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Flow focusing with viscoelastic liquids

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We present an experimental comparative study of the effect of elasticity of the
continuous liquid in generation of droplets in microfluidic flow focusing devices.
For a wide range of values of dynamic viscosity of the Newtonian droplet phase
we compare the dynamics of the formation of droplets in three different Newtonian
and non-Newtonian, viscoelastic (Boger) fluid pairs of the same (and nearly constant)
shear viscosities. In both Newtonian and viscoelastic systems we find similar regimes
of operation of the system: (i) dripping without satellites, (ii) dripping with single
satellites, (iii) formation of multiple satellites, and (iv) jetting. We find that the
elasticity of the focusing liquid stabilizes the jets facilitating formation of smaller
droplets, and leads to transitions between various regimes at lower ratios of flow and
at lower values of the capillary numbers in comparison to the Newtonian focusing
liquids. We also show that the polydispersity of Newtonian droplets produced in non-
Newtonian continuous phases depend significantly on the viscosity of the droplet
phase while this dependence is minor when the continuous liquid is Newtonian.
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I. INTRODUCTION

Microfluidics handles liquids at small (usually micrometer) length scales1–3 where the surface
tension and the viscous dissipation of energy dominate while the inertial effects are negligible. As
a key consequence, the Reynolds number – the dimensionless expression of the ratio of inertial
to viscous forces – has low values and the flow is laminar without turbulence. As opposed to the
simplistic nature of laminar flow of Newtonian liquids, viscoelastic fluids produce more interesting
behaviors. For example, in elastic liquids, viscoelastic turbulences may occur even at low Reynolds
numbers4, 5 and even a small amount of long-chain polymer additives can significantly reduce the
drag6, 7 in the turbulent regime. Elastic liquids exhibit a range of exotic behaviors such as the well-
known die-swell effect (in which the liquid expands downstream of an exit into air), the Weissenberg
effect (climbing of liquid onto a spinning rod) or among others, the tubeless siphon effect (of liquid
being drawn by the flow into an opening).

The surprising and complicated rheological behavior of elastic liquids can be utilized in mi-
crofluidic systems to perform non-trivial functions. For example, a microfluidic device that realizes
bistable flip-flop memory,8 a flow stabilizer using nonlinear resistance8 or a fluid mechanic equivalent
of a diode—a microfluidic rectifier.9–11 The use of non-Newtonian (NN) liquids in microfluidic
devices may also allow for miniaturization of the existing macrodevices that use or examine non-
Newtonian liquids—especially rheometers.12, 13

The use of viscoelastic liquids in flow focusing devices has recently gained attention as a
possible method for focusing and separation of particles. First proposed by Leshansky et al.,14 it
was shown that microparticles driven with viscoelastic medium migrate toward the centerline due

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to the first normal stress difference between the centerline and the walls. Leshansky et al.\textsuperscript{14} showed that in slit microchannels at negligible inertia (2D elasticity-driven focusing) the spatial distribution of particles depends on their size and on the rheological properties of the suspending medium. Later Yang et al.\textsuperscript{15} expanded this technique to microchannels of square cross section, and showed that three-dimensional particle focusing can be achieved via a synergistic combination of inertial and elastic forces (3D Elasto-Inertial Focusing). Elastic forces themselves are not sufficient to generate three-dimensional focusing. This is because the low normal stress region in the corners of the lumen of the microchannel generates multiple equilibrium positions for the particles. Very recently, elasto-inertial particle focusing was successfully applied for separation of (i) solid microparticles of various sizes,\textsuperscript{16} (ii) RBCs and WBCs from platelets in diluted whole blood sample,\textsuperscript{16} and (iii) flexible DNA molecules.\textsuperscript{17}

Here we explore the idea of use of viscoelastic focusing liquids to enhance the formation of Newtonian droplets in square cross section flow focusing droplet generators. The dynamics of breakup and the formation of Newtonian droplets in Newtonian continuous liquid are already relatively well known. Microfluidic flow focusing junctions\textsuperscript{2,3} operated with Newtonian liquids are known to produce monodisperse bubbles, droplets, and multiple emulsions with perfect control over the size and structure of the droplets. Many different variants of these systems have been studied: for the formation of oil-in-water (O/W) and water-in-oil (W/O) simple emulsions and for the formation of multiple (O/W/O and W/O/W) droplets.\textsuperscript{18–20} Formation of monodisperse droplets holds the potential for multiple applications, some of which may require the formation of droplets of NN liquids (e.g., of samples of physiological liquids such as blood, synovial fluid or saliva). The problem of the formation of droplets of NN liquids has been researched by a number of groups.\textsuperscript{21–23} For example the formation and the pinch-off mechanism of viscoelastic droplets in Newtonian continuous phase was investigated in various geometries of the flow-focusing device by Steinhaus et al.\textsuperscript{23} The effect of the molecular weight of the polymer in the droplet phase on filament thinning was studied by Arratia et al.\textsuperscript{21,22} Typically, it is observed that elasticity of the droplet liquid prolongs the processes of thinning of the filament and significantly increases the interval required for breakup to complete. As an effect, often, before the elongated and thin jet of the polymeric liquid breaks, the capillary forces form a number of bulges—interconnected “droplets” often called Beads-On-A-String (BOAS).

Here we focus on the—previously never studied—situation in which the non-Newtonian fluid is the outer (continuous) liquid, while the inner (droplet) phase is Newtonian. There are two motivations for this study. One is that the use of elasticity in the focusing liquid could potentially be useful in achieving more efficient focusing of the inner stream within a given geometry of the junction. To this end we characterize the effect of the elasticity of the focusing liquid on the formation of droplets, satellite droplets, dripping-jetting transition, and on the polydispersity of droplets generated in the junction. The second motivation is that microfluidics has been demonstrated as a useful tool for the generation of compound (or multiple) droplets that comprise polymeric shells. In such a case the inner (core) liquid breaks-up while being shielded by a typically non-Newtonian polymeric liquid.

In order to extract the role of elasticity of the focusing liquid, we conduct two series of experiments. In one we use Boger fluids\textsuperscript{24} (or near-Boger fluids), i.e., a constant viscosity elastic fluids, in the second we use a standard Newtonian liquids, both liquids having a very similar value of the shear viscosity coefficient. In order to comprehensively explore the effects of elasticity of the continuous phase on the process of the formation of drops in a microfluidic flow-focusing geometry we test a wide range of shear viscosities of the inner phases (4-500 cP) and the outer phases (30-1780cP), a wide range of rates of flow of the outer phases, a range of elasticity of the non-Newtonian continuous phases by using three liquids of low, medium, and high elasticity and, finally, three distinct classes of geometries of the flow-focusing junction to elucidate the role of geometry in the process.

II. MATERIALS AND METHODS

A. Microfabrication

We fabricated the flow-focusing devices using three layers of polycarbonate (Fig. 1). The middle layer is a spacer that has the same thickness (250 μm) as the height of the channels. We prepared the patterns of channels of the desired 2D layout by milling this intermediate layer of polycarbonate.
FIG. 1. Schematic illustration of the layers of polycarbonate constituting the microfluidic device and of the geometries of the flow-focusing junctions. (a) We assembled all the devices by sealing three layers of polycarbonate. The top layer (of thickness $d = 5$ mm) comprised through-holes for both of the phases and for the outlet; the middle layer ($d = 0.25$ mm) contained the layout of the channels; the system was closed with a flat bottom plate ($d = 5$ mm). (b) Geometries of the channels: (FF-1) simple cross junctions of widths $w_{in} = w_{out} = 250$ $\mu m$, (FF-2) cross junction with additional orifice of widths $w_{or} = 100$ $\mu m$, $l_{or} = 100$ $\mu m$, (FF-3) cross junction with an orifice and a wider outlet channel $w_{out} = 800$ $\mu m$. The scale bar is 250 $\mu m$.

(PC) (Makrolon®, Bayer, Germany) through using MSG4025 CNG milling machine (ErgWind, Poland). We then placed this layer between two other PC plates of the same material of thickness of 5 mm. On the upper plate we drilled holes for inlets and outlet, respectively. The pieces were cleaned with detergent and water and dried with pressurized air prior to the sealing process (Fig. 1(a)). We swelled the pieces with vapour of DCM (Dichloromethane pure p.a.; Chempur, Poland) in vacuum and sealed the 3-layer device under appropriate temperature and pressure as described by Ogonczyk et al. This scheme of fabrication of the device provides smooth top and bottom walls of the channels guaranteeing better transparency than if the channels were milled into a thicker material because the milling procedure leaves surface roughness that reduces the resolution of imaging. We rendered the surface of the microchannels hydrophilic using PAH/PSS polyelectrolytes. This surface treatment allows for production of organic droplets without the use of surfactants, which could rigidify the interface of the two liquids blurring the intended separation of the viscous and elastic effects.

B. Liquids

We used three pairs of Newtonian/viscoelastic liquids with the same shear viscosities. In the first one the Newtonian continuous liquid was a 90% (w/w) aqueous solution of glycerin (referred to as N1). Polymeric fluid counterpart was made by adding small amount (100 ppm) of flexible polymer polyacrylamide (PAM, $M_w = 1.8 \times 10^7$; Polysciences, Inc., USA) to the Newtonian phase (referred to as NN1, strongly elastic liquid, $E_l = 328.2$). The second Newtonian continuous phase was an 81.5% aqueous solution of glycerin (referred to as N2) and a corresponding polymetric liquid was a 300 ppm w/w polyacrylamide (PAM, $M_w = 5 \times 10^6$, 1% soln. in water; Polysciences, Inc., USA) dissolved in 80% aqueous glycerin (referred to as NN2, moderate elasticity liquid, $E_l = 14.8$). In the third set the Newtonian liquid was a 75.5% w/w aqueous solution of glycerin (N3) and its viscoelastic pair was a 50 ppm polyacrylamide (PAM, $M_w = 5 \times 10^6$, 1% soln. in water; Polysciences, Inc., USA) dissolved in 75% aqueous glycerin (NN3, low elasticity liquid, $E_l = 3.42$). The elasticity numbers $E_l$ of the viscoelastic liquids were calculated as $E_l = \frac{\lambda \mu}{\rho w^2}$ ($\lambda$, $\mu$, $\rho$ being the relaxation times, dynamic viscosities, and densities of the liquids, respectively, and $w$ is the typical length scale—in our case the width of the channels). All the polymeric solutions have constant or nearly constant shear viscosities and exhibit high extensional viscosity (see Fig. S1 of the supplementary material). The NN1 showed minor shear thinning (Fig. 2), the other two polymeric liquids can be treated as true Boger liquids. The concentration of the polymers in all three viscoelastic liquids were kept in the dilute regime, i.e., $c/c^* < 1/3$ (where $c^*$ is the overlap concentration of the polymer), to avoid polymer chain entanglement. Since the shear viscosity is nearly constant, the shear thinning
FIG. 2. Measured flow curves of the viscoelastic liquids (full symbols) and their corresponding Newtonian pairs with the same shear viscosities (empty symbols) used in our experiments. The strongly elastic liquid (NN1, $E_i = 328.2$, full squares) exhibits minor shear thinning the other two non-Newtonians have a constant viscosity.

effect is minimized (Fig. 2) and it is possible to investigate only the effect of the elasticity on the flow behavior.

For the dispersed phase we used silicone oil of kinematic viscosity of 5 cSt (Xiameter® PMX-200 Silicone fluid; Dow Corning, USA) (only for the highest viscosity Newtonian/non-Newtonian continuous phase liquids) and PDMS oils of kinematic viscosities of 20, 50, 200, and 500 cSt (Poly(dimethylsiloxane) 200® fluid; Sigma-Aldrich, Germany) as received from the manufacturers.

We measured the dynamic viscosity coefficients of the 5 and 20 cSt oils with B6-BS/IP/MSL Suspended Level Viscometer and the rest of the fluids used in our experiments with the Anton Paar® Physica MCR 301 stress controlled rheometer at 23 °C using 1°/40 mm cone and plate geometry (diameter of 40 mm and cone angle of 1°). The relaxation times of the polymeric liquids were measured with capillary breakup extensional rheometer (CaBER1, Thermo Scientific, Karlsruhe, Germany). The surface tension of the dispersed phase against both the Newtonian and the viscoelastic fluids were measured using the pedant drop technique. We show the measured rheological flow curves in Fig. 2 and detail the properties of the liquids in Table I.

TABLE I. Measured physicochemical properties of the fluids: density $\rho$, dynamic viscosity $\mu$, concentration of the polymer $c$, overlap concentration $c^*$, relaxation time $\lambda$ and the interfacial tension between the droplet phase and Newtonian $\sigma_N$ and viscoelastic $\sigma_{NN}$ outer phase. All measurements were performed at 23 °C.

<table>
<thead>
<tr>
<th>Continuous phase</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$\mu$ (Pa.s)</th>
<th>glycerin (%)</th>
<th>c (ppm)</th>
<th>$c^*$ (ppm)</th>
<th>$\lambda$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>N1 – 90%glycerin</td>
<td>1235.7</td>
<td>0.178</td>
<td>90</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>NN1 – PAM18M</td>
<td>1234.02</td>
<td>0.175</td>
<td>90</td>
<td>100</td>
<td>350</td>
<td>1.43E-01</td>
</tr>
<tr>
<td>N2 – 81.5% glycerin</td>
<td>1204.7</td>
<td>0.0604</td>
<td>81.5</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>NN2 – PAM5M</td>
<td>1205.3</td>
<td>0.0600</td>
<td>80</td>
<td>300</td>
<td>975</td>
<td>1.72E-02</td>
</tr>
<tr>
<td>N3 – 75.5%glycerin</td>
<td>1189.5</td>
<td>0.0301</td>
<td>75.5</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>NN3 – PAM5M</td>
<td>1188.8</td>
<td>0.0301</td>
<td>75</td>
<td>50</td>
<td>975</td>
<td>8.05E-3</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Disperse phase</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$\mu$ (Pa.s)</th>
<th>$\sigma_{N1}$ (mN/m)</th>
<th>$\sigma_{NN1}$ (mN/m)</th>
<th>$\sigma_{N2}$ (mN/m)</th>
<th>$\sigma_{NN2}$ (mN/m)</th>
<th>$\sigma_{N3}$ (mN/m)</th>
<th>$\sigma_{NN3}$ (mN/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xiameter 5 cSt</td>
<td>776.15</td>
<td>0.0041</td>
<td>22.1</td>
<td>22.3</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>PDMS 20 cSt</td>
<td>959.2</td>
<td>0.0196</td>
<td>27.0</td>
<td>27.0</td>
<td>27.2</td>
<td>24.9</td>
<td>28.5</td>
<td>28.7</td>
</tr>
<tr>
<td>PDMS 50 cSt</td>
<td>973.14</td>
<td>0.0499</td>
<td>28.9</td>
<td>28.1</td>
<td>28.3</td>
<td>25.9</td>
<td>29.4</td>
<td>29.7</td>
</tr>
<tr>
<td>PDMS 200 cSt</td>
<td>979.4</td>
<td>0.2074</td>
<td>27.6</td>
<td>27.4</td>
<td>28.5</td>
<td>25.9</td>
<td>30.6</td>
<td>30.0</td>
</tr>
<tr>
<td>PDMS 500 cSt</td>
<td>986.95</td>
<td>0.5072</td>
<td>26.6</td>
<td>27.3</td>
<td>29.0</td>
<td>25.5</td>
<td>...</td>
<td>...</td>
</tr>
</tbody>
</table>
For the formation of the droplets we used syringes of small diameter (1 ml Braun Injekt®-F) and syringe pumps (Harvard Apparatus, PHD 2000) to inject the liquids into the microchannels. The small diameters of the syringes served to reduce the fluctuations of the rate of flow.29

### C. Imaging and analysis

For monitoring and recording we used the Nikon SMZ800 stereo microscope and a high speed camera (Photron Fastcam SA-5, model 1000-C1). We analyzed the images with Image J software (Fiji, ImageJA 1.42e).

### III. RESULTS AND DISCUSSION

Our main goal is to characterize the influence of the viscoelastic character of the continuous liquid on the process of the formation of droplets in microfluidic flow focusing junctions. In order to clearly verify the influence of the elastic component we compare the processes of generation of droplets in Newtonian and viscoelastic (Boger) liquids of the same shear viscosity. Boger fluids,24 are usually polymeric liquids which exhibit elasticity but their shear viscosity within a good approximation can be treated as constant (Fig. 2). The choice of such liquids is severely limited by the fact that typically if they posses significant elasticity they are also very viscous30 and vice versa if they are less viscous23,31 they have negligibly low elasticity. Here we use high molecular weight (MW = \(5 \times 10^6 - 1.8 \times 10^7\)) polyacrylamides of various concentrations dissolved in aqueous glycerin of various shear viscosities as strongly elastic (NN1, \(E_l = 328.2\)), moderate elasticity (NN2, \(E_l = 14.8\)) and weak elasticity (NN3, \(E_l = 3.42\)) liquids, and glycerin-water mixtures with adjusted viscosities as their Newtonian counterparts. For a given Newtonian/non-Newtonian continuous phase we tested silicone oils of a range of viscosities (\(\mu_d = 5-500\text{cSt}\)) as the dispersed (droplet) liquids.

As a reference geometry of the junction, for which we report the most systematic screen of the dynamics of break-up and the quality of emulsions, we use the simplest cross-slot geometry of square cross section of the channels (FF-1, see Fig. 1(b)). In all experiments we fixed the rate of flow of the droplet liquid \(Q_d\) (to 100 and 250 \(\mu\text{L/h}\) for experiments with continuous liquids N1/NN1 and N2/NN2; and to 500 \(\mu\text{L/h}\) for the third pair N3/NN3). We changed the rate of flow of the continuous phase \(Q_c\) (from 25 \(\mu\text{L/h}\) to 4000 \(\mu\text{L/h}\) for N1/NN1; between 50 and 5000 \(\mu\text{L/h}\) for N2/NN2; and 75-10 000 \(\mu\text{L/h}\) for N3/NN3). We define the ratio of flow \(q\) as the ratio of the flow rate of the continuous phase to the rate of flow of the dispersed phase \(q = Q_c/Q_d\) to preserve direct proportionality between the non-dimensional ratio \(q\) and the variable \(Q_c\). Since for a given (fixed) viscosity of the continuous phase we tested a range of viscosities of the dispersed phase—we define the ratio of viscosities as the viscosity of the disperse phase divided by the viscosity of the continuous phase. To avoid confusion we mark the symbol of the ratio of viscosities \(\beta\) with a prime, i.e., \(\beta' = \mu_d/\mu_c\).

The capillary number calculated for the flow of the continuous phase (\(Ca_c = \mu_c \cdot Q_c/A \cdot \gamma\)) varied between \(10^{-4}\) and \(10^{-1}\). For the dispersed phase (\(Ca_d = \mu_d \cdot Q_d/A \cdot \gamma\)) the capillary number ranged from \(10^{-5}\) to \(10^{-2}\). In these expressions, \(\rho, \mu,\) and \(Q\) are the density, the dynamic viscosity and the rate of flow of the liquids, \(\gamma\) is the interfacial tension between the droplet and the continuous phase and \(A\) is the area of the lumen of the channels. Subscripts “c” and “d” indicate the continuous and the dispersed phases, respectively. The value of the Reynolds numbers \(Re_c = \rho_c \cdot Q_c/\mu_c \cdot w\), where \(w\) is the width of the channel, spanned approximately three orders of magnitude from \(Re_c \sim 10^{-4}\) to \(Re_c \sim 10^{-1}\). We calculated the Weissenberg number for the non-Newtonian liquids as \(Wi = \epsilon\lambda\), where \(\epsilon\) is the rate of extension \(\epsilon = \frac{Q_d}{w^2}\) as generally used in the literature for cross slot geometries32-35 and \(\lambda\) is the relaxation time of the viscoelastic liquids. In our experiments \(Wi\) ranged between 0.01 and 10 s\(^{-1}\). The Reynolds numbers of the droplet phase \(Re_d = \rho_d \cdot Q_d/\mu_d \cdot w\) covered a similarly wide range of values from \(\sim 10^{-4}\) to \(\sim 10^{-2}\).

### A. Flow patterns

While the elasticity of the droplet phase has a pronounced qualitative effect on the dynamics of break-up21,22 the non-Newtonian character of the continuous liquid has more subtle effects. Although
FIG. 3. Examples of flow patterns at selected ratios of flow (columns) for Newtonian (N1 – 90% glycerine) and non-Newtonian focusing phases (NN1 – 100 ppm PAM-18M) of the same viscosity and Newtonian disperse phase of various viscosities (rows). The disperse phase has a fixed rate of flow $Q_d = 100 \mu L/h$. In the dripping regime the non-Newtonian outer phase deforms the shape of the droplets. The rate of deformation grows with increasing rate of flow of the continuous phase. Elasticity of the non-Newtonian continuous phase stabilizes the jet which results in longer treads of the jet than in the Newtonian continuous phase.

the detailed analysis of, e.g., the frequency of breakup does show systematic and potentially important changes in the dynamics of the droplet generator the appearance of systems operated with Newtonian and non-Newtonian continuous liquids is quite similar (Fig. 3).

A closer look at the micrographs shown in Fig. 3 reveals several differences. The first one we note is that in the case of NN focusing, the droplets are much more deformed. This becomes apparent when the “diameter” of the droplet becomes smaller than the width of the outlet channel (see the panel $Q_c/Q_d = 5$ in Fig. 3). In Newtonian liquid we observed only spherical, or nearly spherical, droplets and their shape does not change in the flow through the outlet channel. In the non-Newtonian continuous phase the droplets assume elliptical shapes after the pinch-off and relax slowly $\mathcal{O}(\lambda)$ into bulleted shapes in the flow. The elongation (length-to-width ratio of the droplet) and the time of subsequent relaxation increases with increasing ratio of flows $q$ until the transition to the jetting regime. This observation aligns with earlier reports$^{36}$ where elasticity of the suspending liquid was stated to promote the deformation of the Newtonian droplets at ratios of viscosity $\beta' \leq 1$. In our study we noticed significant deformation of droplets up to $\beta' \approx 2.8$.

The second important difference is that the transition from the dripping to the jetting regime occurs at lower ratios of flow in the viscoelastic continuous media than in the Newtonian one. Also the length of the thread of the inner phase in the jetting regime is longer in the non-Newtonian liquids than in the Newtonian ones. Figure 3 shows the N1-NN1 pair of continuous liquids at fixed rate of flow of the disperse phase $Q_d = 100 \mu L/h$.

As can be seen in Fig. 3 for all combinations of the rates of flow of the two phases, the tips of the droplet phase focused by the viscoelastic liquid are thinner and more pointed than the ones surrounded by the Newtonian liquid. This has the simple explanation that in the flow-focusing junction due to the large strain fields, molecules are extended and they align with the flow.

In the NN focusing liquids, the elastic effects cause the tips of the Newtonian droplet phase to be more efficiently focused (or contracted) and the droplets broken off from these tips are elongated even at relatively low ratios of flow. As the rate of flow of the continuous phase increases and the rate of extension increases the tips become thinner and they are more difficult to break because, just like the case of the non-Newtonian inner phase,$^{22}$ the elasticity of the outer phase also opposes the extension needed to fill the troughs in the breaking ligament. As a result in the non-Newtonian liquid we observe longer and more stable jets than in the viscous outer liquid at the same rates of flow.

B. Flow regimes

We define the dripping regime as the regime in which breakup occurs at the junction or in close proximity of the junction and the tip of the droplet phase recoils into the junction after the pinch-off. In the dripping regime typically large droplets are produced at low rates $O(10^0 - 10^1 \text{ s}^{-1})$. 

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FIG. 4. Flow map as a function of capillary number of the continuous phase $C_{a_c}$ versus the capillary number of the disperse phase $C_{a_d}$. The evolution of transitions between the regimes are very similar in both Newtonian and Boger phase focusing, but elasticity promotes transitions at lower $C_{a_c}$. Remarkably, in the viscoelastic focusing system the ratio of viscosities $\beta = \frac{\mu_d}{\mu_c} \approx 1$ favors the formation of multiple satellites rather than single satellites, while at $\beta > 3$ this regime becomes very narrow and vanishes.

Jetting regime ensues at higher rates of flow of the continuous phase. In the jetting regime the stream (jet) of the inner liquid extends into the outlet channel, droplets pinch-off further downstream and the tip of the inner phase remains in the outlet channel after break-up. The frequency of generation of droplets in the jetting regime may be several orders of magnitude higher than in the dripping regime (typically by a factor of $O(10^2)$).

We demonstrate the transitions between the dripping and jetting regimes on a flow map plotted as the function of the capillary number of the continuous phase versus the capillary number of the dispersed phase in Fig. 4. We distinguish four regimes: (a) dripping without satellites, (b) dripping with a single satellite, (c) dripping with multiple satellites, and (d) jetting.

The dripping regime in which droplets are produced without satellites is limited to low values of $C_{a_d}$ (both low rate of flow and low viscosity of the droplet phase (Fig. 4(a))). Introduction of elastic effects into the continuous liquid shifts the transition to the satellite regime to lower ratios of rates of flow (Fig. 4(b)).

We found that satellites start to form above a capillary number of the focusing liquid $C_{a_c}$ which is related to capillary number of the inner phase $C_{a_d}$ as $C_{a_c} \propto \alpha_1 \cdot C_{a_d}^{-2.2}$ when focusing with Newtonian liquid and $C_{a_c} \propto \alpha_1 \cdot C_{a_d}^{-1.57}$ in the case of non-Newtonian focusing. The values of the fitting parameters $\alpha_1 = 2.24 \times 10^{-11}$ and $\alpha_2 = 2.24 \times 10^{-9}$ may depend on the geometry of the junction. In general, the satellite-free dripping regime is typical when low viscosity dispersed phase is focused with a high viscosity outer phase below $\beta' \approx 0.15$ and at low rates of flows (low values of the $Ca$).

Upon increase of $C_{a_c}$ the number of satellites increases. At small values of $C_{a_c}$ only a single satellite accompanies the primary droplet (Fig. 4(b)). Above a certain value of $C_{a_c}$ the system starts to form a pack of satellites (Fig. 4(c)). Typically, the volumes of the satellites decrease monotonically in the sequence that follows the primary droplet. We classified this regime as the “multiple satellite regime.” The range of $C_{a_c}$—at which a sequence of satellite droplets is formed—strongly depends of the rheological properties of the non-Newtonian phase. While in the Newtonian focusing system $C_{a_c}$ scales with $C_{a_d}$ similarly for all tested Newtonian continuous phases, in the viscoelastic systems at viscosity ratio $\beta' = 1$ we observed that packs of satellites start to form at much lower $C_{a_c}$ than in the corresponding Newtonian phase (Fig. 4, right graph). Moreover, the formation of multiple satellites at $\beta' = 1$ found to be dependent on the elasticity number of the continuous phase. In strongly elastic liquid (NN1) we observed the multiple satellite regime to start at $C_{a_c}$ one order of magnitude lower than in the corresponding Newtonian system. With decreasing elasticity (moderate elasticity liquid NN2) multiple satellites starts to form still at lower $C_{a_c}$ than is the corresponding N2, but the effect is less pronounced. For low elasticity (NN3) it showed similar behavior as in the Newtonian system. As the viscosity of the droplet phase exceeds the viscosity of the continuous phase the number of satellites decreases in both Newtonian and non-Newtonian systems and at $\sim \beta' > 3$ completely vanishes with the system producing only single satellites.
When the number of the satellites reaches a certain threshold (5-12 depending on the ratio of viscosities and the type of the focusing phase) the dripping regime transits into the jetting regime.

C. The dripping-jetting transition

Considering the balance of viscous shear, surface tension and inertial forces at the dripping-jetting transition of Utada et al.\textsuperscript{37} showed that the dripping-jetting transition for co-flowing liquid streams can be drawn in a state diagram as a function of $Ca_c$ versus $We_d$, when both dimensionless numbers are small. The analysis concluded with a general expectation that the flow behavior may depend on other dimensionless parameters, such as $(Wi_d, Ca_c, Re_d, Re_c, q)$.

Even though the flow-focusing design that we study here differs from the co-flowing geometry\textsuperscript{37} they share common characteristics: both designs the shearing stresses, capillary instabilities and a similar set of parameters govern the formation of droplets.\textsuperscript{38-40}

In our experiments we found that the state diagram of $Ca_c$ versus the $We_d$ does not yield suggest that the $We_d$ group controls the dripping-jetting transition. We found that with increasing velocity of the inner phase $U_d = Q_d/A$, the critical (transition) $We_d$ number increases with the second power of $U_d$ (see Fig. S2 of the supplementary material\textsuperscript{26}). Also no quantitative difference in the dripping-jetting transition expressed in the capillary and Webber numbers (see Fig. S2 of the supplementary material\textsuperscript{28}), can be directly seen between the Newtonian and viscoelastic focusing, although we observed that at a fixed rate of flow of the disperse phase the dripping-jetting transition occurs at much lower rates of flow of the outer liquid in viscoelastic continuous media than in the Newtonian one of the same shear viscosity.

Since the densities of all droplet phases are nearly same and the interfacial tensions between the two phases are also very similar in value, the variable parameters in our experiments were the viscosities and the rates of flow of the two immiscible phases and the presence or absence of the elasticity in the continuous phase. Therefore, we consider the balance of shear and interfacial forces on the disperse phase (without taking into account the inertial effects) immediately before the dripping-jetting transition takes place. For Newtonian focusing the viscous shear stress of the outer phase is balanced by the surface tension—and this balance can be expressed by the capillary number $Ca_c$. When $Ca_c$ is small, surface tension dominates, forcing break-up immediately at the junction (the dripping regime). By contrast as $Ca_c$ increases the shear stress on the droplet becomes large enough to overcome surface tension, causing the transition to the jetting regime. We note that the viscosity of the inner phase also retards break-up and facilitates the transition to jetting. In other words, an increase of $Ca_d$ (at fixed $Ca_c$) promotes jetting.

In general the dripping-jetting transition takes place when the sum of forces exerted on the inner fluid ultimately overcome the surface tension forces.\textsuperscript{37} While for the Newtonian continuous phase only the viscous shear balances the surface tension forces in viscoelastic continuous phase, additional elastic forces have an important contribution and facilitate transition to jetting at smaller magnitudes of the viscous forces, i.e., in the elastic liquids the transition occurs at lower capillary number (lower $Q_c$). The distribution of the elastic forces of the continuous phase in square microchannels has been discussed previously by Yang et al.\textsuperscript{15}

To generalize this considerations we used 5 liquids as the disperse phase and three pairs of (N and NN) liquids as the outer phase varying the ratio of viscosity $\beta' = \mu_d/\mu_c$ between 0.02 and 16.9 and the ratio of flows $q$ between 0.25 and 40 at nearly constant surface tension (25-30 mN/m). We plotted the state diagrams as $Ca_c$ versus $Ca_d$ for both Newtonian and viscoelastic focusing phases in Fig. S3 of the supplementary material\textsuperscript{28} to show that the dripping-jetting transition has a power-law character and scales with the capillary numbers of the two phases as $Ca_c = B \cdot Ca_d^A$. By fitting experimental data over all the observed dripping-jetting transition points (see Figs. S3 and S4 of the supplementary material\textsuperscript{28}) we found that $A_N = -0.35$ and $B_N = 6.71 \times 10^{-3}$ when focusing with Newtonian liquid and $A_{NN} = -0.47$ and $B_{NN} = 1.6 \times 10^{-3}$ when elastic forces are present.

As a results of the elastic forces the dripping-jetting transition occurs at significantly lower $q$ when focusing with elastic liquid (depending on $\beta'$ and $Ca_d$ in our experiments the critical value of $q$ (and $Ca_d$) were different by 35% to 70% between the N and NN systems). Figure 5 shows the impact of the elastic forces on the transition.
D. The effect of the elasticity

It is known that above a critical length of the molecule, linear polymers can exist in two stable states: coiled and stretched.\textsuperscript{41} Theory predicts the coil-stretch transition to occur\textsuperscript{42, 43} at $\dot{\varepsilon} \lambda \approx 0.5$. Various observations and numerical simulations affirmed the onset of polymer stretching and the consequent non-Newtonian effects to emerge at this condition.

For example, Perkins et al.\textsuperscript{44} demonstrated that DNA molecules in the hyperbolic point (stag-nation point) of a microfabricated cross-slot geometry start to uncoil above a critical De number of $D_{e\text{crit}} = 0.4$. Schroeder et al.\textsuperscript{41} confirmed this observation and showed a hysteretic coil-stretch transition between $D_{e} = 0.4$ and 0.5. For bulk liquids, rather than for single polymer molecule Poole et al.\textsuperscript{45} demonstrated by numerical simulations that the flow of the simplest differential viscoelastic fluid model (so-called upper-convected Maxwell model, UCM) undergoes spatial symmetry breaking and bistability at $D_{e\text{crit}} \approx 0.31$ when flowing in a cross-slot geometry. In their work they concluded that this asymmetry is purely elastic in nature and that the effect of inertia is a stabilizing one. Arratia et al.\textsuperscript{21} previously experimentally observed this instability at higher values of $D_{e}$ ($D_{e} \approx 1.8$).

We note that the mentioned studies use the Deborah number for characterization of the viscoelastic flow defined as $D_{e} = \lambda / \tau$, where $\lambda$ is the relaxation time of the polymer and $\tau$ is the typical flow time of the fluid usually defined as the inverse value of the extensional rate $\dot{\varepsilon}^{-1}$. The resulting $D_{e}$ number is then actually identical with our Weissenberg number $W_{i} = \dot{\varepsilon} \lambda$, allowing us to make direct comparisons.\textsuperscript{45}

In our experiments the dripping-jetting transitions in viscoelastic continuous phases occur between $D_{e} \approx 0.3$ and 1.5 (Fig. 6). This suggest that the dripping-jetting transition occurs when the polymer molecules starts to uncoil. Although the long polymer chains are most likely only partially stretched\textsuperscript{46} the effect still gives rise to elastic forces in the bulk liquid and enhance the effect of the focusing. Surprisingly, we found the same quantitative difference of reduction of the capillary number for all three elastic liquids (regardless of the $E_{l}$ number varying over two orders of magnitude and relaxation times varying over a similar span) with respect to the Newtonian phase of the same shear viscosity (Fig. 5). Both of these results suggest that the $W_{i} (D_{e})$ dimensional group govern the dynamics of our systems.

E. Rate of generation of droplets

For practical applications the important parameters of the dynamics of the droplet generator include the size of the droplets, their dispersion, and the rate (frequency) at which they are produced. In general, the size (diameter) of the droplets is inversely proportional to the frequency of the breakup.\textsuperscript{47} The satellite droplets can make it difficult to scale the average diameter of the droplets,
especially when the size of the satellites cannot be neglected and/or multiple satellites are formed. We thus choose to investigate the frequency of formation instead of the droplet diameter and we define the frequency of formation as the frequency of generation of the large (primary or mother) droplets, while the satellites enter only into the widened distribution of sizes. We found it informational to decouple the frequency of formation of the primary droplets from characterization of the number and distribution of volumes of the satellites. We recorded high speed videos of the processes of generation of droplets and recorded the instances at which the primary droplets were formed. Then we calculated the average frequency of formation as \( f = N/\tau \), where \( N \) is the number of primary droplets recorded within a particular interval \( \tau \). In the dripping regime we did not observe any significant differences between the Newtonian and viscoelastic continuous liquids in the formation of droplets. In both cases we observed the same power law dependence of the frequency of breakup \( f \) on the ratio of flows \( q \), which scales as \( f \sim A_q^{2/3} \) and approaches to \( f \sim A_q \) in the case of the most viscous outer phases (N1, NN1) (see Fig S5 of the supplementary material)\(^{28}\), where constant \( A \) depends on the geometry of the device and on the capillary number of the inner phase as reported by others.\(^{48}\)

This observations are consistent with earlier reports,\(^{49, 50}\) that also found that droplet size exhibit power law behavior at low values of the capillary numbers. These forms of the scaling of the droplet size suggest that not only the pressure drop along the growing droplet (which dominates in purely squeezing regime\(^{3,51-54}\)) but also shear stress exerted on the growing droplet by the flow of the continuous phase play a role in the process of the formation of droplets. The frequency of breakup in the dripping regime is relatively low (on the order of several hertz’ to tens of Hz) and the system produces large drops of diameter larger than the width of the outlet channel or comparable to it.

When the speed of flow in the channel becomes sufficiently large, the tip of the droplet phase no-longer recoils into the junction after break-up, but rather extends into the outlet channel to form a jet. Further increase of the rate of flow results in an elongation of the jet, so that it can be several times longer than the orifice is wide. The mechanism of breakup of droplets in the jetting regime for falling liquid jets (in surrounding air) is driven by the Plateau-Rayleigh instability and is determined predominantly by the jet radius\(^{55}\) but in co-flowing fluid stream the precise contribution of the rheological properties of the two immiscible liquids, their rates of flows and the geometry of the device is still not fully understood.\(^{37, 56}\)

We investigate the rate of production of droplets (frequency of generation) just after the system transits from the dripping to the jetting regime. We found that depending on the ratio of viscosities \( \beta' = \mu_d/\mu_c \) and on the ratio of flow rates \( q = Q_c/Q_d \) two classes of transition can occur.
FIG. 7. Frequency $f$ of break-up plotted against the ratio of flows $q = Q_c/Q_d$ for selected viscosities of the droplet phase. Silicon oils of viscosity 20 cSt (circles) and 200 cSt (squares) are presented focused with Newtonian liquids (open symbols, dotted lines) and non-Newtonian liquids (full symbols, solid lines) of various viscosity (and elasticity in the case of viscoelastic focusing). We observed two classes of dripping-jetting transition: “continuous” – when the frequency of generation of droplets increases gradually as the system transits to the jetting regime, and “discontinuous” – when the frequency of generation of droplets jumps suddenly upon transition to values order(s) higher as it was in the dripping regime.

At high $q$ and low viscosity outer phase (high viscosity ratio $\beta'$) the breakup is fast (up to two orders of magnitude higher frequency as it was in the dripping regime) and produces small droplets.

In the tested range of $Ca_c$ numbers ($Ca_c \sim 10^{-4} - 10^{-1}$) in Newtonian outer phase we observed only a discontinuous transition, i.e., the frequency of generation of droplets suddenly increases by a factor of 10 or 100. In the NN focusing fluids the transition to jetting occurs at lower capillary numbers and in a continuous manner (Fig. 7.). If in the viscoelastic focusing system the transition is discontinuous than in the Newtonian system we observe only the dripping regime (i.e., we do not observe jetting). For example, at $Q_d = 100 \, \mu L/h$ we observed only discontinuous transition over the whole range of viscosity of the inner phase ($\mu_d = 20 - 500$ cSt; $\beta' = 3 - 0.12$) when we used the NN2 (medium viscosity/medium elastic Boger fluid) and only dripping regime for the Newtonian focusing phase even we increased $q$ to 80 (see Fig. S6 of the supplementary material). Interestingly the scaling of frequency in the Newtonian focusing liquids follows a similar trend as described by the continuous transition to jetting observed in the experiments with the Boger liquids.
FIG. 8. Polydispersity (coefficient of variance, CV) of droplet sizes focused with Newtonian (left column, open symbols) and non-Newtonian (right column, full symbols) liquids. The inner phase was PDMS oil with kinematic viscosities of 5 cSt (diamonds – green), 20 cSt (circles – red), 50 cSt (triangles – blue), 200 cSt (squares – black), and 500 cSt (stars – orange). Droplets produced in the dripping regime are monodisperse (CV < 1.5%) for both focusing systems. The dispersion increases suddenly as jetting occurs and show clear dependence of the ratio of viscosities while focused with viscoelastic continuous phase.

F. Distribution of volumes of the primary droplets

The breadth of the distribution of volumes of the primary droplets is significantly affected by the elastic effects in the flow of the continuous liquid. For each fixed conditions of flow we measured the areas of ~50-100 droplets and calculated the coefficient of variance \( CV(\%) = (\sigma / A_d) \times 100 \), defined as the standard deviation (\( \sigma \)) of the area of the droplets normalized by their average area (\( A_d \)).

Both in Newtonian and non-Newtonian systems the droplets produced in the dripping regime are monodisperse—the value of \( CV \) falls well below 1.5% when droplets are formed in the low-to-medium viscosity outer liquids (\( \mu_d = 30-60\text{cP} \)). In viscous (\( \mu_d = 178\text{cP} \)) continuous phase this value of \( CV \) is somewhat higher (\( CV \sim 5\% \)) and grows gradually with \( q \) both in Newtonian and non-Newtonian phases (N1, NN1) (Fig. 8).

Our observations also confirm the earlier reports\(^{56}\) that in the dripping regime higher viscosity of the inner liquid promotes the formation of more monodisperse droplets. Interestingly, we find that in the jetting regime higher viscosity of the droplet phase results in less monodisperse droplets, and as we show it depends on the rheological properties of the outer liquid.

As the system transit to jetting regime the polydispersity becomes significant, due the nonlinear breakup rising from the Rayleigh-Plateau instability. In the case of a Newtonian focusing phase (Fig. 8, left column) the dispersion of volumes of the droplets in the jetting regime is independent of the ratio of viscosities \( \beta' \) and can be treated as constant with a value of \( CV \sim 20\% \). For highly viscous continuous phase we observed dependence only on the ratio of rates of flow. In the case of
viscoelastic focusing the low elasticity focusing phase (NN3) show similar behavior as its Newtonian counterpart, while in medium elasticity (NN2) and strongly elastic fluids (NN1) the CV in the jetting regime increases both with \( q \) and with the viscosity of the inner phase and is dependent on the elasticity. The polydispersity index is found to reach similar value (\( CV \sim 20\% \)) as in the Newtonian focusing phase of the same viscosity when the ratio of viscosity \( \beta = 1 \), and can reach much higher values when \( \beta' \) is high (Fig. 8, right column). On the other hand, at low ratios of viscosity (\( \beta' < 1 \)) the dispersion is lower than in the corresponding Newtonian liquid which leads to conclusion that viscoelastic liquids with sufficient elasticity can be a useful tool to decrease the dispersion in the jetting regime by adjusting the ratio of viscosity of the two phases.

G. Satellite droplets

Near to the break-up point viscous droplets develop long necks that further split into a series of smaller droplets, while the ratio of viscosity significantly affects the necking and breakup process of the separating droplet thus the formation of satellites droplets.\(^{57}\) At low viscosity ratios the breakup process is fast—droplets detach at the stagnation point of the junction, whereas with increasing viscosity ratio the time of the breakup rises, the breakup point moves further in the direction of flow (downstream channel) and a thread between the primary drop and the inlet channel forms. After the breakup of the primary droplet secondary breakup of the thread occurs under the unbalanced capillary forces—causing the formation of single satellite and at higher rate of flow of the disperse phase a series of satellite droplets.

The number and the size of the satellite droplets increase with increasing \( q \) both in the case of the Newtonian and the non-Newtonian focusing. The individual regimes (and so the transitions between them) occur at lower ratios of flows in the non-Newtonian system, therefore at a given ratio of flow rates the number of satellite droplets is usually higher when focusing with the viscoelastic liquid. Nonetheless, the maximum number of satellite droplets the system can produce is always higher in the Newtonian continuous liquid. At a particular contrast of viscosities (e.g., \( \beta = \mu_d/\mu_c \approx 0.1-1 \)) the number of satellites may be as high as 10 or more. In the non-Newtonian continuous phase, the system transits into jetting earlier—when the number of satellite droplets reaches \( \sim 5 \).

The evolution of the number and the size of the satellites is similar in both the Newtonian and non-Newtonian systems (Fig. 9). The thread between the primary drop and the inlet channel in Newtonian ambient liquid breaks into several drops under capillary insabilities,\(^{58}\) whereas in the presence of additional elastic forces tend to lower the total number of satellites while increasing their size (especially that of the primary satellite droplet). When the viscosity of the droplet phase is less than the viscosity of the focusing phase (\( \beta' < 1 \)), multiple satellites start to form at similar value of \( q \) in both systems and their number increases with increasing \( q \). In contrast, when the viscosity of the droplet phase exceeds the viscosity of the continuous phase (\( \beta > 1 \)) the number of satellites decreases dramatically and at \( \beta' \sim 3.5 \) the multiple satellite regime become very narrow and restricted to the immediate preludium to the dripping-jetting transition. In the strongly elastic continuous phase (NN1) this effect is even more pronounced, starting at ratio of viscosities \( \beta' \sim 2.5 \). An interesting phenomena was observed in the viscoelastic continuous phase at \( \beta' \approx 1 \) as described above—increase of the elasticity of the continuous phase favors the formation of a series of satellites.

For demonstration, we plotted the evolution of sizes and numbers of the satellite droplets with respect to the mother droplet as a function of \( q \) for selected viscosities of the inner phase in Fig. 9. The sizes (area) of the mother droplet are normalized by the cross section area of the channel, while the area of the satellite droplets is non-dimensionalized by the area of the mother droplets. Interestingly, we find that the dripping-jetting transition occurs when the size of the first satellite approaches (to within \( \sim 25\% \)) the area of the primary droplet (i.e., \( \sim 12.5\% \) of the volume of the primary droplet) which is in good agreement with previous observations.\(^{59}\)

H. Effect of the geometry of the junction

When a viscoelastic liquid is forced to flow out a narrow orifice it may expand sideways (as in the die-swell effect). In addition, turbulences, vortices, and eddies may occur at the outlet even at low
FIG. 9. Evolution of numbers and sizes of the satellite droplets with respect to the mother droplet focusing with strongly elastic liquid, NN1 (right, full symbols) and with Newtonian liquid of the same viscosity, N1 (left, open symbols). Micrographs show the distribution of satellite droplets right before the transition to the jetting regime takes place. Elastic effects of the continuous phase tend to lower the number of the satellites while increasing their size (especially of the primary satellite). Dashed lines indicate the dripping-jetting transition.

Reynolds numbers. We tested the idea to use this vortices to increase the frequency of the breakup and to decrease the size of the droplets. Except for the simple cross junction (Fig. 1(b)/FF-1) that had the inlet and outlet channels of the same dimensions we also investigated two other geometries of the flow focusing droplet generator.

In geometry FF-2 we inserted an orifice of dimensions of $100 \times 100 \times 250 \, \mu m$ (width $\times$ length $\times$ height) and left the outlet channel unchanged (Fig. 1(b)/FF-2). We tested the operation of this device with the 20 cSt and 200 cSt silicon oils as the droplet liquids and with either N1 and N2 or NN1 and NN2 liquid as the continuous phase. We varied the rate of flow of the focusing phase from $0.25$ to $4000 \, \mu L/h$ while the rate of flow of the disperse was fixed at $100 \, \mu L/h$. The Reynolds number $Re_c = \rho_c \cdot Q_c \cdot \mu_c \cdot w_{or}$ at the orifice varied between $4.8 \times 10^{-4}$ and $7.7 \times 10^{-2}$, and the corresponding Weissenberg number $Wi = \frac{Q_c}{w_{or}}$ was in the range of $0.4 \leq Wi \leq 62.5$. The capillary number of the droplet phase was $7.9 \times 10^{-4}$ and $8.2 \times 10^{-2}$ for the 20 cSt and 200 cSt PDMS oils, respectively.

In the last system we used an orifice of the same dimension as in geometry FF-2 while we modified the outlet channel to a much larger width (800 $\mu m$) (Fig. 1(b)/FF-3). The rate of flow of the disperse phase $Q_d$ was $100 \, \mu L/h$ as in the previous experiments, but this time we changed the rate of flow of the focusing liquid between $100$ and $10^4 \, \mu L/h$ (ratio of flow rates $q$ varied between 1 and 100). These rates of flow correspond to Reynolds numbers $1.9 \times 10^{-3} \leq Re_c \leq 1.9 \times 10^{-1}$ and Weissenberg numbers of $1.56 \leq Wi \leq 156$, calculated for the flow within the orifice.

In both geometries we observed the same sequence of transitions as in the simple cross junction geometry: dripping regime without satellites, dripping regime with single satellite followed by
multiple satellite regime with an increasing number and size of the satellites as the function of $q$ and—finally—the jetting regime.

Comparing the two contraction geometries we used in our experiments we found that (i) in the dripping regime the dimension of the outlet channel is irrelevant to determine the size of the produced droplets (neither in Newtonian nor in viscoelastic focusing) yet (ii) in the jetting regime the wider outlet channel results in larger droplets (both in the Newtonian and viscoelastic systems). Figure 10 shows the micrographs of the flow patterns in the dripping and in the jetting regimes for both geometries in N1 and NN1 focusing liquids.

The lack of dependence of the droplet size on the geometry of the outlet channel in the dripping regime can be explained by nothing that the break up occurs inside the contraction and hence the dimension of the outlet channel is irrelevant in the determination of the sizes of the droplets. At the same time, in the jetting regime droplets break off downstream in the outlet channel and its width becomes decisive in determination of droplet volumes.

Introduction of an orifice has both qualitative and quantitative impact on the dripping-jetting transition. When the elastic liquid flows through a narrowing it causes the droplet phase to be injected through the orifice in pulses rather than continuously (Fig. 11, Video 1). We speculate that because the tip of the droplet phase occupies a significant portion of the cross section of the orifice it significantly increases the stretching of the focusing liquid. As the viscoelastic liquid opposes such deformation, the pressure in the droplet phase needs to exceed a threshold value to enter the orifice. When a series of droplets is broken off, this pressure is released and the tip recoils back upstream of the orifice. The process repeats creating pulses of droplets (Fig. 11, Video 1). In this mode the system produces series of droplets of decreasing volume generating a wide distribution of volumes in the resulting emulsion. The system with the wider outlet channel yields a smaller number of droplets in a single pulse and these droplets are larger (preserving the descending order) than in the system with the narrower outlet. This pulsatory mode of generation of droplets is most pronounced for the strongly elastic liquid, NN1 and when the ratio of viscosities $\beta_1 = \mu_d/\mu_e$ is low ($\sim 0.1$) while at high values of viscosity of the inner phase ($\beta_1 \approx 1$) the effect is only minor. This suggests that the viscous shear of the inner phase damps the instability. Moreover, even for the low ratio of viscosities the pulsation disappears when the speed of flow of the non-Newtonian liquid is increased. Simultaneously the polydispersity index ($CV$) of the droplets also decreases.

We also observe a second class of instability similar to the one caused by the presence of surfactants in the flow of bubbles in a non-Newtonian liquid. At higher rates of flow the flow in the outlet channel exhibits “beating” in the direction transverse to the axis of the channel. Analysis of the video sequence (see instability2.mov in the supplementary material) shows that this instabilities onset already in the inlet channels, where the elastic liquids in the lateral (focusing) arms exhibit temporal fluctuation—pushing the stream of the Newtonian phase back and forth in the transverse direction. This fluctuation is then transferred to the outlet channel where the droplets form a pseudo-sinusoidal string. We do not observe any drift, i.e., once set the transverse positions of the droplets
FIG. 11. Effect of the contraction of the flow focusing junction. After the transition to the jetting regime in viscoelastic medium the droplet phase is injected through the orifice in pulses. This pulsation with increasing speed of the continuous phase disappears but simultaneously a second class of (transverse) instability appears. The Weissenberg numbers on the right side of the micrographs are calculated for the geometry of the orifice. For the lateral focusing channels the corresponding values are 2.35, 4.68, 7.8, 9.36, 12.5, and 15.6, respectively. The droplets phase was 20cSt PDMS oil, the Newtonian and viscoelastic phases are the N1/NN1 fluid pair. The scale bar is 250 μm (enhanced online). [URL: http://dx.doi.org/10.1063/1.4817995.1]

do not change while they flow downstream (instability2.mov in the supplementary material28). Since this phenomena was observed only in the high elastic (NN1) liquid and only at large rates of flow, we conclude that it is partially caused by the “purely elastic instability of second-type” reported by Arratia et al.60 starting above \( De = 12.5 \). We report very similar values of the \( Wi \) number (\( Wi \equiv De \approx 10 \)) calculated for the lateral (focusing) arm-channels at which we observed the occurrence of this instability (Fig. 11). On the other hand, since this instability was not observed in the classical cross-slot geometry (without orifice) we assume that the sudden pressure drop by the presence of the orifice play also significant role—allowing buildup of elastic forces in the lateral arms.

In an overview, and in comparison to the dynamics in the simple cross slot geometry the systems with narrow orifices: (i) do not differ in the dripping regime, (ii) a smaller number of satellites is produced in the satellite regime (max. 3-4) before the transition into the jetting mode, (iii) the dripping-jetting transition and the volumes of droplets produced in the jetting regime do depend on the geometry of the orifice and of the outlet channel.

IV. SUMMARY AND CONCLUSIONS

We presented a comparative experimental investigation of the effect of the elasticity of the focusing liquid on generation of Newtonian droplets in the flow focusing droplet generators. We found that—in comparison to the Newtonian focusing liquid—elasticity produces a smoother transition between the dripping and jetting regimes, promotes transition to jetting at lower capillary numbers of the continuous phase and slightly reduces the volume of generated droplets.

Elasticity of the focusing liquid significantly affects the breadth of the distribution of volumes of the droplets. In the non-Newtonian focusing liquids, the distribution of diameters of droplets depends on the rheological properties of the outer phase and on the ratio of viscosities. In the junctions comprising a contraction we observed two distinct instabilities. In the first, right after the dripping-jetting transition takes place, droplets are injected through the orifice in pulses. We found that viscous shear of the inner phase damps this class of instability and is characteristic for low ratios of viscosity. With increasing rate of flow of the outer phase a second class of instability arose yet in the inlet (focusing) channels where the flow become unsteady and fluctuates pseudo-periodically in time. Interestingly, we observed both instabilities at similar values of the \( Wi \) number as Arratia et al.21 did in a single phase purely elastic flow in a simple cross-slot junction.

In summary, with careful adjustment of the ratio of viscosities of the two immiscible liquids, viscoelasticity of the focusing liquid can help to lower the dispersion of the emulsions and to decrease the volume of the produced droplets. Still the range of parameters that promote these effects is rather narrow and increased dispersion (with respect to the Newtonian outer phase) is characteristic for the
wide range of capillary numbers. Viscoelastic (Boger) focusing liquids may also be used to promote higher frequency of droplet production at low rates of flow.

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28 See supplementary material at http://dx.doi.org/10.1063/1.4817995 for the additional information about (i) the measured apparent extensional viscosity of the used viscoelastic liquids, (ii) the dripping-jetting transitions in our experiments in the $C_{\alpha}$-$\omega_{\alpha}$ plane, (iii) experimental data fit for the dripping-jetting transition, (iv) the rate of the formation of droplets in the dripping regime as the function of the ratio of flows, and (v) the frequency of droplets break-up plotted against the ratio of flows, respectively. The video “instability2.mov” demonstrates the formation of oil droplets in a highly elastic Boger liquid and in a Newtonian liquid of the same shear viscosity. In the Newtonian continuous phase droplets form at the centerline of the channel, while in the elastic liquid droplets form a pseudo-sinusoidal string. The droplet phase is a
20:St PDMS oil, the Newtonian and viscoelastic phases are the N1/NN1 fluid pair. The flow rates of the droplet phase and the continuous phases are $Q_d = 100 \mu$L and $Q_c = 2500 \mu$L/h, respectively.


