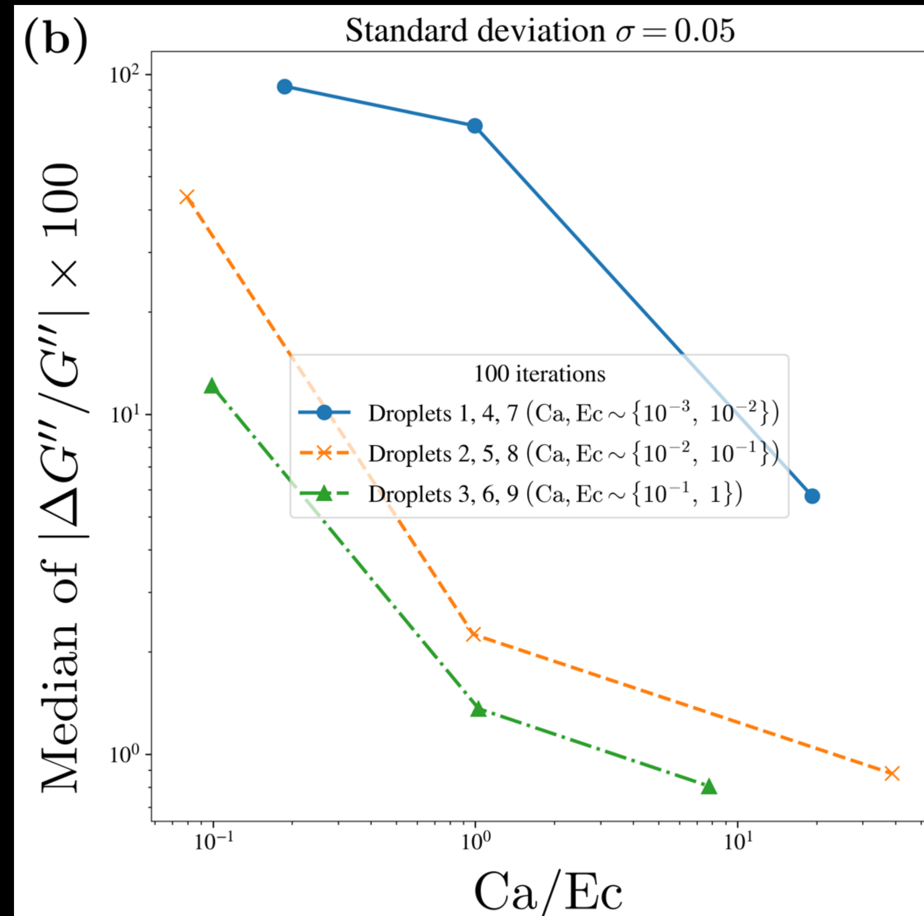
(c) $\mathrm{Ca/Ec} \sim 10^{1}$: Fluid-like

Accuracy in Determining Storage and Loss Moduli is Highest for Solid-Like and Fluid-Like Droplets, respectively

Median % relative error in G' vs. Ca/Ec (log-log)



Median % relative error in G'' vs. Ca/Ec (log-log)



Thumb rule

(a) $\mathrm{Ca/Ec} \sim 10^{-1}$: Solid-like

(b) $\mathrm{Ca/Ec} \sim 10^{0}$: Equally fluid-like and solid-like

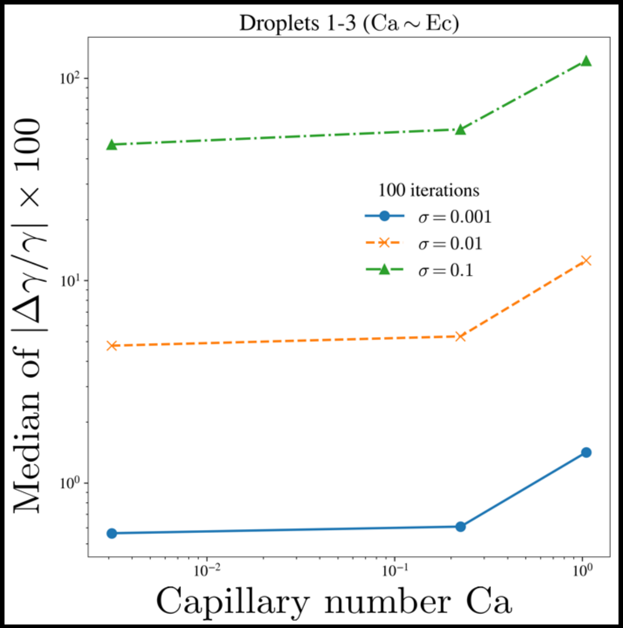
(c) $\mathrm{Ca/Ec} \sim 10^{1}$: Fluid-like

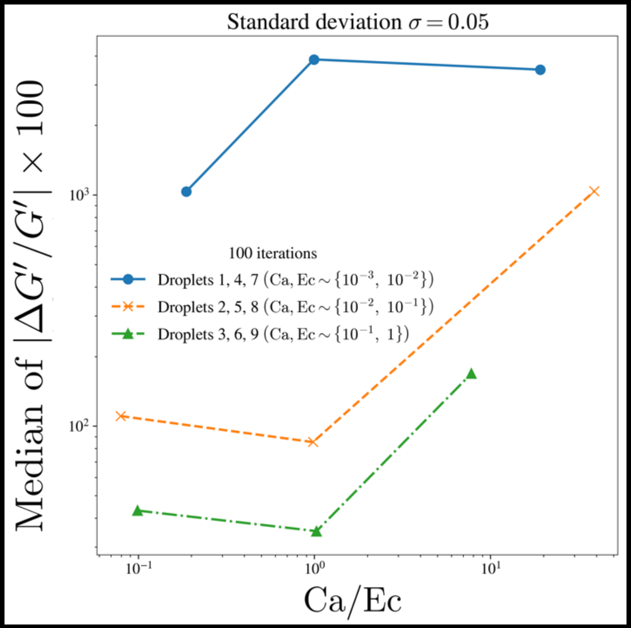
Key Takeaways

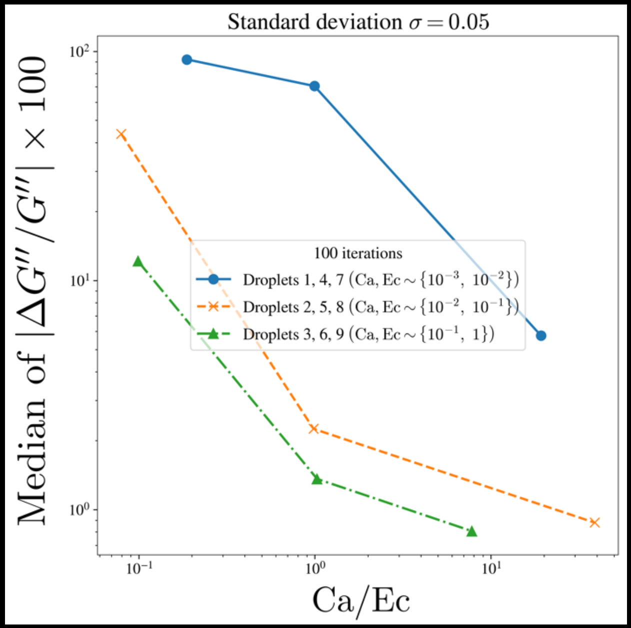
1. Developed an analytical framework to decouple rheology of viscoelastic microdroplets.

2. Accuracy of determining rheology is influenced by the droplet's capillary and elasto-capillary numbers.

3. Droplet's material type also plays a role in determining rheological accuracy.

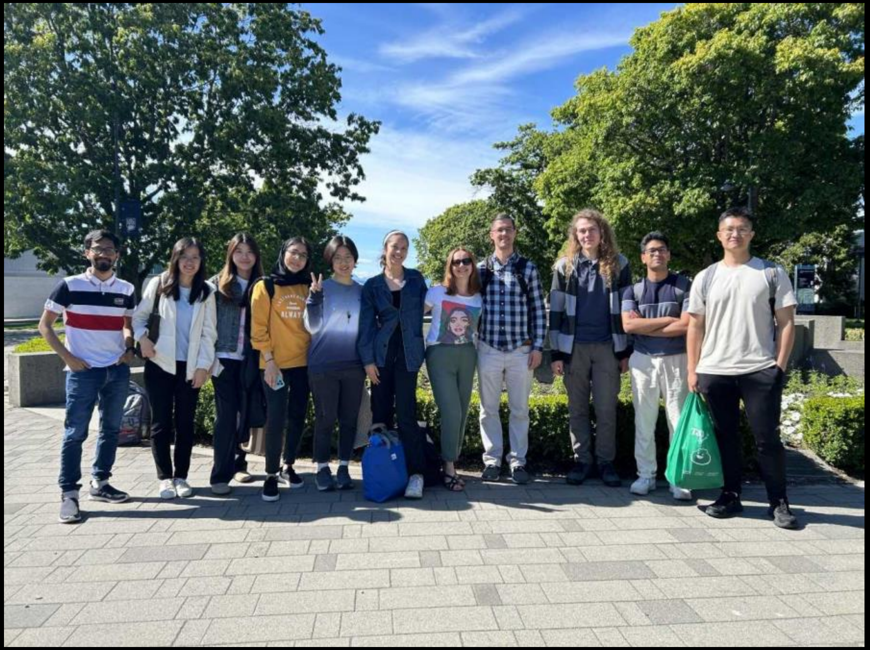






Acknowledgements

The Soft Matter Group



Frostad Research Group

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Thank you for your feedback!

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