

# Competition between shear and biaxial extensional viscous dissipation in the expansion dynamics of Newtonian and rheo-thinning liquid sheets

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## ABSTRACT

When a drop of fluid hits a small solid target of comparable size, it expands radially until reaching a maximum diameter and subsequently recedes. In this work, we show that the expansion process of liquid sheets is controlled by a combination of shear (on the target) and biaxial extensional (in the air) deformations. We propose an approach toward a rational description of the phenomenon for Newtonian and viscoelastic fluids by evaluating the viscous dissipation due to shear and extensional deformations, yielding a prediction of the maximum expansion factor of the sheet as a function of the relevant viscosity. For Newtonian systems, biaxial extensional and shear viscous dissipation are of the same order of magnitude. On the contrary, for thinning solutions of supramolecular polymers, shear dissipation is negligible compared to biaxial extensional dissipation and the biaxial thinning extensional viscosity is the appropriate quantity to describe the maximum expansion of the sheets. Moreover, we show that the rate-dependent biaxial extensional viscosities deduced from drop impact experiments are in good quantitative agreement with previous experimental data and theoretical predictions for various viscoelastic liquids.

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## I. INTRODUCTION

Drop impact process on solid surfaces is an extremely active research area because of the development of high-speed imaging technology,<sup>1</sup> and its implication in many industrial applications, such as the impact of pesticide drops on plant leaves<sup>2</sup> or in inkjet printing.<sup>3</sup>

We can distinguish two main experimental configurations. On the one hand, drops are impacted on a flat solid surface with a size much larger than the drop size, in a way that the whole expansion event occurs on the target.<sup>4–10</sup> The sheet spreads thus in intimate contact with a uniform surface. Hence, viscous dissipation is mainly shear-induced. In that case, it has been reported that the shear rate-dependent viscosity is the pertinent parameter to describe the dissipation process in the expansion dynamics of non-Newtonian fluids.<sup>8,10–14</sup> Note, however, that the retraction dynamics was suggested

to be dominated by the nonlinear extensional viscosity for a drop of fluid containing polymer additives.<sup>15</sup>

The other distinguishable configuration, albeit sparsely studied, involved impacting drops on repellent surfaces where there is no contact with the surface, hence eliminating shear dissipation. Such surfaces may include superhydrophobic surfaces,<sup>16–19</sup> hot plates above the Leidenfrost temperature,<sup>20,21</sup> and, more recently, cold plates covered with liquid nitrogen to exploit the cold Leidenfrost effect.<sup>22–25</sup> We have demonstrated elsewhere<sup>24</sup> that, in the so-called viscous regime, biaxial extensional dissipation dominates the sheet dynamics produced using the cold Leidenfrost effect.

Nature and pertinent industrial problems are more complex than the above two extreme cases. Hence, we may consider a third scenario, where drops impact a solid target of size comparable to that of the

drop. In this case, a part of the expanding sheet is in contact with the target surface and the other part is expanding freely in the air, which, depending on the sample viscosity, may be the largest part of the sheet.<sup>26–36</sup> Although the situation of impacting drops on small targets was originally conceived to reduce the friction dissipation,<sup>26</sup> it turns out that it actually highlights the competition between shear dissipation (on the target) and biaxial extensional dissipation (in the air). In this work, we assess this competition by evaluating both dissipations (shear and biaxial extensional) for Newtonian and rheologically thinning non-Newtonian fluids. We show that, for Newtonian fluids, the shear and biaxial extensional dissipations are of the same order of magnitude. However, for the non-Newtonian thinning samples, the biaxial extensional dissipation is found to control the sheet expansion dynamics. We compare our data with experimental measurements of biaxial extensional viscosity of thinning fluids<sup>24,37,38</sup> and model semi-quantitatively the sheet expansion dynamics using a biaxial extensional thinning viscosity.

## II. MATERIALS AND METHODS

### A. Materials

#### 1. Newtonian fluids

We investigate two classes of Newtonian fluids, silicone oils, and mixtures of water and glycerol. Silicone oils, with zero-shear-rate viscosities from 4.5 to 339 mPa s, an average surface tension of 20 mN/m,<sup>39</sup> and densities ranging from 0.913 to 0.97 g/mL, are purchased from Sigma-Aldrich and used as received. Glycerol/water mixtures with concentrations ranging from 22 to 100% g/g glycerol are used, yielding zero-shear rate viscosities from 1.7 to 1910 mPa s, densities from 1.05 to 1.25 g/mL,<sup>40</sup> and an average surface tension of 65 mN/m (as measured with a pendant drop setup).

#### 2. Non-Newtonian fluids

As non-Newtonian system, we choose wormlike micellar solutions (WLM) made of 2,4-bis(2-ethylhexylureido)toluene, abbreviated as EHUT, dispersed in dodecane. This monomer has the ability to self-associate by means of hydrogen bonding in an apolar solvent (dodecane) forming supramolecular polymers.<sup>41–47</sup> As schematically shown in Fig. 1, EHUT molecules self-assemble into small and long rod-like structures whose cross section is solvent- and temperature-dependent. Above critical concentration and temperature, the monomers self-assemble into thin filaments with a diameter (1.3 nm) comparable to the size of one EHUT monomer. At lower temperatures, thick tubes with a cross section equivalent to about three EHUT monomers form. In this work, we investigate samples in the tube region of the phase diagram, in the temperature range between 20 °C and 25 °C, and concentration range from  $C = 0.37$  to  $C = 3$  g/L, where the tubes are long enough to entangle, allowing us, hence, to explore a wide range of viscoelastic properties. Although this phase diagram is made for EHUT supramolecular assemblies formed in toluene, Fourier transform infrared spectroscopy (FTIR), small-angle neutron scattering (SANS), and rheology studies confirm that the range of temperatures and concentrations investigated here also corresponds to the tube region of the EHUT assemblies formed in dodecane.<sup>43–45</sup> The surface tension for EHUT solutions is assumed to be independent of concentration. We take  $\Gamma = 25 \pm 2$  mN m<sup>-1</sup> as measured with a pendant drop experiment for a dilute sample ( $C = 0.37$  g/L). The density

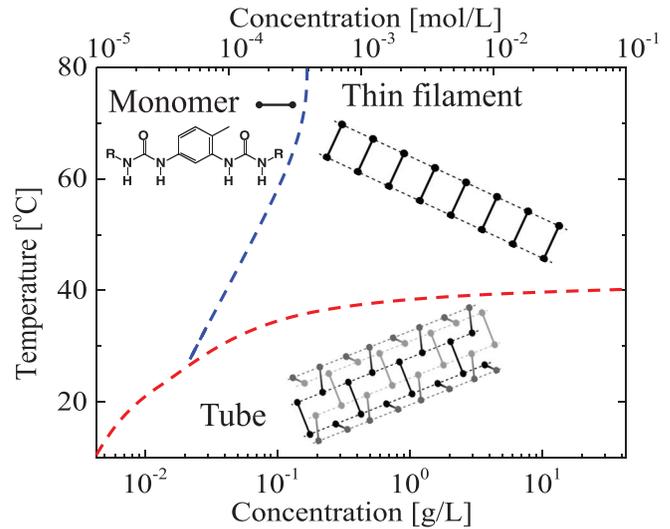


FIG. 1. Phase diagram for EHUT solutions in toluene<sup>43</sup> showing the transition between monomers and supramolecular filaments. The structure of the EHUT monomer is shown, along with a schematic illustration of the various aggregates formed (out of scale), that is, filaments and tubes. Hydrogen bonds are represented by dotted lines connecting the urea functions (black and gray circles).

of the EHUT solutions is assumed to be equal to the density of the solvent,  $\rho = 0.75$  g/mL.

### B. Methods

#### 1. Solution rheology

Linear and nonlinear shear rheology measurements are performed with an MCR501 stress-controlled rheometer (Anton Paar, Austria), operating in the strain control mode and equipped with a stainless-steel cylindrical Couette geometry. Temperature control ( $\pm 0.2$  °C) is achieved by means of a Peltier element.

For Newtonian and non-Newtonian samples, the zero-shear-rate viscosity,  $\eta_0$ , is measured by applying a ramp of steady shear rate varying from 0.01 to 1000 s<sup>-1</sup>.

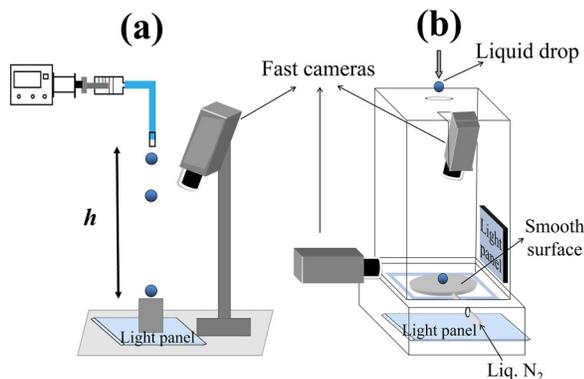
For EHUT solutions, the linear viscoelastic spectra are obtained by applying a small-amplitude sinusoidal strain ( $\gamma = 10\%$ ) with varying angular frequency,  $\omega$ , from 100 to 0.01 rad/s, and measuring the storage,  $G'(\omega)$ , and loss,  $G''(\omega)$ , moduli. The complex viscosity is also calculated from the linear viscoelastic spectra as  $|\eta^*(\omega)| = \frac{\sqrt{(G'(\omega))^2 + (G''(\omega))^2}}{\omega}$ . All experiments are performed in open air without any particular humidity precaution (the relative humidity is about 40%). Since hydrogen bonding systems such as EHUT self-assemblies are very sensitive to humidity,<sup>48,49</sup> and because we do not control the drop impact environmental conditions, we ensure that both bulk rheology and drop impact experiments are conducted under the same temperature and humidity conditions.

#### 2. Drop impact experiment

To study the competition between shear and biaxial extensional viscous dissipations, two drop impact experimental setups are used.

The main setup based on the impact on a small cylindrical target was originally designed by Vignes-Adler *et al.*<sup>26</sup> and subsequently modified by several groups.<sup>30,31,50</sup> We use the same setup as in,<sup>31,32</sup> schematically shown in Fig. 2(a). In brief, a hydrophilic glass target of diameter  $d_T = 6.5$  mm is fixed on top of an aluminum rod with the same diameter. The liquid is injected from a syringe pump with a flow rate of 1 ml/min through a needle placed vertically above the target. The initial diameter of the falling drop is constant,  $d_0 = 3.9 \pm 0.2$  mm, as measured by image analysis and by weighting the drops. Note that, considering the difference in fluids' surface tensions, we have adapted the needle diameter to produce a constant drop diameter (a needle with a diameter of 2 mm for fluids with a surface tension of 65 mN/m and a needle diameter of 4 mm for fluids with  $\Gamma = 20\text{--}25$  mN/m). The drop falls from a height  $h = 91$  cm, yielding an impact velocity  $v_0 = \sqrt{2gh} = 4.2$  m s<sup>-1</sup> ( $g$  is the acceleration of gravity). The corresponding Weber numbers,  $We = \frac{v_0 \rho d_0}{\Gamma}$ , are  $We \sim 1200$  for glycerol/water mixtures,  $We \sim 3400$  for silicone oils, and  $We \sim 2400$  for EHUT-based samples. The target is mounted on a transparent Plexiglas plate, illuminated from below by a high-luminosity backlight (Phlox LLUB, luminance of 20 cd/m<sup>2</sup>). The drop impact is recorded from the top using a high-speed camera (Phantom V 7:3) operating at 6700 frames/s with a resolution of  $800 \times 600$  pixels<sup>2</sup>. The angle between the camera axis and the horizontal plane is fixed at about 10°.

Additional experiments are performed with Newtonian fluids under cold Leidenfrost conditions.<sup>22</sup> The relevant setup is described elsewhere<sup>24</sup> and schematized in Fig. 2(b). In brief, the drop maintained at room temperature impacts a smooth surface (polished silicon wafer or quartz slide with a diameter larger than the maximum sheet diameter) covered with a thin layer of liquid nitrogen (temperature  $T = -196$  °C). Upon impact, a vapor cushion forms at the liquid interface due to the evaporation of N<sub>2</sub>, providing a unique scenario of non-wetting and slip conditions that eliminates shear viscous dissipation.<sup>22,51</sup> Details on the heat transfer involved in the impact of drops in cold Leidenfrost conditions have been reported elsewhere,<sup>25</sup> and it does not change significantly the temperature of the drops during the expansion phase of the sheet.



**FIG. 2.** (a) Schematic illustration of the main experimental setup configuration, consisting of a drop impacting a small solid target as described in the text. (b) Schematic illustration of the second experimental setup showing a drop falling on a liquid nitrogen thin layer. The expansion event is recorded using two fast cameras allowing concomitant top and side visualizations.

### 3. Image analysis

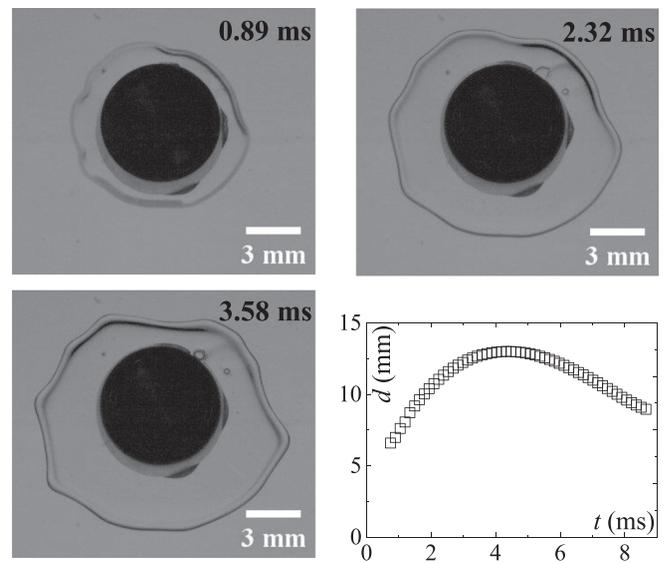
The time evolution of the diameter of the sheet is measured with ImageJ software. We first subtract the background image from the expansion movie and highlight the rim by a binary thresholding. This allows us to determine the contour of the sheet and measure its area  $A$ . An apparent sheet diameter is simply deduced as:  $d = \sqrt{\frac{4A}{\pi}}$ . The results are obtained by averaging for each sample the time evolution of the sheet diameter from three different experiments. Note that for the small target setup, the dark black central disk is the target and precludes to study the expansion of sheets that do not cross the edge of the target. For the cold Leidenfrost setup, in addition to the top view images, systematic side view images are also collected, though not shown here, to correct the maximum expansion diameter for the corona effect observed for the low viscosity samples (viscosity smaller than 100 mPa s), as detailed in Ref. 24.

## III. RESULTS AND DISCUSSION

### A. Newtonian fluids

#### 1. Drop impact experiments

Once hitting the target, the drop spreads first on the target and then expands freely in the air until reaching a maximum expansion. It then retracts because of surface tension and eventually bulk elasticity for non-Newtonian fluids (representative videos of the impact process are available in the multimedia). This general phenomenology has been observed for many types of fluids impacting a small solid target.<sup>26,30–32,34,50,52</sup> The overall behavior is illustrated in Fig. 3



**FIG. 3.** Snapshots taken at different times (as indicated on each image) during the expansion of the sheet for a glycerol/water mixture with a viscosity  $\eta_0 = 160$  mPa s produced upon impact on a solid target with a diameter  $d_T = 6.5$  mm. The maximum expansion occurs at  $t_{Max} = 3.58$  ms. The black area represents the surface of the target. Video of a drop of glycerol and water mixture (160 mPa s) impacting on a target of diameter 6.5 mm at an impact velocity of 4.2 m/s. The impact was filmed with a high-speed camera operating at 6700 fps and has been slowed down 200 times for the videos. Multimedia view: <https://doi.org/10.1063/5.0057316.1>

(Multimedia view) that depicts snapshots of the drop after its impact for a Newtonian sample (glycerol/water) with  $\eta_0 = 160 \pm 10$  mPa s at different times. In the bottom right of Fig. 3, we show the corresponding time evolution of the effective sheet diameter,  $\tilde{d}$ , where the expansion and retraction regimes can be clearly determined (we choose for the origin of time, the time when the drop hits the target). Similarly, in Fig. 4 (Multimedia view), we present snapshots for the same Newtonian sample at different times, when it expands and retracts under cold Leidenfrost conditions obtained by impacting a drop on a thin layer of liquid N<sub>2</sub>. The origin of time is set when the droplet starts expanding on the nitrogen vapor cushion.

Despite qualitatively similar behavior, we find that, for a same sample and a same impact velocity, the maximum expansion diameter of the sheet,  $d_{Max}$ , is significantly larger under cold Leidenfrost conditions ( $d_{Max} = 34.6$  mm) than with the small target ( $d_{Max} = 12.9$  mm). When the expansion results from the impact on the target, only the inner portion of the sheet is at intimate contact with the target surface, while its outer part is free in the air, resulting in a combination of two potential sources of viscous dissipation: shear on the target and biaxial extensional everywhere but predominantly in air. The situation is less complex when the sheet is produced under cold Leidenfrost conditions because the sheet is expanding on the liquid nitrogen vapor, ensuring biaxial extensional deformation as the unique source of viscous dissipation.<sup>24</sup>

The cold Leidenfrost conditions allow also the observation of the sheet central region, usually obscured by the target. It is worth noting

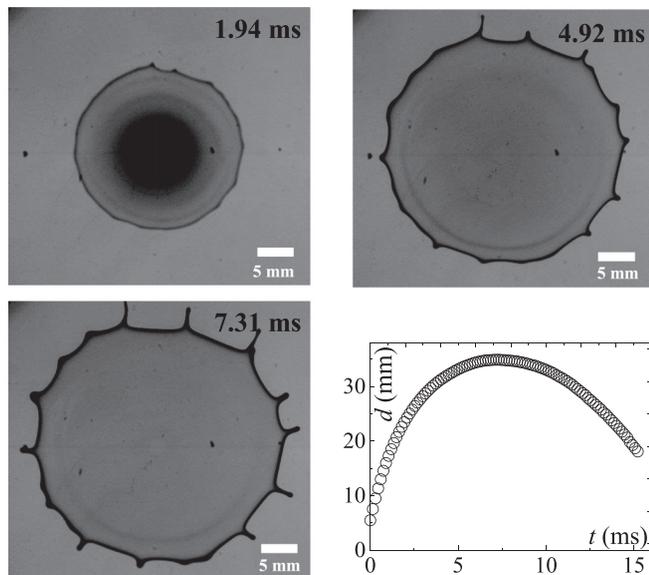


FIG. 4. Snapshots taken at different times (as indicated on each image) during the expansion of the sheet for a glycerol/water mixture with a viscosity  $\eta_0 = 160$  mPa s produced upon impact under cold Leidenfrost conditions. The maximum expansion occurs at  $t_{Max} = 7.31$  ms. Video of a drop of glycerol and water mixture (160 mPa s) impacting on liquid nitrogen at an impact velocity of 4.2 m/s and video of a drop of EHUT solution ( $C = 0.37$  g/L) impacting a target of diameter 6.5 mm at an impact velocity of 4.2 m/s. The impact was filmed with a high-speed camera operating at 6700 fps and has been slowed down 200 times for the videos. Multimedia view: <https://doi.org/10.1063/5.0057316.2>

the occurrence of a thicker rim for the sheet when impacted on the target and, on the other hand, the appearance of a larger number of better defined fingers instabilities under cold Leidenfrost conditions. However, we will not discuss further these two interesting features, which are out of the scope of this work. Moreover, in the rest of the paper, we will focus on the sheets expansion dynamics up to their maximum expansion and not consider the retraction regime.

In order to follow the expansion dynamics of different samples, we focus on the maximum expansion diameter  $d_{Max}$ . We use a normalized maximum expansion diameter,  $\tilde{d}$ , adopting the same definition as in Refs. 24 and 32:

$$\tilde{d} = \frac{d_{Max}}{d_{Max}^{cap}} \quad (1)$$

Here,  $d_{Max}$  is the maximum diameter of the sheet and  $d_{Max}^{cap}$  is the maximum expansion diameter in the capillary regime (at low viscosity), where viscous dissipation does not reduce significantly the maximum expansion diameter. This normalized quantity allows us to compare drops with different initial diameters  $d_0$  and different surface tensions. In addition, to account for the variation of surface tensions among samples, the data are plotted against the dimensionless zero-shear-rate Ohnesorge number, which expresses the ratio of viscous forces to inertial and surface tension forces:  $Oh_0 = \frac{\eta_0}{\sqrt{\rho\Gamma d_0}}$ , with  $\rho$  the

sample density,  $\Gamma$  the surface tension, and  $\eta_0$  the zero-shear-rate viscosity. Results for different Newtonian fluids obtained using two experimental conditions (target and cold Leidenfrost) are presented in Fig. 5. Note that, for the more viscous glycerol/water mixtures ( $\eta_0 > 200$  mPa s), experiments on the solid target are not shown because the maximum expansion diameter is smaller than the target diameter itself. We find two regimes for both experimental configurations. The capillary regime where the dynamics is independent of viscosity resulting in a constant  $\tilde{d} \approx 1$ , and the viscous regime, where

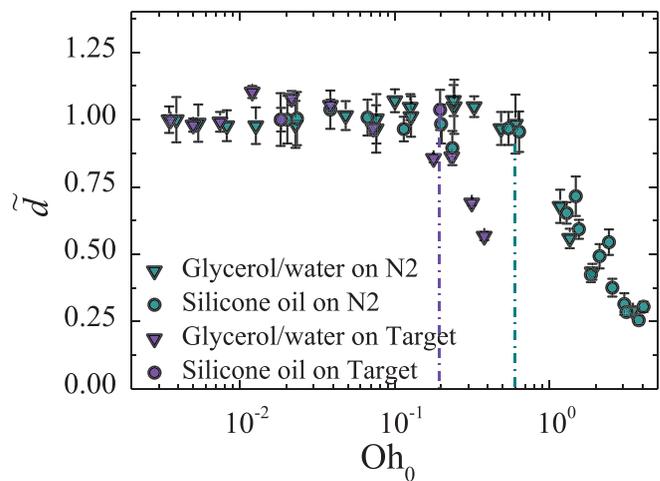


FIG. 5. Normalized maximum expansion as a function of the zero-shear-rate Ohnesorge number,  $Oh_0$ , for the two classes of Newtonian samples, as indicated in the legend, and for the two experimental configurations: small solid target (purple symbols) and cold Leidenfrost conditions (green symbols). Dotted lines indicate the onset of the viscous regime for each configuration. For the Newtonian samples (glycerol/water) presented in Figs. 3 and 4,  $Oh_0 = 0.31$ .

$\tilde{d}$  decreases with increasing viscosity. Importantly, the transition from capillary to viscous regime occurs at a larger  $Oh_0$  number (higher viscosity) for the sheets produced under cold Leidenfrost conditions ( $Oh_0 \approx 0.6$ ) compared to those produced by the impact on a target ( $Oh_0 \approx 0.2$ ). This implies that viscous dissipation is more important with the target setup than with the cold Leidenfrost setup, as exemplified by comparing the expansion dynamics of the Newtonian sample presented in Figs. 3 and 4.

In addition, Fig. 5 confirms the fact that even in the absence of friction on a solid surface, a dissipation regime exists resulting in the decrease in  $\tilde{d}$  with  $Oh_0$ . In this configuration, the deformation of the sheet is biaxial extensional.<sup>24</sup> The dissipation on a small solid target is by contrast more complex because it results from the contact with a solid surface and from free expansion in the air (similar to that under cold Leidenfrost conditions). Thus, in Sec. III A 2, we will analyze the contribution of each dissipation mechanism to the expansion dynamics of a Newtonian drop impacting a small solid surface.

## 2. Viscous dissipation processes

*a. Shear dissipation energy.* To estimate the shear dissipation energy, we first define the shear rate experienced by the sheet during its expansion on the small solid target. We approximate the expanding sheet by a disk. The shear flow velocity field on the target<sup>53,54</sup> reads

$$\begin{cases} v_r = \frac{2}{RH} \frac{dR}{dt} rz & \text{(radial),} \\ v_z = \frac{-2}{RH} \frac{dR}{dt} z^2 & \text{(axial),} \end{cases} \quad (2)$$

where  $R(t) = \frac{d(t)}{2}$  is the radius of the expanding sheet,  $r$  denotes the radial direction,  $z$  denotes the axial direction, and  $H(t) = \frac{d_0^3}{6R(t)^2}$  is the mean thickness of the expanding sheet in the disk shape approximation with  $d_0$  the drop diameter. The instantaneous shear rate reads

$$\dot{\gamma} = \frac{1}{2} \left( \frac{\partial v_r}{\partial z} + \frac{\partial v_z}{\partial r} \right) = \frac{1}{RH} \frac{dR}{dt} r. \quad (3)$$

The viscous shear energy dissipated,  $E_S$ , during the sheet expansion process can be written as

$$E_S \approx \int_0^{t_{\max}} \int_V \sigma_S \dot{\gamma} dV dt, \quad (4)$$

where  $V$  denotes the sheared volume and  $\sigma_S$  is the shear stress. During the expansion of the drop sheet, the viscous boundary layer thickness,  $\delta(t) = \alpha \sqrt{\frac{\eta_S t}{\rho}}$  ( $\alpha$  is an unknown prefactor and  $\eta_S = \eta_0$  is the shear viscosity), which quantifies the thickness of the sheet that is actually sheared.<sup>32,55,56</sup> At a short time,  $\delta < H$ , but the viscous boundary layer grows with time, while the film thickness,  $H(t)$ , decreases. Hence, at some time  $\delta$  exceeds  $H$  and the relevant thickness to be considered is  $H$  and not  $\delta$  anymore.

For simplicity, here, we consider the two limits for the viscous shear energy: (i)  $E_{S,H}$ , where the relevant thickness for the sheared volume is  $H(t)$ ; and (ii)  $E_{S,\delta}$ , where the relevant thickness for the sheared volume is  $\delta(t)$ .  $E_{S,H}$  reads

$$\begin{aligned} E_{S,H} &= \int_0^{t_{\max}} dt \int_0^{d_T/2} \eta_S \dot{\gamma}^2 2\pi r H dr \\ &= \frac{3\pi\eta_S d_T^4}{16d_0^3} \int_0^{t_{\max}} \left( \frac{dR}{dt} \right)^2 dt. \end{aligned} \quad (5)$$

With the approximations  $\int_0^{t_{\max}} \left( \frac{dR}{dt} \right)^2 dt \approx t_{\max} \langle \left( \frac{dR}{dt} \right)^2 \rangle$ ,  $t_{\max} \approx \frac{d_{\max}}{v_0}$ , and  $d_{\max} \gg d_0$ ,  $E_{S,H}$  reads

$$E_{S,H} \approx \frac{3\pi\eta_S d_T^4 v_0}{64d_0^3} d_{\max}. \quad (6)$$

On the other hand,  $E_{S,\delta}$  reads

$$\begin{aligned} E_{S,\delta} &= \int_0^{t_{\max}} dt \int_0^{d_T/2} \eta_S \dot{\gamma}^2 2\pi r \delta dr \\ &= \frac{9\pi\alpha\eta_S^{3/2} d_T^4}{8\sqrt{\rho} d_0^6} \int_0^{t_{\max}} R^2 \left( \frac{dR}{dt} \right)^2 t^{1/2} dt. \end{aligned} \quad (7)$$

With the approximations  $\int_0^{t_{\max}} R^2 \left( \frac{dR}{dt} \right)^2 t^{1/2} dt \approx t_{\max} \left\langle \frac{1}{4} \left( \frac{dR}{dt} \right)^2 \right\rangle \langle t^{1/2} \rangle$ ,  $t_{\max} \approx \frac{d_{\max}}{v_0}$ , and  $d_{\max} \gg d_0$ ,  $E_{S,\delta}$  reads

$$E_{S,\delta} \approx \frac{3\pi\alpha\eta_S^{3/2} d_T^4 v_0^{1/2}}{256\sqrt{\rho} d_0^{5/2}} \left( \frac{d_{\max}}{d_0} \right)^{7/2}. \quad (8)$$

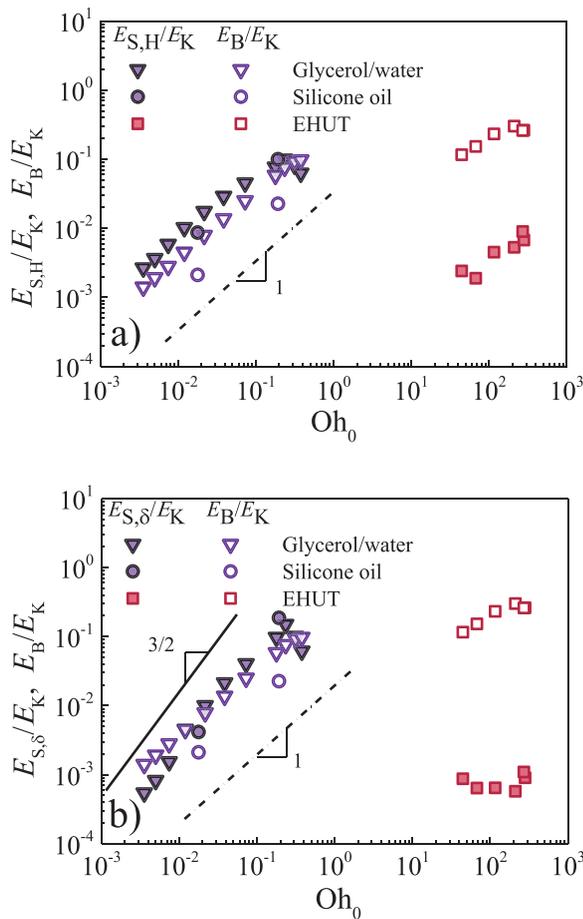
*b. Biaxial extensional dissipation energy.* A typical example of a biaxial extensional flow is the compression of a sample sandwiched between two (continuously) lubricated surfaces.<sup>57</sup> The biaxial viscosity is defined as  $\eta_B = \frac{\sigma_{rr} - \sigma_{zz}}{\dot{\epsilon}}$ , where  $\dot{\epsilon}$  is the strain rate, and  $\sigma_{rr}$  and  $\sigma_{zz}$  are the stress tensor components in cylindrical coordinates.<sup>57</sup> The stress tensor difference  $\sigma_{rr} - \sigma_{zz}$  is commonly expressed as the biaxial stress  $\sigma_B$ . Hence, the biaxial viscosity reads  $\eta_B = \frac{\sigma_B}{\dot{\epsilon}}$ . For a Newtonian fluid,  $\eta_B = 6\eta_S$ .

The biaxial extensional energy,  $E_B$ , dissipated during the expansion of the sheet can be written as<sup>24</sup>

$$\begin{aligned} E_B &= \int_0^{t_{\max}} dt \int_V \sigma_B \dot{\epsilon} dV \\ &\approx \eta_B \frac{\pi d_0^3}{6} \int_0^{t_{\max}} \left( \frac{1}{d} \frac{\partial d}{\partial t} \right)^2 dt \\ &\approx \eta_B \frac{\pi d_0^3}{6} t_{\max} \left\langle \left( \frac{\partial \ln(d)}{\partial t} \right)^2 \right\rangle \\ &= \eta_B \frac{\pi v_0 d_0^3}{6} \frac{\ln^2 \left( \frac{d_{\max}}{d_0} \right)}{d_{\max}}, \end{aligned} \quad (9)$$

where  $\dot{\epsilon} = \frac{1}{d} \frac{\partial d}{\partial t}$  is the biaxial extensional strain rate.

*c. Comparison between shear and biaxial extensional viscous dissipations.* In order to assess the respective roles of viscous dissipation due to shear and biaxial extensional deformations during the expansion of a drop, we compare in Fig. 6 the variations of the biaxial extensional viscous energy  $E_B$  [Eq. (9)] with the two limits for the shear viscous dissipation energy  $E_{S,H}$  [Eq. (6)] [Fig. 6(a)] and  $E_{S,\delta}$  [Eq. (8)] [Fig. 6(b)]. The dissipation energies have been normalized by the initial kinetic energy of the drop  $E_K = \frac{1}{2} m v_0^2$ , with  $m$  the mass of the drop, and plotted as a function of the zero-shear rate Ohnesorge



**FIG. 6.** Variations of the biaxial extensional viscous energy  $E_B$  calculated from Eq. (9) and the two limits of the shear viscous dissipation energy, respectively, (a)  $E_{S,H}$  from Eq. (6), and (b)  $E_{S,\delta}$  from Eq. (8) during the expansion of a drop impacting a small solid target, as a function of the zero-shear rate Ohnesorge number,  $Oh_0$ , for the two classes of Newtonian fluids (purple symbols) and EHUT supramolecular polymer solutions (red squares). The dissipation energies have been normalized by the initial kinetic energy of the drop  $E_K = \frac{1}{2}mv_0^2$ . The dashed and solid lines indicate a slope of 1 and 1.5, respectively.

number,  $Oh_0$ . We find that  $E_{S,H}$  and  $E_{S,\delta}$  exhibit power law evolution with the viscosity, which mirrors their explicit dependence with  $\eta_0$ ,  $E_{S,H} \sim \eta_0$  [Eq. (6)] and  $E_{S,\delta} \sim \eta_0^{3/2}$  [Eq. (8)], reflecting the fact that the dependence of  $d_{max}$  with viscosity is much weaker. This is also the case for  $E_B$ , which increases as a power law with  $\eta_0$ ,  $E_B \sim \eta_0$  [Eq. (9)], since  $\eta_B = 6 \eta_0$  for Newtonian fluids.

Moreover,  $E_{S,\delta}$  and  $E_{S,H}$  are of the same order of magnitude. Hence, for convenience, we choose the simplest one,  $E_{S,H}$  for further discussion on the competition between shear and biaxial extensional viscous dissipation. From Eqs. (6) and (9), we can compute the ratio of the two sources of viscous dissipation energy as

$$\frac{E_B}{E_S} = \frac{64}{3} \left( \frac{d_0}{d_{Max}} \right)^2 \left( \frac{d_0}{d_T} \right)^4 \ln^2 \left( \frac{d_{max}}{d_0} \right). \quad (10)$$

This ratio strongly depends on the drop-to-target size ratio,  $\frac{d_0}{d_T}$ . It does not depend explicitly on  $\eta_0$ , but indirectly because of the dependence of the maximal expansion diameter on the sample viscosity. Furthermore, as evidenced by the superposition of  $E_{S,H}/E_K$  and  $E_B/E_K$  in Fig. 6,  $\frac{E_B}{E_{S,H}}$  is close to one and, as expected, depends only weakly on  $\eta_0$ .

### B. Viscoelastic supramolecular polymers

In this section, we present the results for non-Newtonian fluids. In contrast to their Newtonian counterparts, these samples have shear and biaxial extensional viscosities that depend on the applied deformation rate.

#### 1. Viscoelasticity

Figure 7(a) shows the dynamic moduli ( $G'$ ,  $G''$ ) as a function of oscillatory frequency ( $\omega$ ) for some representative concentrations. As discussed previously,<sup>44</sup> these systems behave as Maxwell fluids at low frequencies. This is confirmed by the good agreement between the data and the Maxwell fits [lines in Fig. 7(a)] where  $G' = G_0 \frac{\omega^2 \tau_0^2}{1 + \omega^2 \tau_0^2}$  and  $G'' = G_0 \frac{\omega \tau_0}{1 + \omega^2 \tau_0^2}$ , with  $G_0$  being the elastic modulus and  $\tau_0$  the characteristic relaxation time. In Fig. 7(b), we plot the relevant viscoelastic quantities ( $G_0$ ,  $\tau_0$ , and  $\eta_0$ ) for the range of concentrations studied here, extracted from the linear viscoelastic spectra. We find that these quantities follow scaling laws with concentration ( $G_0 \sim C^{1.9}$ ,  $\tau_0 \sim C^{0.8}$ , and  $\eta_0 \sim C^{2.6}$ ) in agreement with.<sup>46</sup>

Figure 7(c) depicts the complex viscosity,  $|\eta^*|$ , as a function of frequency, along with the steady shear viscosity,  $\eta_S(\dot{\gamma})$ , as a function of shear rate. The collapse of the dynamic and steady data (especially above 0.37 g/L) validates the Cox–Merz rule.<sup>58</sup> We describe the data with a fit by means of the Cross model,<sup>59</sup> one of many options for empirical models, all giving comparable results:

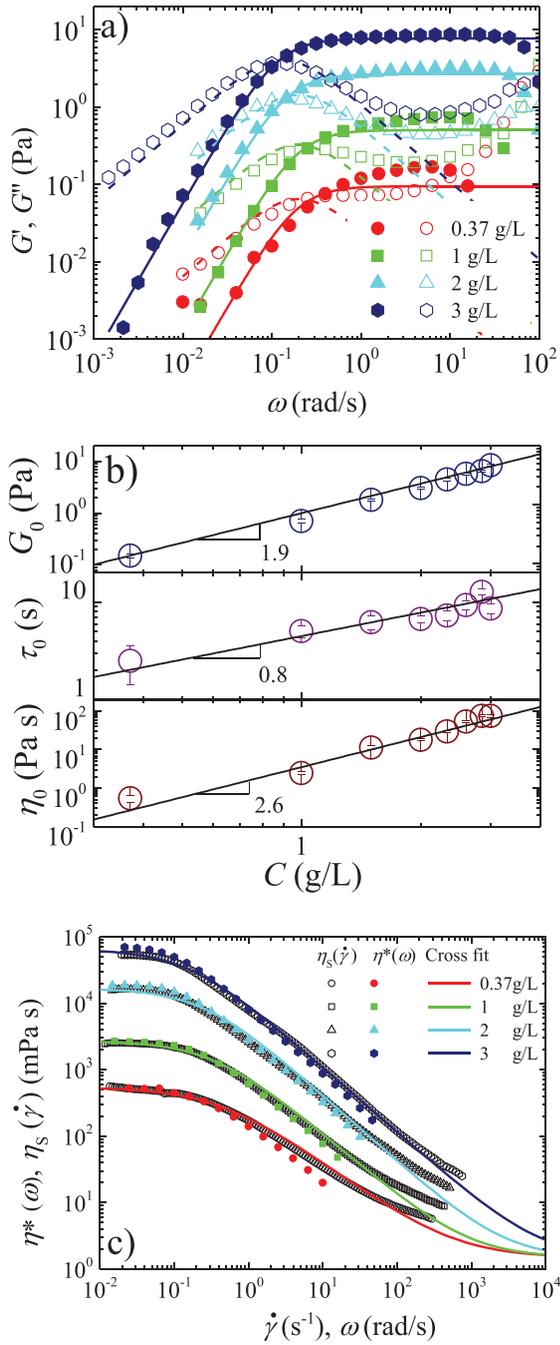
$$\eta_S(\dot{\gamma}) = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + (k\dot{\gamma})^n}. \quad (11)$$

Here,  $\eta_\infty$  is the viscosity at very large shear rate that we set equal to the solvent viscosity ( $\eta_\infty = 1.5$  mPa s),  $\eta_0$  is the zero-shear-rate viscosity [plotted in Fig. 7(b)],  $n$  is the shear-thinning exponent, and the parameter  $k$  is the inverse of a critical shear rate.<sup>57</sup> We find that the Cross model provides a good description of the samples' shear-thinning behavior [continuous lines in Fig. 7(c)], except at very large shear rates where some deviation is systematically measured. All data can be fitted with a very similar shear-thinning exponent  $n = 0.97 \pm 0.05$ .

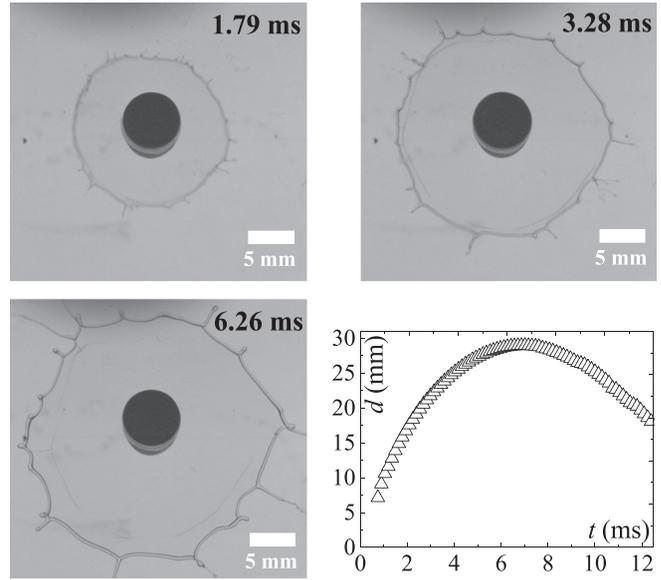
#### 2. Drop impact experiments

Figure 8 (Multimedia view) depicts snapshots of the drop after its impact for an EHUT solution with  $C = 0.37$  g/L at different times. The general phenomenology that has been observed for Newtonian samples (see Figs. 3 and 4) is also found here, and the expansion process does not exhibit any pertinent qualitative differences compared to that of Newtonian samples. This suggests common physical processes driving the expansion dynamics of sheets of different kinds of systems.

In Fig. 9,  $\bar{d}$  is plotted as a function of the zero-shear-rate Ohnesorge number,  $Oh_0$ , for both EHUT solutions (red half plain

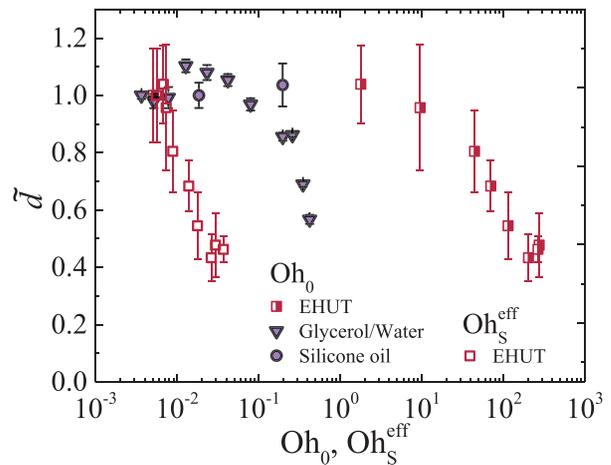


**FIG. 7.** (a) Frequency dependence of the storage ( $G'$ , filled symbols) and loss ( $G''$ , open symbols) moduli of different EHUT supramolecular polymer solutions. The lines (solid for  $G'$ , dashed for  $G''$ ) are Maxwell model fits for samples with different concentrations as indicated in the legend. (b) Evolution of the storage modulus (top), terminal relaxation time (middle), extracted from the Maxwell fits, and zero-shear-rate viscosity (bottom) with sample concentration. The concentration dependence is indicated by the slopes in each figure. (c) Complex viscosity as a function of frequency (closed symbols) and steady shear viscosity as a function of shear rate (open symbols), and fits (lines) using the Cross equation [Eq. (11)], for samples with different concentrations as indicated in the legend.



**FIG. 8.** Snapshots taken at different times (as indicated on each image) during the expansion of a sheet for EHUT sample with concentration  $C = 0.37$  g/L. Multimedia view: <https://doi.org/10.1063/5.0057316.3>

squares) and Newtonian samples (green plain triangles) impacted on a small target. We find that for the thinning fluids, the onset of the viscous regime takes place at much higher  $Oh_0$ , hence at much larger  $\eta_0$  (by factor of 100) as compared to Newtonian samples, a clear indication that the zero-shear-rate viscosity, is not the relevant quantity to characterize the dissipation process for viscoelastic fluids. We first argue that elasticity does not play an important role in the expansion dynamics of the present viscoelastic sheets. Indeed, even if the characteristic relaxation time of the EHUT is always much larger than the



**FIG. 9.** Normalized maximum expansion factor  $\tilde{d}$  as a function of the zero-shear-rate Ohnesorge number,  $Oh_0$ , for an EHUT supramolecular polymer solutions (closed squares) and for the Newtonian water/glycerol mixtures and silicone oils (purple symbols) and of the effective shear Ohnesorge number,  $Oh_S^{eff}$  (see the text), for the EHUT supramolecular polymer solutions (open squares).

characteristic time of the impact experiment [see the regime where storage modulus overwhelms the loss modulus in Fig. 7(a)] so that in principle elasticity should play a role in drop impact experiments, bulk elastic energy contribution is negligible with respect to the capillary contribution if the elastocapillary length, defined as  $l_{ec} = \frac{3\Gamma}{G_0}$ , is larger than the drop diameter  $d_0$ .<sup>23</sup> Taking  $G_0 = 8.5$  Pa,  $\Gamma = 25$  mN/m, and  $d_0 = 4$  mm, for the most concentrated sample investigated [Figs. 7(a) and 7(b)], one gets  $l_{ec}/d_0 = 2.2 > 1$ . This ratio is even larger for less concentrated polymer solutions. Therefore, bulk elastic energy contributions can be safely neglected in the present framework. Hence, the contrasted results for Newtonian and non-Newtonian samples must stem from the thinning character of EHUT solutions. Actually, different groups have successfully accounted for the shear-thinning upon impact on a solid surface with a size larger than the expanding maximum sheet diameter.<sup>8,11,13,32</sup>

In our case, the non-stationary shear rate experienced by the part of the expanding liquid sheet in contact with the target can be analyzed as following. We estimate the spatial average shear rate  $\dot{\gamma}(\bar{r})$  experienced by the sheet on the target at a given time  $t$ . We assume a constant thickness  $H(t)$  throughout  $r$  at a given time. We define by  $t^*$ , the time at which the expanding sheet reaches the edge of the target, that is,  $R(t^*) = R_T = \frac{d_T}{2}$ .

For  $t < t^*$ ,

$$\dot{\gamma}(\bar{r}) = \frac{1}{\pi R^2(t)} \int_0^{R(t)} \frac{1}{R(t)H(t)} \frac{dR}{dt} 2\pi r^2 dr = \frac{4R^2(t)}{d_0^3} \frac{dR}{dt}. \quad (12)$$

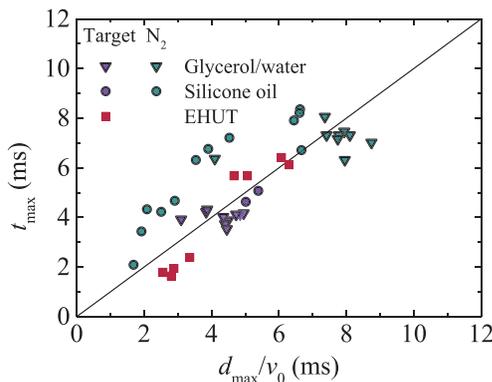
For  $t > t^*$ ,

$$\dot{\gamma}(\bar{r}) = \frac{1}{\pi R_T^2} \int_0^{d_T/2} \frac{1}{R(t)H(t)} \frac{dR}{dt} 2\pi r^2 dr = \frac{2d_T R(t)}{d_0^3} \frac{dR}{dt}. \quad (13)$$

Finally, we calculate the time-averaged shear rate experienced by the sheet on the target during the entire expansion process as

$$\langle \dot{\gamma} \rangle = \frac{1}{t_{\max}} \left( \int_0^{t^*} \frac{4R^2}{d_0^3} \frac{dR}{dt} dt + \int_{t^*}^{t_{\max}} \frac{4R_T R}{d_0^3} \frac{dR}{dt} dt \right), \quad (14)$$

$$\langle \dot{\gamma} \rangle = \frac{4}{t_{\max} d_0^3} \left( \frac{d_T^3}{24} + \frac{d_T (d_{\max}^2 - d_T^2)}{16} \right). \quad (15)$$



**FIG. 10.** Time to reach the maximal expansion,  $t_{\max}$ , as a function of  $d_{\max}/v_0$ , with  $d_{\max}$  the maximal expansion of the sheet and  $v_0$  the impact velocity, for all impact experiments. The line corresponds to  $t_{\max} = \frac{d_{\max}}{v_0}$ .

A good approximation, well supported by the experimental results (see Fig. 10), is  $t_{\max} \approx \frac{d_{\max}}{v_0}$  with  $v_0$  the impact velocity. Moreover, by assuming that  $3d_{\max}^2 \gg d_T^2$ , the time-averaged shear rate,  $\langle \dot{\gamma} \rangle$ , reads:

$$\langle \dot{\gamma} \rangle \approx \frac{d_T d_{\max} v_0}{4d_0^3}. \quad (16)$$

Having shown in Fig. 7(c) that the flow curves are well fitted by the Cross model [Eq. (11)], the effective shear viscosities,  $\eta_S^{\text{eff}}$ , are evaluated from the Cross fits at the relevant shear rate,  $\langle \dot{\gamma} \rangle$ . The latter vary from  $\langle \dot{\gamma} \rangle = 1420 \text{ s}^{-1}$  to  $3220 \text{ s}^{-1}$  leading to effective viscosities varying between  $\eta_S^{\text{eff}} = 10$  and  $2 \text{ mPa s}$ .

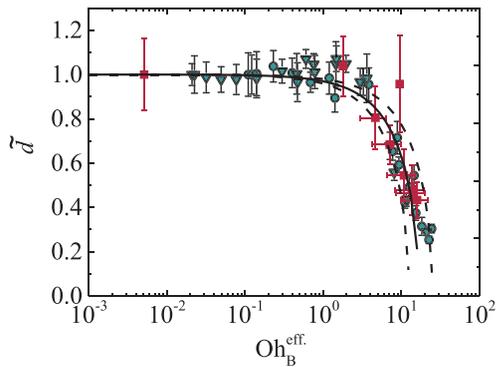
In Fig. 9, we plot the normalized maximum expansion as a function of an effective Ohnesorge number,  $\text{Oh}_S^{\text{eff}}$ , where  $\eta_0$  is replaced by  $\eta_S^{\text{eff}}$  for the non-Newtonian fluids (open red squares). The plot shows that viscoelastic samples expand much less than one would expect based only on their shear viscosity at the relevant shear rate, when taking as reference the data for Newtonian fluids. Moreover, taking into account the shear-thinning viscosity generates an unrealistic situation where samples with almost the same shear-thinning viscosity exhibit different expansions. Therefore, the shear-thinning viscosity fails to properly describe the maximum expansion of the viscoelastic sheets. Instead, the biaxial extensional viscous dissipation during the expansion in the air must be considered. The deformation field during the sheet expansion upon impacting a small target is essentially biaxial extensional, except on the small target where the sheet experiences both shear and biaxial extensional deformations. Given the very low value of the shear-thinning viscosity, we anticipate that the dissipation is mainly due to the biaxial extensional flow.

### 3. Viscous dissipation processes

In order to estimate the shear and biaxial extensional dissipation energy of rheo-thinning fluids, one must assess the relevant rate-dependent viscosities involved in these processes.

*a. Effective shear viscosity.* In order to account for the rate-dependent viscosity in the shear dissipation energy of non-Newtonian fluids, we will consider the effective shear viscosity,  $\eta_S^{\text{eff}}$ , resulting from the shear thinning of the EHUT solution and accessible by mean of classical rotational rheometry (see Sec. III B 2).

*b. Effective biaxial extensional viscosity.* Measuring properly, the biaxial extensional viscosity is a difficult task for viscoelastic fluids of low viscosity such as the present EHUT solutions.<sup>37,38,60–66</sup> In the framework of the impact drop problem, we have very recently established a successful, yet simple strategy in order to address this challenge.<sup>24</sup> Our approach consists of mapping the viscoelastic systems to Newtonian samples impacted under cold Leidenfrost conditions where they experience a purely biaxial extensional deformation. Thus, we attribute to the viscoelastic sample an effective biaxial extensional viscosity equal to the biaxial extensional viscosity of a Newtonian fluid with the same maximum expansion factor. Operationally, we shift horizontally the data of  $\bar{d}$  obtained for the viscoelastic samples so that they overlap the reference data for the Newtonian fluids (Fig. 11). The shift factor yields directly an effective biaxial extensional Ohnesorge



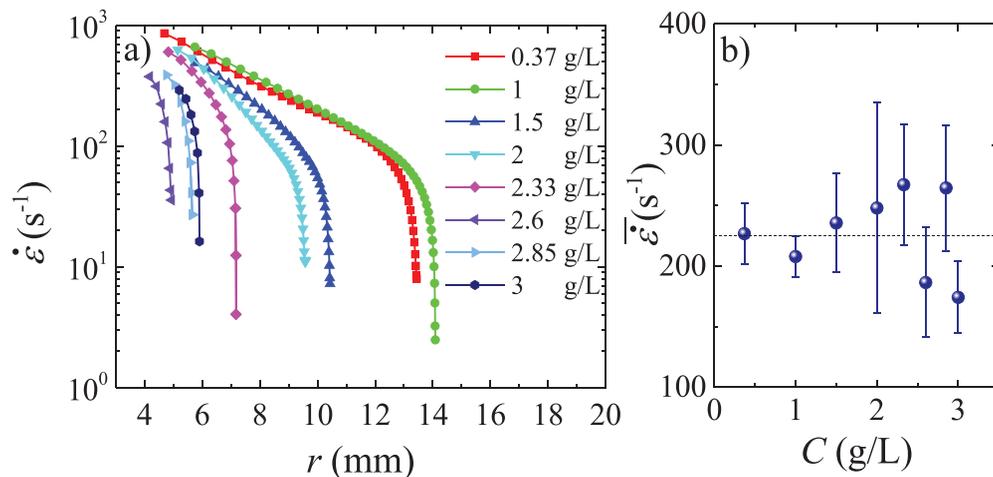
**FIG. 11.** Normalized maximum expansion factor for drops of Newtonian fluids impacting a surface covered with liquid nitrogen as a function of Ohnesorge number based on  $\eta_B^0$  (green symbols), together with that of EHUT samples (red squares) impacting a small target as a function of an effective Ohnesorge number based on an effective biaxial extensional viscosity  $\eta_B^{eff}$  chosen to collapse on the two sets of data in the viscous regime. The thin continuous line is the best fit with Eq. (17), and the dash lines show error bars on the fitting parameter  $\beta$ .

number,  $Oh_B^{eff} = \frac{\eta_B^{eff}}{\sqrt{\rho\Gamma d_0}}$ , from which an effective biaxial extensional viscosity  $\eta_B^{eff}$  is derived.  $\eta_B^{eff}$  is different from the biaxial extensional viscosity in the Newtonian limit,  $\eta_B^0 = 6\eta_0$ . Note that only  $\tilde{d}$  data in the viscous regime are shifted for EHUT systems (data in the capillary regime are not subjected to the shift).

In Fig. 11, we show the fit of the in Ref. 24: in:

$$\tilde{d} = \sqrt{1 - \beta Oh_B^{eff}}. \quad (17)$$

Here,  $\beta = 0.060 \pm 0.020$  is the only fitting parameter. The model in Eq. (17) is based on an energy conservation balance, assuming that the initial kinetic energy,  $E_K$ , is fully converted into surface energy,  $E_\Gamma$ , and biaxial extensional viscous dissipation energy,  $E_B$  (the shear viscous dissipation energy,  $E_S$ , is neglected).



**FIG. 12.** (a) Evolution of the biaxial extensional strain rate as a function of the radius of the sheet during its expansion for EHUT samples with different concentrations, as indicated in the legend. (b) Average strain rate calculated according to Eq. (18) as a function of concentration. Error bars represent the standard deviation of three different experiments.

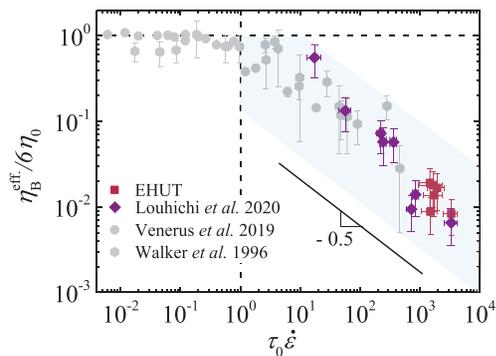
In the following, we will verify our assumption that the biaxial extensional dissipation is the relevant source of viscous dissipation in the present experimental conditions. First, we validate the rheological origin of  $\eta_B^{eff}$ . The effective biaxial extensional viscosity,  $\eta_B^{eff}$ , is expected to depend on the strain rate. We use here the same approach as the one developed in Ref. 24 to extract a relevant strain rate for the impact experiment. The time-dependent biaxial extensional strain rate in the expansion regime (Fig. 12) is averaged as

$$\bar{\dot{\epsilon}} = \frac{\int_0^{R_{max}} r \dot{\epsilon} dr}{\int_0^{R_{max}} r dr}. \quad (18)$$

Although the average strain rate (Fig. 12) does not vary significantly with sample concentration ( $\bar{\dot{\epsilon}}_{Mean} = 225 \pm 35 \text{ s}^{-1}$ ), in the following we use the computed average strain rate ( $\bar{\dot{\epsilon}}$ ) for each concentration.

We report in Fig. 13 the variation of the normalized effective biaxial extensional viscosity,  $\eta_B^{eff}/\eta_B^0$ , of EHUT supramolecular polymers as a function of the effective Weissenberg number,  $Wi^{eff}$ . The latter is defined as  $Wi^{eff} = \tau_0 \bar{\dot{\epsilon}}$ , where  $\tau_0$  is the relaxation time obtained from linear shear rheology measurements (Fig. 7) and  $\bar{\dot{\epsilon}}$  is computed using Eq. (18) (Fig. 12).

On the same plot, we report previous experimental data for normalized biaxial extensional thinning viscosities measured for different viscoelastic fluids using different setups.<sup>37,38</sup> We also report polyethylene oxide (PEO) solutions data impacted under cold Leidenfrost conditions from our previous work.<sup>24</sup> The EHUT experimental data follow remarkably well the experimental results reported in the literature for stationary biaxial extensional deformation flows of different systems (surfactant wormlike micelles in Ref. 37, polystyrene solutions in Ref. 38, and PEO solutions in Ref. 24) At low Weissenberg numbers ( $Wi = \dot{\epsilon} \tau_0 < 1$ ), the biaxial extensional viscosity is rate independent and reaches the Newtonian limit:  $\eta_B = \eta_B^0 = 6\eta_0$ . By contrast, when  $Wi > 1$ , the biaxial extensional viscosity decreases with the strain rate as  $\eta_B \sim 6\eta_0 (\dot{\epsilon} \tau_0)^{-0.5}$



**FIG. 13.** Effective thinning biaxial extensional viscosity, extracted from the manual shift of the data from Fig. 11 in order to collapse with the data for Newtonian samples, normalized by the plateau biaxial extensional viscosity as a function of the Weissenberg number for the different EHUT solutions studied here (red squares), together with the literature data as indicated in the legend. A unique master biaxial flow curve is obtained with a thinning exponent of  $-0.5$  in agreement with theoretical predictions (see the text). The gray zone is the region of data uncertainties.

with a thinning exponent of  $-0.5$  as validated by theoretical predictions in the framework of the classical tube-based model.<sup>67</sup>

*c. Comparison between shear and biaxial extensional viscous dissipation.* After assessing  $\eta_S^{\text{eff}}$  and validating the rheological origin of  $\eta_B^{\text{eff}}$ , we can now compute and compare the two viscous dissipation processes for the supramolecular polymers. In order to account for the rate-dependent viscosity in the shear dissipation energy of non-Newtonian fluids, we have replaced, in Eqs. (6) and (8),  $\eta_S$  by  $\eta_S^{\text{eff}}$ . On the other hand, we replaced, in Eq. (9),  $\eta_B$  by  $\eta_B^{\text{eff}}$ , to evaluate the biaxial extensional viscous energy.

Results are reported in Fig. 6. The dissipation energies have been normalized by the initial kinetic energy of the drop  $E_K = \frac{1}{2}mv_0^2$  and plotted as a function of the zero-shear-rate Ohnesorge number. We observe that the biaxial extensional viscous dissipation energy is larger by two orders of magnitude than the shear dissipation energy (for both limits in the calculation of the shear dissipation energy) despite the fact that the zero-shear viscosity of the less concentrated complex fluid sample (about 10 Pa s) is two orders of magnitude larger than the more viscous Newtonian samples we have investigated (about 0.2 Pa s). Note that in the same experimental conditions, a Newtonian viscous sample with a viscosity of 10 Pa s will not expand outside of the target after impact.

## IV. CONCLUSIONS

Drop impact experiments on a small solid target have been performed with Newtonian fluids and solutions of entangled supramolecular polymers as rheo-thinning viscoelastic fluids. Upon impact on the surface, a drop expands into a sheet. A part of the expanding sheet is always at intimate contact with the target, while the rest of the sheet freely expands in the air. We have measured the maximum expansion of sheets for the two classes of fluids. We have quantitatively assessed the energy dissipation due to shear and biaxial extensional deformations, and have highlighted the distinctive roles of shear and biaxial extensional viscosities. We have shown that the expansion process is controlled by a combination of shear (on the target) and biaxial extensional (in the air) deformations. For Newtonian fluids, the two sources of dissipation remain of the same order of magnitude. In sharp

contrast to the findings for Newtonian fluids, for the rheo-thinning viscoelastic fluids, the dominant source of viscous dissipation is the biaxial extensional deformation during the sheet expansion, and consequently, the biaxial thinning extensional viscosity is found to control the maximum expansion of the sheets. The physical reason is that the thinning behavior of the fluid strongly depends on the flow type, since nonlinear viscosities scale differently with the relevant deformation rate. In other words, polymeric fluids exhibit a different thinning behavior in shear and in biaxial extension, justifying the distinction we made between the two viscous dissipation modes. On the contrary, for Newtonian fluids, both viscosities are proportional to each other, and thus, the distinction of the nature of the viscous dissipation is less important. We have also shown that the ratio between the two dissipation modes strongly depends on the size of the target. It would therefore be interesting to study the contribution of the shear dissipation at different drops to target ratios. This will be the task of future work.

## AUTHORS' CONTRIBUTIONS

A.L. and C.-A.C. contributed equally to this work.

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## DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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