Rheology of Viscoelastic Microdroplets Under Oscillatory Compression

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BACKGROUND: Droplets with radii between $10\ \mu m$ and $100\ \mu m$ that exhibit both fluid-like (viscous) and solid-like (elastic) behavior are known as *viscoelastic microdroplets*. Their rheology plays a critical role in designing drug carriers for targeted cancer therapies, stabilizing emulsified food products (e.g., dressings and sauces), and enhancing the performance of cosmetic formulations (e.g., moisturizers and foundations). However, due to their small size and deformable nature, very few devices—other than the Cantilevered Capillary Force Apparatus (CCFA)—are capable of measuring their rheological properties. The CCFA is designed to accurately measure the force response of viscoelastic microdroplets (with magnitudes of at most $100\ \mu$ N) under oscillatory compression (see Fig. 1). We present an analytical framework that decouples the interfacial tension, storage modulus (elasticity), and loss modulus (viscosity) of such droplets from this force response.

METHODOLOGY

1. Using mass and momentum conservation [Eqns. (1) and (2)] along with the Maxwell model [Eqn. (3)], we derive the flow field of the oscillating microdroplet (Fig. 1):

$$\nabla \cdot \mathbf{u} = \mathbf{0}, \qquad (1)$$

$$-\nabla p + \rho \mathbf{g} + \nabla \cdot \boldsymbol{\tau} = \mathbf{0}, \qquad (2)$$

$$\boldsymbol{\tau} + \lambda \frac{\partial \boldsymbol{\tau}}{\partial t} = 2\mu_0 \mathbf{E}. \qquad (3)$$

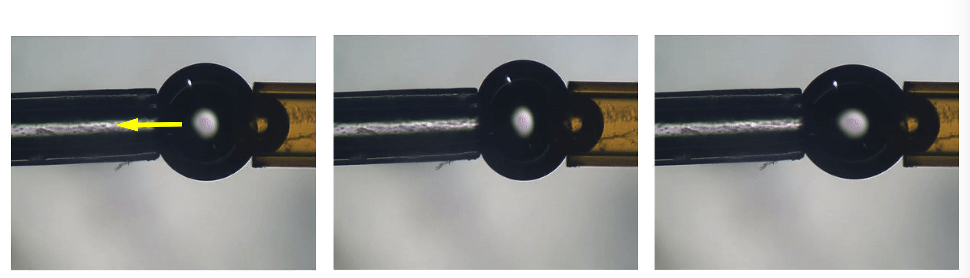
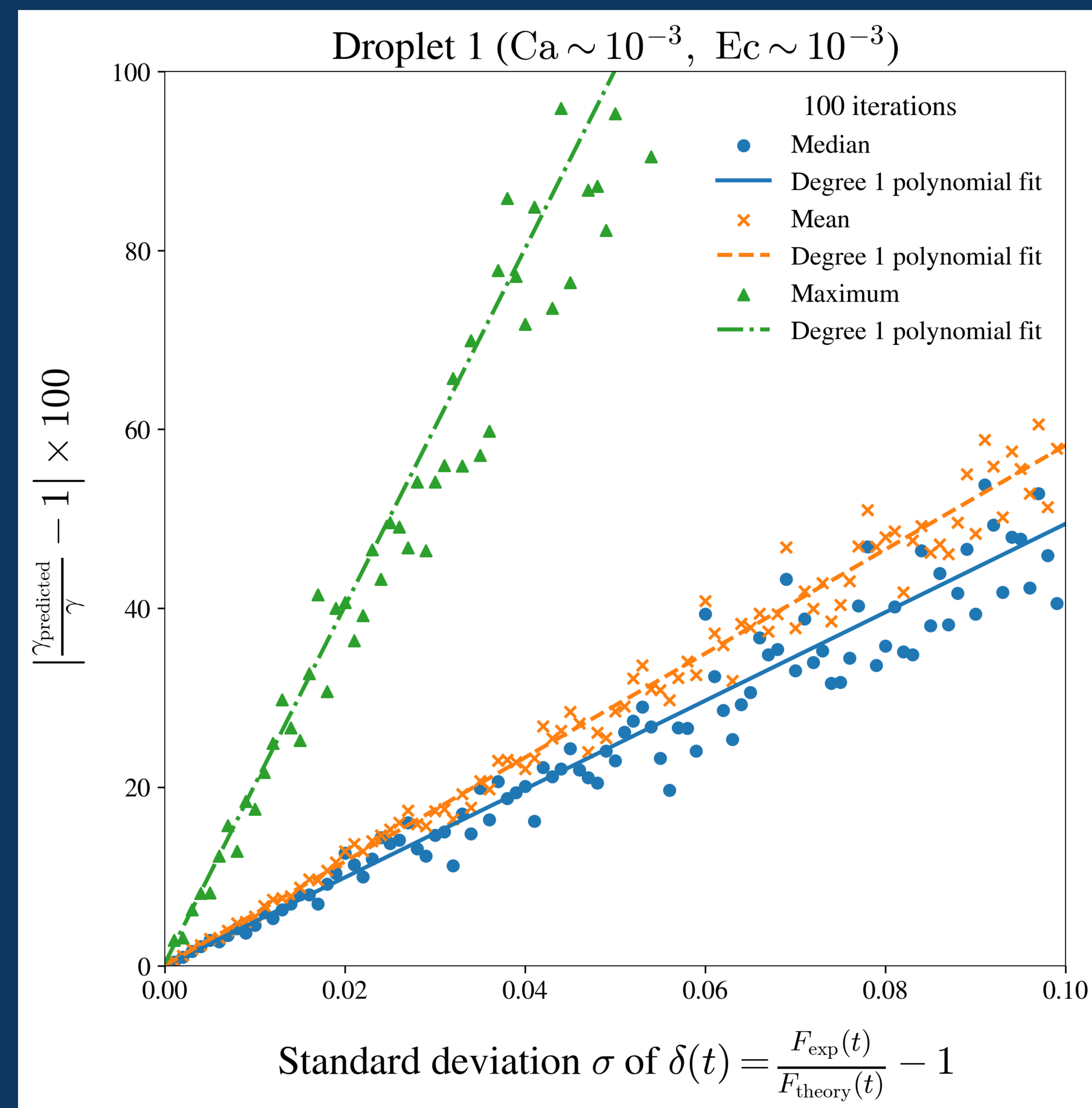


Fig. 1: A viscoelastic microdroplet undergoing oscillatory compression in the CCFA. The arrow indicates the force direction.

2. The force exerted by the droplet on the capillaries is given by:

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

By simply **squeezing** a **viscoelastic microdroplet** between two substrates in an **oscillatory fashion**, we can accurately characterize its…



Interfacial tension γ

Fig. 3: % relative error in interfacial tension γ vs. relative error in force response for a microdroplet (Droplet 1) with $\gamma = 0.022$ N/m, $G' = 25.120$ Pa, and $G'' = 24.960$ Pa.

Storage modulus G' (elasticity)

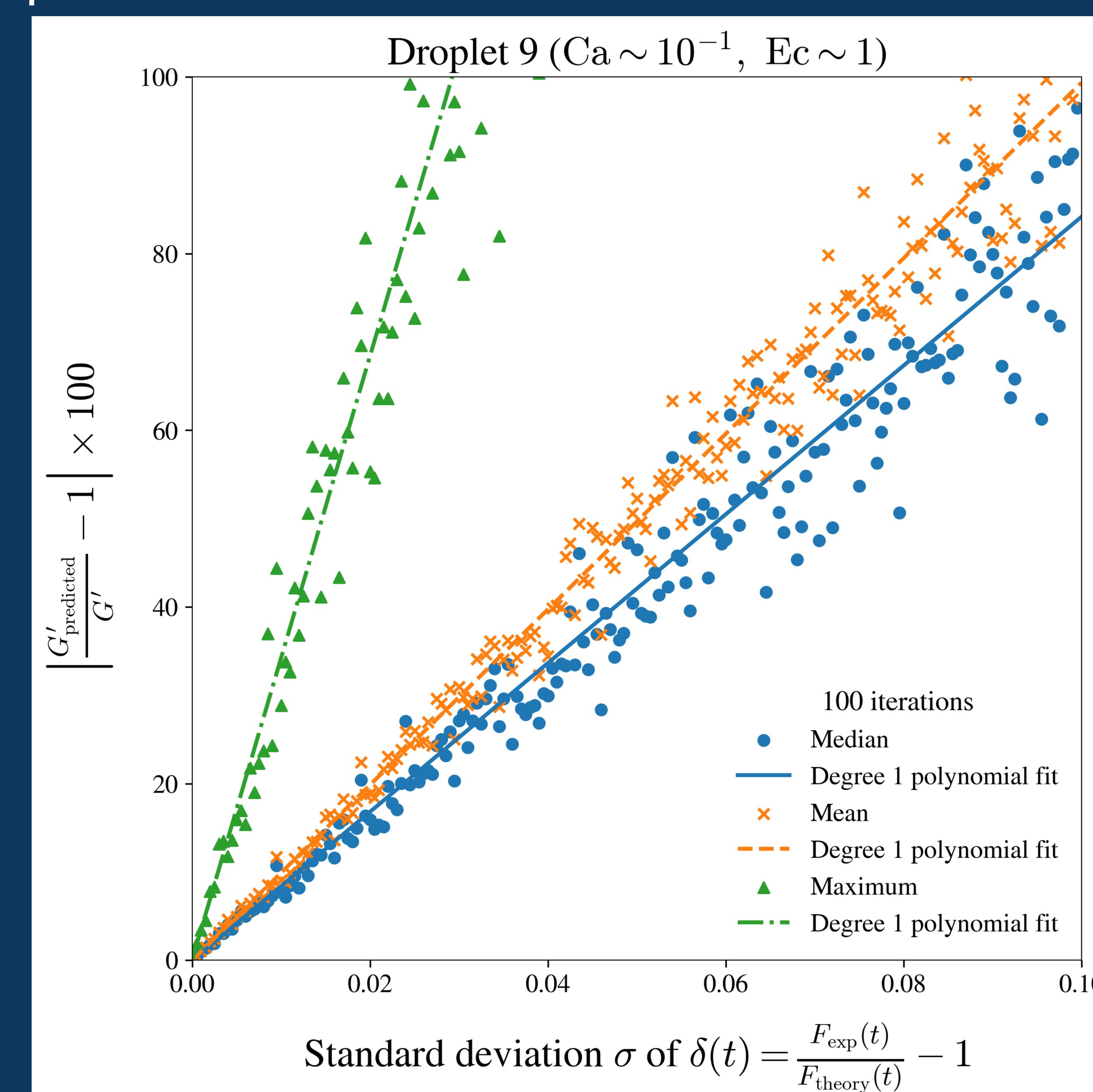


Fig. 4: % relative error in storage modulus G' vs. relative error in force response for a microdroplet (Droplet 9) with $\gamma = 0.019$ N/m, $G' = 7116.364$ Pa, and $G'' = 704.727$ Pa.

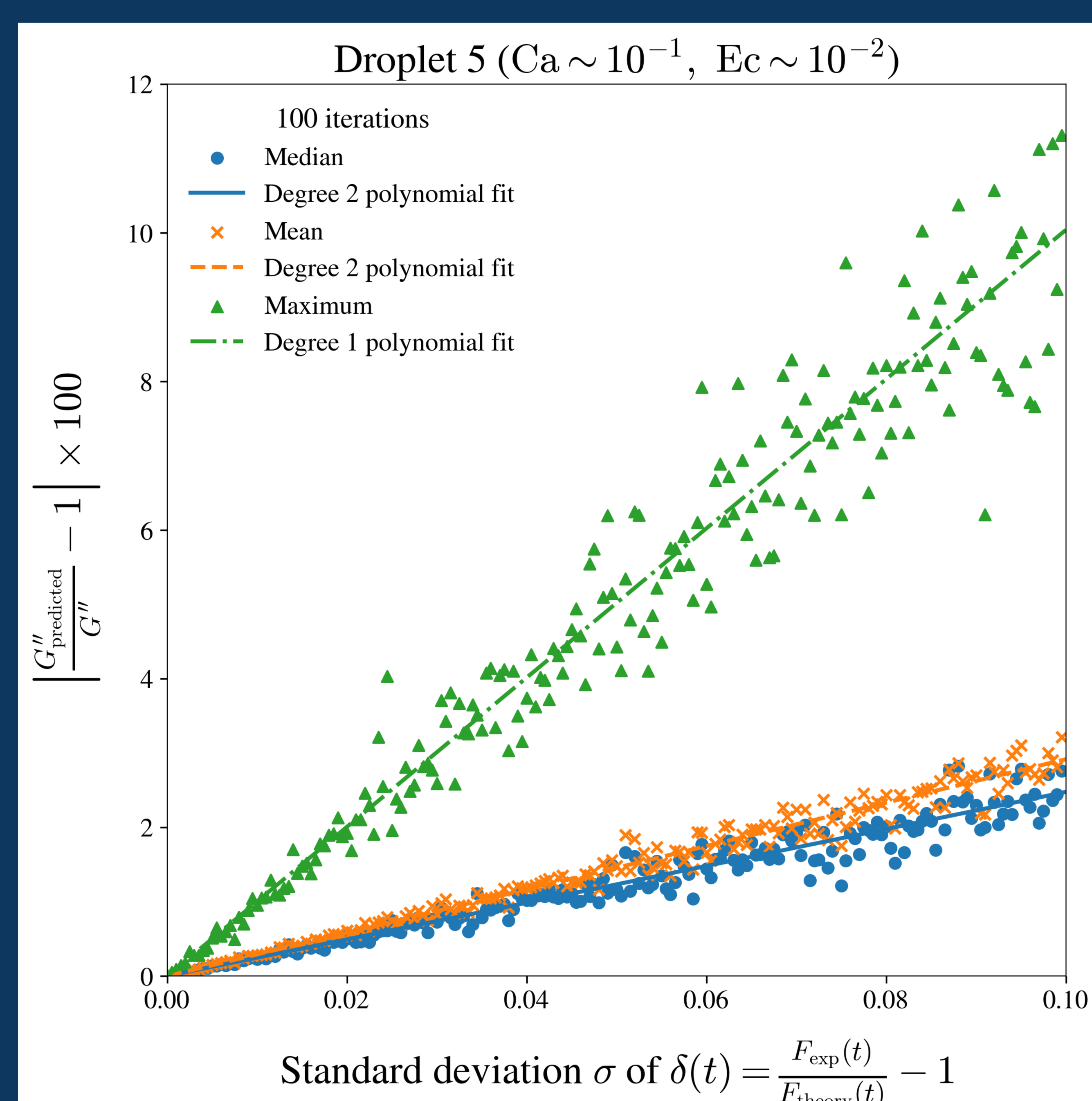


Fig. 5: % relative error in loss modulus G'' vs. relative error in force response for a microdroplet (Droplet 5) with $\gamma = 0.031$ N/m, $G' = 152.182$ Pa, and $G'' = 5906.909$ Pa.

Loss modulus G'' (viscosity)

…even from highly noisy experimental data!

METHODOLOGY CONTD…

3. The force-time data, $[t, F(t)]$, is transformed into force-normalized strain data, $[\epsilon, \mathcal{F}(\epsilon)]$ (refer to Fig. 2), and then fitted with an ellipse of the form:

$$B_1\epsilon^2 + B_2\epsilon\mathcal{F} + \mathcal{F}^2 + B_4\epsilon + B_5\mathcal{F} + B_6 = 0. \qquad (5)$$

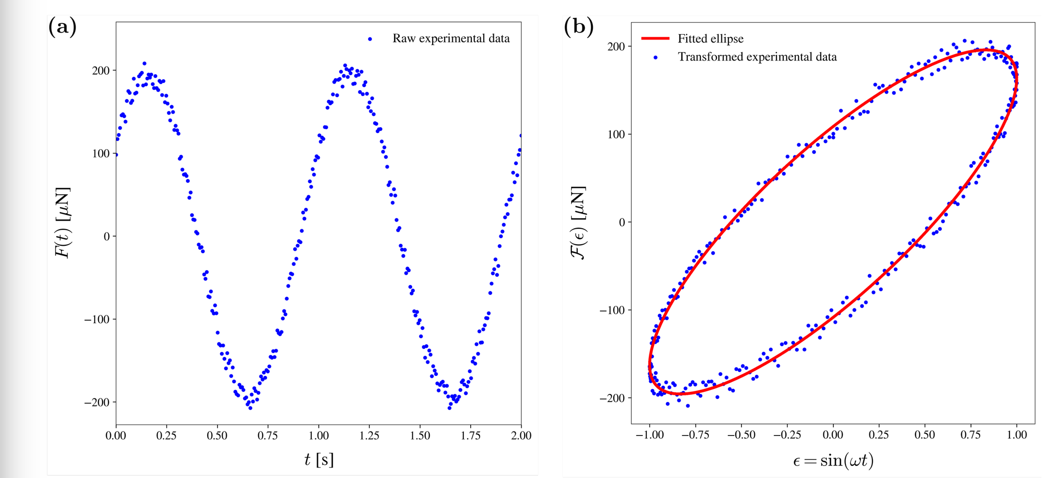


Fig. 2: (a) Raw force-time experimental data, and **(b)** transformed force-normalized strain data, fitted to an ellipse of the form in Eqn. (5).

4. The rheological properties are extracted from the coefficients B_1, B_2, and B_5 using the relations:

$$\gamma = \rho g H_0 R_0 - \frac{B_5}{2\pi R_0}, \qquad (6a)$$

$$G' = -\frac{\Lambda}{H_0}\left(\frac{B_2\Lambda^2}{3\pi a R_0} + 4\gamma\right), \qquad (6b)$$

$$G'' = \frac{2\Lambda^3}{3\pi a H_0 R_0}\left(B_1 - \frac{B_2^2}{4}\right)^{1/2}. \qquad (6c)$$

RESULTS

- We assess the accuracy of our analytical model by determining the rheological properties of viscoelastic droplets of radius $R_d = 10\ \mu$ m and varying magnitudes of capillary number (Ca) and elasto-capillary number (Ec) [see Figs. 3, 4, and 5].
- The experimental force response for all droplets is modeled as:

$$F_{\text{exp}}(t) = F(t)\left[1 + \delta(t)\right], \qquad (7)$$

where $\delta(t)$ is a Gaussian-distributed random variable with mean 0 and standard deviation σ, representing the noise in the experimental data.

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