How capillarity affects the propagation of elastic waves in soft gels

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Elastic waves propagating at the interface of soft solids can be altered by the presence of external forces such as capillarity or gravity. We measure the dispersion relation of waves at the free surface of agarose gels with great accuracy, revealing the existence of multiple modes as well as an apparent dispersion. We disentangle the role of capillarity and elasticity by considering the 3D nature of mechanical waves, achieving quantitative agreement between theoretical predictions and experiments. Notably, our results show that capillarity plays an important role for wavenumbers much smaller than expected from balancing elastic and capillary forces. We further confirm the efficiency of our approach by including the effect of gravity in our predictions and quantitatively comparing it to experiments.

Introduction. – Mechanical waves propagating in biological tissues have been at the center of attention since the development of ultrasonic imaging more than 50 years ago [1]. Using soft elastic materials to mimic the physics of wave propagation inside the body has enabled to develop technological innovations, such as elastography, allowing for a direct measurement of the bulk elastic properties [2, 3]. Soft solids have also been used as a model for fracture dynamics [4, 5] and for geological systems, in particular the role of friction and fault structure on rupture dynamics during earthquakes [6, 7]. Wave propagation at interfaces raises the question of additional forces competing with elasticity. Indeed, solid interfaces, like liquid ones, possess a surface tension $\gamma$ that dominates bulk elasticity at small scales, below the elastocapillary length $\ell_{ec} = \gamma/\mu$ where $\mu$ is the solid shear modulus [8–10]. Depositing liquid drops on soft substrates allows to probe the competition between elasticity and capillarity, as the wetting ridge induced by the contact line sets the drop's statics and dynamics [11]. For very soft solids, $\ell_{ec}$ can be as large as 1 mm. Capillary phenomena then become macroscopically visible at free surfaces: edges are rounded [12] and thin cylinders develop undulations reminiscent of the classical Plateau-Rayleigh instability for liquids [13]. Waves existing at the interface of soft materials have been only partially described so far. The existence of two regimes, dominated by elasticity or capillarity, theoretically predicted [14] and initially probed experimentally in the late 90s [15] has been at center of discussion [16, 17]. Recent work focussed on the transition between the two regimes, yet with limited resolution on the wave measurements [18]. In this letter, we propose to combine accurate wave field measurements and a theoretical analysis in order to discriminate the influence of capillarity on the propagation of mechanical waves at the free surface of soft gels.

Experimental setup. – We make agarose gels by heating a solution of water and agarose (Sigma A4550-500G) at 95 °C. The hot solution is then poured in a rectangular container with dimensions 8.5 cm × 26 cm and left to cool at room temperature for 2 hours. We determined the rheology of the hydrogels for concentrations of 2 g/L and 3 g/L which gives shear moduli, $\mu$, of respectively 95 Pa and 380 Pa (See Supplemental Material). We generate plane waves at the air/gel interface by locally imposing a vertical sinusoidal motion with frequency $f$ at the free surface of the sample. To do so, we deposit a rectangular magnet on the surface of the gel and activate it with an electromagnet, or alternatively we use a vibration exciter (figure 1a). We measure the out of plane displacement field generated at the interface using a Synthetic Schlieren Imaging (SSI) technique, based on the apparent displacement of a pattern placed below the tank caused by refraction at the surface [19]. We record from the top at a frame rate of 350 Hz for $f$ ranging from 10 Hz to 160 Hz. We use continuous sweeps at a rate of 1.6 Hz/s, which we checked to be small enough to consider the excitation as monochromatic when analysing small signal windows.

Dispersion relation. – We show in figure 1d a typical height field measured with this technique, obtained at $f = 40$ Hz in a gel with $\mu = 95$ Pa and depth $h = 1.1 \pm 0.1$ cm. We extract the wave field at any frequency by Fourier filtering a signal window around the corresponding $f$. We then use spatial 2D Fourier transforms to extract the spatial spectra along the direction of propagation that we normalize by their maximum amplitude. By stacking the spectra, obtained at each $f$, we can construct a map of the dispersion relation, which we show in figure 1e for a gel with $\mu = 380$ Pa and thickness $h = 3.4 \pm 0.4$ cm. The dispersion relation shows the co-existence of two distinct behaviors. (i) For $f < 120$ Hz, we observe multiple branches, which start at increasing cutoff frequencies. (ii) At higher frequencies, the different branches merge, and a single line is observed. We
interpret the presence of several cutoff frequencies (at $k_z = 0$) as a signature of the finite thickness: in a confined sample the vertical component of the wave vector can only take discrete values. We investigate this effect by decreasing $h$ to 1.1 ± 0.1 cm while keeping $\mu$ constant. The dispersion relation is shown in figure 1d. One can readily see the strong effect of the depth: there still are several branches but with markedly different cutoff frequencies. The fundamental mode appears at a higher frequency, as expected for a shallower gel. The following branches start existing at larger $f$ and are further apart. Then, we probe the effect of the gel’s elastic properties by decreasing the agarose concentration to obtain a gel with $\mu = 95$ Pa and $h = 1.1$ cm (fig. 1e). The cutoff frequencies are now lower, which shows the influence of $\mu$ on the position of the branches. We also see that the local slope of each branch is significantly smaller than that of the other two gels (fig. 1f and 1g). Plotting on figures 1c-e the dispersion relation of shear waves $w = k\sqrt{\mu/\rho}$ (black dashed-lines) [20] suggests that the local slope of the branches is controlled by the speed of elastic shear waves. Conversely, the slope of the single line observed at high frequency, is larger than that of shear waves. The dispersion relation can be regarded as an apparent dispersion curve whose group velocity progressively increases. An effect that hints at the presence of capillarity that could stiffen the interface at large wave numbers.

**In-depth displacements.** – Surface measurements suggest that the finite thickness of our samples has a strong influence on mode selection at low $f$. We further confirm this hypothesis by measuring in-depth displacement fields using Digital Image Correlation (DIC). We seed the gel with micro-particules (diameter ~ 10 $\mu$m and density 1100 kg/m$^3$) and illuminate a plane parallel to the direction of propagation using a laser sheet (figure 2a). We measure the local displacement field at 250 fps, using a standard DIC algorithm [21], in a window with dimensions 1.8 x 1.6 cm whose top left corner is approximately 2 cm away from the source. Figures 2b-c present a quiver plot of the displacement vector superimposed over a map of its magnitude for a gel with $\mu = 95$ Pa and $h = 1.9$ ± 0.1 cm excited at $f = 40$ Hz (fig. 3b) and $f = 120$ Hz (fig. 3c). The amplitude of the displacements is on the order of 10 $\mu$m, and both a vertical and a horizontal component are present. For $f = 40$ Hz, we observe displacements in the entire sample, without a significant decay in the vertical direction. The apparent square structure of the displacement field is caused by the interference between an incident shear wave and its reflection at the bottom (See Supplemental Material). At $f = 120$ Hz, the amplitude seems to decay faster in the vertical direction than in the horizontal direction so that propagation occurs mostly at the surface. The wave travelling downwards is damped before it reaches $z = -h$. These experiments reveal that the motion is not confined at the free surface and confirm that the multiple modes observed at low frequency result from the vertical confinement. At high frequency, they suggest that dissipation plays a significant role by preventing the incident wave to propagate all the way down to the bottom of the sample.
Both $\Phi$ and $H$ing advantage of the incompressibility of the hydrogels linearize the boundary conditions at $z$ 

At the free surface, assuming small deformations to transverse part by a vector potential $\Phi$ and the transverse divergence free contribution $u_3$. The longitudinal part can be described by a scalar potential $\Phi$ and the transverse part by a vector potential $H$.

$$u = u_x + u_3 = \nabla \Phi + \nabla \times H.$$ 

Both $\Phi$ and $H_y$ verify a wave equation [20]:

$$\nabla^2 \Phi - \frac{1}{c_l^2} \frac{\partial^2 \Phi}{\partial t^2} = 0, \quad \nabla^2 H_y - \frac{1}{c_l^2} \frac{\partial^2 H_y}{\partial t^2} = 0$$

where $c_l = \sqrt{\frac{\rho}{\mu}}$ and $c_t = \sqrt{\frac{\lambda + 2\mu}{\rho}}$ are, respectively, the shear and longitudinal wave speeds. We look for solutions of the form $\Phi = \Phi(x)e^{ikx-\omega t}$ and $H_y = H_y(x)e^{ikx-\omega t}$ and impose the following boundary conditions. (i) At the bottom of the sample, we assume that the gel is bounded to the container, so that:

$$u_x(z = -h) = u_z(z = -h) = 0.$$ 

(ii) At the free surface, assuming small deformations to linearize the boundary conditions at $z = 0$ and taking advantage of the incompressibility of the hydrogels ($c_l \to \infty$) that allows to absorb bulk gravity into the hydrostatic pressure, we impose:

$$\sigma_{xx}(z = 0) = 0, \quad \sigma_{zz}(z = 0) = \frac{\partial^2 u_z}{\partial x^2} - \rho gu_z.$$ 

Only the boundary condition at the interface sets this problem apart from the purely elastic one: capillarity and gravity are taken into account by respectively relating the Laplace and hydrostatic pressure to the normal stress. Using the four boundary conditions, and substituting the chosen solution forms for $\Phi$ and $H_y$, we obtain the dispersion relation for the gravito-elastocapillary waves (see Supplemental Material). This relation can be written in dimensionless form by introducing the variables $\tilde{k} = kh$ and $\tilde{\omega} = \omega h/c_l$:

$$\tilde{k}^2 \sin \tilde{\alpha} \sin \tilde{\beta} \left( \left( \tilde{k}^2 - \tilde{\beta}^2 \right)^2 + 4\tilde{\alpha}^2 \tilde{\beta}^2 \right) - \tilde{\alpha} \tilde{\beta} \cosh \tilde{\alpha} \cos \tilde{\beta} \left( 4\tilde{k}^4 + \left( \tilde{k}^2 - \tilde{\beta}^2 \right)^2 \right) + 4\tilde{\alpha} \tilde{\beta}^2 \tilde{k}^2 \tilde{k}^2 - \left( \Gamma + \frac{G}{k^2} \right) \tilde{\alpha} \tilde{k}^2 \tilde{k}^2 + \tilde{\beta}^2 \right) \left( \tilde{k}^2 \cosh \tilde{\alpha} \sin \tilde{\beta} + \tilde{\alpha} \tilde{\beta} \sinh \tilde{\alpha} \cos \tilde{\beta} \right) = 0 \quad (1)$$

where $\tilde{\alpha}^2 = -\tilde{k}^2$ and $\tilde{\beta}^2 = \tilde{\omega}^2 - \tilde{k}^2$. We identify two dimensionless parameters $\Gamma = \gamma/\mu h$ and $G = \rho gh/\mu$ that compare the elastocapillary length $\ell_{xc} = \gamma/\mu$ and the elastogravity length $\ell_{eg} = \mu/pg$ to the thickness $h$ of the sample. Using a secant method algorithm, we determine the zeros of equation (1) (assuming that the surface tension of the gels is similar to that of water, i.e. $\gamma = 70$ mN/m). We overlay the obtained curves on the experimental dispersion maps in figure 3b-c (red lines). The data of figure 3a-c are extracted from experiments where the thickness of the sample was precisely controlled so that only $\mu$ was used as an adjustable parameter in equation (1). The model is in good agreement with the measured data: it captures very well the existence of multiple modes/branches, their cutoff frequencies and local slope when we vary both $\mu$ and $h$. The values $\mu_{th}$ used to fit the predicted dispersion relations to the experimental data are always slightly larger than the expected $\mu$. We explain this discrepancy qualitatively by the evaporation of the hot agarose solution during cooling. Evaporation occurs over a larger surface in the samples than during rheology measurements which leads to increased agarose concentrations and thus to stiffer gels. Yet, we do not explain the apparent dispersion: the signal is localized only on a finite part of the predicted branches. We gain more insight by deriving the displacement field associated to each mode: for any couple $(\omega, k)$ verifying equation (1) we can compute the displacement field at the interface up to a multiplicative

![FIG. 2. (a) Sketch of the experimental setup used to measure in depth displacement fields. (b-c) Displacement field inside a gel with $\mu = 95$ Pa and $h = 2.3 \pm 0.3$ cm for (b) $f = 40$ Hz and (c) $f = 120$ Hz.](image-url)
constant (see Supplemental Material). We plot in figure 3 the norm of the vertical displacement normalized by the magnitude of the displacement vector at the interface, $|u_z|/|u|_{|z=0}$, as a function of $k$ (red lines) for the gel presented in figure 1e ($\mu_{th} = 120$ Pa and $h_{th} = 1.3$ cm). The normal displacement at $z = 0$ varies in a similar fashion for each mode: it increases sharply until it reaches a maximum for $k = k_m$ (red diamonds) and then decreases at a slower rate. As SSI only detects out of plane motion, we expect to measure waves only when $k > k_m$ and that the signal intensity decays along each mode as $k$ increases. We report with red symbols in figure 3e the couples $(\omega_m, k_m)$ obtained from the model for each sample. Our prediction now captures the apparent dispersion, the red diamonds act as lower bounds for the presence of signal for each mode. The gradual blurring of the modes into a single line can be qualitatively explained by the significant effect of dissipation at high frequency, an effect that would deserve a separate study.

**Elasto-capillary effect.** – Although the apparent dispersion suggests that surface tension plays a role at large $k$, the experimental values of $\Gamma$ range from 0.08 to 0.001 indicating that elasticity is the dominant force. To evidence the role of capillarity, we report in figure 3 the normalized vertical displacement at the interface for the same parameters as in figure 3 without taking into account surface tension. The variations of the out of plane displacement are markedly different at high $k$, where we now observe a plateau. This difference can be ascribed to the dominant role of capillarity at large wavenumbers (i.e. for $k\ell_{cc} > 1$). The nature of the dispersion fields is modified, reducing the relative weight of the out-of-plane contribution. Physically, there is an extra energetical contribution due to surface tension that tends to favour in-plane displacements. We also notice that the values of $(\omega_m, k_m)$ are shifted so that we no longer recover the apparent dispersion in figures 1d-e (blue circles); they align on a line with slope $\sqrt{2\mu}$, corresponding to Lamé modes (see Supplemental Material). It shows that the apparent dispersion is caused by capillarity for wavenumbers much lower than $1/\ell_{cc}$. To further probe the effect of surface tension, we investigate wave propagation in a very shallow sample ($\Gamma \sim 1/h$). We report in figure 4 the dispersion relation of a gel with $\mu = 95$ Pa and $h = 0.23 \pm 0.05$ cm for which $\Gamma = 0.4$. The red lines represent the prediction of equation 3. (a) $\mu = 380$ Pa, $h = 2.90 \pm 0.05$ cm, $\mu_{th} = 400$ Pa (b) $\mu = 380$ Pa, $h = 0.98 \pm 0.05$ cm, $\mu_{th} = 420$ Pa and (c) $\mu = 95$ Pa, $h = 0.99 \pm 0.05$ mm, $\mu_{th} = 120$ Pa. (d-e) Normalized vertical displacement $|u_z|/|u|$ at the interface plotted as a function of $k$ for the gel presented in 1e ($\mu_{th} = 120$ Pa and $h_{th} = 1.3$ cm) with (d, red lines) and without (e, blue lines) taking into account capillarity.

**Elasto-gravity effect.** – Finally, we check the influence of gravity on the dispersion relation. To do so, we measure wave propagation in a sample whose interface normal points upwards or downwards. In the first case, gravity acts as a restoring force on the free interface whereas in the second it tends to deform it and can even make it unstable. Figures 3 and 4 present the dispersion relations obtained for a sample.
FIG. 4. (a) Dispersion map obtained from a gel with $\mu = 95$ Pa and $h = 0.23 \pm 0.05$ cm. The red lines represent the prediction of equation 1 ($\mu = 95$ Pa, $h = 0.26$ cm) with (solid line) and without (dashed line) taking into account capillary effects. (b) Dispersion map for a gel with $\mu = 95$ Pa and $h = 0.99 \pm 0.05$ cm when the interface points downwards. The solid line (respectively dashed line) corresponds to the prediction of equation 1 ($\mu = 120$ Pa) with the interface pointing up (respectively down). In both cases the black dashed lines show the dispersion relation of shear waves: $\omega = k \sqrt{\frac{\mu}{\rho}}$.

with $\mu = 95$ Pa and $h = 0.99 \pm 0.05$ cm when the interface points respectively up or down. For such a sample $|G| = 1.02$, so that we expect to see the effect of gravitational forces but remain below the instability threshold. The model accurately predicts the influence of gravity as shown by the overlay of the red solid line (respectively dashed line) corresponding to the prediction of equation 1 ($\mu = 120$ Pa) with the free surface pointing up (respectively down). This experiment shows that by tuning $G$ below the value of the instability threshold, we can control the dispersion of the fundamental mode of the wave propagating at the surface of the gels.

Discussion. – In this letter, we use state-of-the-art measurement techniques to probe the propagation of surface waves in agarose gels with great accuracy, revealing the existence of multiple modes at low frequencies as well as an apparent dispersion. We quantitatively predict the dispersion relation using an elastic model including capillary forces. In particular, we capture the major role of surface tension, that alters the balance between in and out-of-plane interfacial displacements, even at wavenumbers lower than expected from simple scaling arguments. We confirm the validity of our approach by including gravity in the model and successfully testing it against experimental data. The influence of gravity opens new perspectives: $G$ can be tuned to take negative values creating materials in which the phase and group velocity have opposite signs, a sought-after property that allows perfect lensing [24]. Furthermore, $G$ also depends on depth enabling to tune the medium properties down to sub-wavelength scales and thus the creation of metamaterials for elastic wave control [25].

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