Motion and deformation of a droplet in a microfluidic cross-junction

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In this paper we investigate computationally the transient deformation of a droplet flowing along the centerline of a microfluidic cross-junction device. We consider naturally buoyant droplets with size smaller than the cross-section of the square channels comprising the cross-junction, and investigate low-to-strong flow rates and a wide range of fluids viscosity ratio. Our investigation shows that the intersecting flows at the cross-junction act like a constriction, and thus the droplet shows a rich deformation behavior as it passes through the micro-junction. Our work highlights the three-dimensional effects of the asymmetric microfluidic geometry on the droplet deformation, and the different effects of the viscosity ratio on the droplet's overall length scales and the local length scales at the droplet edges. The large edge curvatures and thus the small local length scales developed transiently, especially at the tail of low-viscosity droplets, reveal that the current investigation is a multi-length interfacial dynamics problem.

1. INTRODUCTION

The study of the interfacial dynamics of droplets and bubbles in micro-channels of various configurations has been intrigued by the recent development of droplet-based (or digital) microfluidic devices. These devices have a wide range of applications including well-controlled micro-reactors and micro-engines [2, 35], fabrication of micro-particles with desirable properties [5, 6, 22, 30, 43], and lab-on-a-chip devices for clinical and environmental monitoring and analysis [11, 29]. Digital microfluidics utilize significantly reduced fluids volumes and processing times, while they offer easy parallel processing and thus allowing large data sets to be acquired efficiently [2, 11, 35].

Besides microfluidics, understanding the behavior of non-wetting droplets in micro-channels is also of great interest for many industrial and biomedical processes. In enhanced oil recovery, aqueous liquids or gaseous foams flow through the underground porous media to displace the trapped oil, and the success of such operations depends on understanding of the multiphase flows through the porous matrix [9, 24]. In the lubrication process, lubricants usually contain a small amount of immersed bubbles altering the performance of journal bearings and squeezing film dampers [31]. The fundamental physics of bubble behavior in micro-channels is also essential in the operation and design of fuel cells, e.g. the removal of CO₂ bubbles in the anode channel of a direct methanol fuel cell (DMFC) [21]. In addition, micro-bubbles are utilized as drug delivery carriers in vascular microvessels [38].

A plethora of studies has investigated the motion of droplets and bubbles in cylindrical tubes and rectangular channels, considering small- and large-size droplets or fluid fingers, e.g. [1, 3, 15, 25, 41]. In addition,

microfluidic co-axial injections along with T- and cross-junctions have been commonly used for the generation of controlled-sized droplets, e.g. [2, 7, 12, 20, 36, 42]. A typical co-flow device contains an inner tube aligned with an outer channel and two streams flowing in parallel near the nozzle, while the inner fluid breaks into droplets due to dripping or jetting mechanisms [2]. Droplets of controllable size can also be formed when two immiscible fluids enter a T- or cross-junction through different inlet channels [12, 19, 20]. By combining droplet generation with a cross-linking agent in a microfluidic device, several studies achieved fabrication of microparticles or microcapsules with desirable size and properties, e.g. [6, 22, 28, 30].

In this paper we investigate computationally the transient deformation of a droplet flowing along the centerline of a microfluidic cross-junction device. In particular, we consider droplets with constant surface tension which are naturally buoyant in the surrounding fluid, and have size smaller than the cross-section of the square channels comprising the cross-junction. Our investigation involves low-to-strong flow rates and a wide range of fluids viscosity ratio.

Studying the shape of multi-phase soft particles like droplets in micro-junctions provides useful information on the utilization of these particles in chemical, pharmaceutical and physiological processes. For example, understanding the stability of soft particle shapes provides helpful insight on the hydrodynamic aggregation and the effective viscosity of suspensions [1, 17]. Such studies also provide useful insight for a wide range of industrial and biomedical applications including enhanced oil recovery, lubrication processes, design of fuel cells, drug delivery, micro-particle fabrication as well as lab-on-a-chip devices for clinical and environmental monitoring and analysis [2, 24, 38].

The shape of a droplet flowing inside a micro-junction is determined by the nonlinear coupling of the restoring interfacial forces and the deforming hydrodynamic forces

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of the intersecting flows. Therefore, the droplet's transient conformational behavior depends strongly on the strength of the surrounding flow rates, the droplet's surface tension, and the fluids viscosity ratio. Our work highlights the three-dimensional effects of the asymmetric microfluidic geometry on the droplet deformation, and the different effects of the fluids viscosity ratio on the droplet's overall length scales and the local length scales at the droplet edges. In addition, our study highlights the multi-length nature of the current interfacial investigation.

2. PROBLEM DESCRIPTION

We consider a three-dimensional droplet flowing along the centerline of a microfluidic cross-junction as illustrated in figure 1. The cross-junction is constructed from two intersecting square microfluidic channels with cross-section half-lengths $\ell_y = \ell_z$. The origin of the coordinate system is placed at the center of the junction. To facilitate our description, we imagine one channel as horizontal and the other as vertical, as illustrated in figure 1. Thus, the x -direction corresponds to the channel's or droplet's length, the z -direction will be referred to as height while the y -direction will be referred to as width (of the channel or the droplet). The flow is incoming at the left (upstream) inlet of the horizontal channel as well as at the two inlets of the vertical channel, and exits only at the right (downstream) outlet of the horizontal channel.

The droplet's interior and exterior are Newtonian fluids, with viscosities $\lambda\mu$ and μ , respectively, and the same density. The droplet has a constant surface tension γ while its size a is specified by its volume $V = 4\pi a^3/3$ and is comparable (or smaller) to the channel half-height ℓ_z .

At time $t = 0$ the droplet is located upstream of the cross-junction in the horizontal channel having a spherical shape with radius a and centroid $x_c = -3\ell_z$. That instant, the flow is turned on inside the microfluidic device and we investigate the transient dynamics of the droplet as it enters and exits the microfluidic cross-junction which occupies the x -region $[-\ell_z, \ell_z]$. We emphasize that the specific choice for the droplet's initial position upstream of the junction does not affect the droplet deformation and motion inside the junction or downstream of it, i.e. we obtained identical results even for droplets placed further upstream of the junction.

Far upstream from the droplet in the horizontal microchannel, the flow approaches the undisturbed flow $\mathbf{u}^\infty$ in the square channel characterized by an average velocity $\mathcal{U}$ and constant flow rate $Q = 4\mathcal{U}\ell_z^2$. (The exact form of the channel's velocity field $\mathbf{u}^\infty$ and its average velocity $\mathcal{U}$ are given in the supplementary material of this paper.) The incoming flow rate at each of the two inlets of the vertical microchannel is Q_v , and thus the horizontal channel after the cross-junction has a constant flow

rate $(Q + 2Q_v)$. Due to symmetry, the neutral droplet flows along the centerline of the horizontal microchannel but its motion and deformation is affected by the flows in both channels of the cross-junction. We assume that the Reynolds number is small for both the surrounding and the inner flows, and thus the droplet deformation and motion occurs in the Stokes regime.

The present problem depends on four dimensionless parameters: the droplet size (relative to the channel height) a/ℓ_z , the relative lateral flow rate Q_v/Q , the viscosity ratio λ , and the capillary number Ca defined as

$$Ca = \frac{\mu\mathcal{U}}{\gamma} \quad (1)$$

where $\mathcal{U}$ is the average undisturbed velocity in the horizontal channel before the cross-junction. It is of interest to note that the capillary number, as defined by Eq.(1), does not contain any length scale, and thus it may be considered as a dimensionless flow rate. In this study, the channel's half-height ℓ_z is used as the length scale, the velocity is scaled with the average undisturbed velocity $\mathcal{U}$ of the upstream horizontal channel, and thus the time scale is $\tau_f = \ell_z/\mathcal{U}$.

To determine the droplet deformation and motion in the microfluidic device, we employ our fully-implicit spectral boundary element algorithm for interfacial dynamics in Stokes flow [8]. The interested reader is referred to the supplementary material of this paper and our earlier papers for more details on our spectral boundary element algorithms for interfacial problems of free suspended droplets, droplets in proximity to solid surfaces and droplet motion in micro-channels [8–10, 32, 40].

We emphasize that our spectral boundary element methods have been used to study droplet motion in solid channels of different cross-sections and we have made successful comparisons with published experimental and computational results, as we discussed in detail in our earlier paper on droplet motion in a square microfluidic channel [40]. In addition, our spectral boundary methods have been used to study capsule motion in different microfluidic geometries, including square and rectangular channels and constrictions [16, 17, 26].

To verify the accuracy of our results for the present problem, we performed convergence runs covering the entire interfacial evolution with different grids for several representative cases. Our convergence runs showed that our results for the interfacial shape are accurate to at least 3 significant digits except for the most challenging cases studied in this work (e.g. the largest, very deformed droplets with $a = 0.7$, $Ca = 0.4, 0.5$ and $Q_v/Q = 0.5$) where the interfacial shape was determined with an accuracy of at least 2 significant digits.

In our computational work, we have investigated a wide range of all the problem parameters, including flow rate $Ca \leq O(1)$, droplet size $a/\ell_z = 0.5\text{--}0.9$, viscosities ratio $\lambda = 0\text{--}20$, and relative lateral flow rate $Q_v/Q \leq O(1)$. We emphasize that we consider flow rates which do not result in interfacial breaking. This range of

dimensionless parameters can readily be used in experimental microfluidic systems. For example, Tice *et al.* investigated microfluidic droplet formation and mixing at low capillary numbers $Ca = 0.0014\text{--}0.011$ [37]. In their synthesis of CdSe nanocrystals in microfluidics, Chan *et al.* generated micro-droplets up to $Ca = 0.81$ at low viscosity ratios [6]. Cubaud and Mason studied the formation of threads containing highly viscous liquids utilizing viscosity ratios $\lambda = 20\text{--}1500$ [7]. Ha and Leal investigated micro-droplet deformation with capillary number $Ca = O(1)$ and viscosity ratio $\lambda = 0.005\text{--}2.2$, by employing polydimethylsiloxane (PDMS) as the surrounding fluid and polyalkylene glycols as the droplet fluid [13]. Nie *et al.* studied microfluidic emulsification of silicone oils in water for viscosity ratio in the range $\lambda = 10\text{--}500$ [23].

A. Definition of Geometric and Physical Variables

To describe the droplet's three-dimensional deformation, we consider several geometric properties including the droplet dimensions and profile curvatures at the droplet edges. In particular, we determine the droplet projection lengths along the three axes, L_x , L_y and L_z as the maximum distance in the x , y and z coordinates of the droplet surface. For a given droplet shape, we determine accurately these three dimensions by employing a Newton method to solve the optimization problems using the spectral discretization points on the droplet interface. Our results are expressed as functions of the droplet centroid $\mathbf{x}_c = (x_c, y_c, z_c)$ where $y_c = z_c = 0$ for this problem owing to the centerline motion.

In addition, to account for the local interfacial length scales at the droplet edges, we calculate the curvatures C_{xz} and C_{xy} at both the downstream and upstream edges of the droplet (i.e. its intersections with the x -axis) by employing our spectral discretization at the middle point of the downstream and upstream spectral elements. Observe that, at each droplet edge, both C_{xz} and C_{xy} are line curvatures, determined along the interfacial cross-section with the $y = 0$ and $z = 0$ planes, respectively, while they take on positive values for the spherical quiescent shape of the droplet where $C_{xz} = C_{xy} = a^{-1}$.

As the droplet moves in the channel, its volume-average velocity is determined from surface properties, i.e.

$$\mathbf{U} \equiv \frac{1}{V} \int_V \mathbf{u} dV = \frac{1}{V} \int_{S_d} (\mathbf{u} \cdot \mathbf{n}) \mathbf{x} dS \quad (2)$$

where V and S_d are the droplet's volume and surface area, respectively, $\mathbf{u}$ is the velocity field (either inside the droplet volume or on its surface), and $\mathbf{n}$ is the unit normal on the droplet surface pointing into the surrounding fluid.

To describe the droplet shape, we adopt the standard definition of geometric shapes (e.g. polygons). Thus we call the droplet's rear edge as convex when the radius of curvature at the middle of the rear edge points inside the

droplet (i.e. the local curvature is positive); in the opposite case the edge shape is concave. In addition, seeing the droplet from the negative y -axis or the positive z -axis represents a front view or a top view, respectively. The three-dimensional droplet views included in this paper were derived from the actual spectral grid by spectrally interpolating to a denser grid and using orthographic projection in plotting.

3. RESULTS

We begin the presentation of our results by considering the transient shape of a droplet with size $a/\ell_z = 0.7$, viscosity ratio $\lambda = 0.2$, capillary number $Ca = 0.4$, and relative lateral flow rate $Q_v/Q = 0.5$, as it deforms inside the micro-device from its original spherical shape, shown in figure 2.

At this flow rate, the droplet shows a rich deformation behavior as it moves inside the microfluidic device. First, the droplet assumes a bullet-like shape before the cross-junction, as shown in figure 2 for $x_c/\ell_z = -1.5$, which is a typical shape for droplet motion in axisymmetric-like solid ducts, such as cylindrical or square channels [2, 24, 40]. When the droplet is inside the cross-junction, its deformation increases owing to the intersecting vertical flows, and the droplet obtains a pointed downstream tip and a flattened rear, as seen in figure 2 for $x_c/\ell_z = -0.07$. Both actions result in an increased surface-tension force on the droplet surface which tries to balance the increased deforming hydrodynamic forces inside the microfluidic cross-junction. To explain this, observe that with respect to the droplet centroid, the surrounding fluid flows in the negative horizontal direction (i.e. towards upstream), and thus on the droplet interface, the surface tension force along the horizontal flow direction scales as

$$F_\gamma \sim \gamma \left(\frac{1}{R_d} - \frac{1}{R_u} \right) \quad (3)$$

where R_d and R_u are the radius of curvature at the droplet's downstream and upstream tips, respectively [2, 16, 18]. Since both droplet edges are convex, the local surface-tension force points inside the droplet, and thus it is positive for the downstream tip and negative for the upstream tip, as shown in Eq.(3). Observe that for a spherical droplet the net surface-tension force is zero, while the droplet needs to increase its downstream tip curvature and decrease its upstream tip curvature so that the restoring surface tension force is increased. Thus, the droplet obtains a deformed bullet shape with a pointed front and a flattened rear as seen in figure 2 for $x_c/\ell_z = -0.07$.

As the droplet exits the junction, it becomes elongated with a pointed downstream tip and a flattened rear along the front view, as shown in figure 2(a, c) for $x_c/\ell_z = 0.63$, owing to the incoming flows from the vertical channels

and thus the increased hydrodynamic forces exerted on the droplet interface.

When the droplet moves further downstream while its rear exits the cross-junction, the droplet's downstream edge becomes more rounded and it is its rear which is now pointed, as seen in figure 2(a, c) for $x_c/\ell_z = 2.45$. Owing to the fast incoming vertical flows, the droplet now moves slower than the surrounding fluid (as we will show shortly later in figure 4). Thus, with respect to the droplet centroid, the surrounding fluid flows in the positive horizontal direction (i.e. towards downstream), and in this location the droplet needs to obtain an inverse bullet-like shape for hydrodynamic stability reasons. It is of interest to note that at $x_c/\ell_z = 2.45$, the droplet obtains a very pointed rear with an increased tip curvature along the front view to account for the increased local hydrodynamic forces since the droplet rear is at the end of the cross-junction.

Far downstream of the cross-junction, the droplet gradually obtains a more deformed bullet-like shape owing to the increased flow rate in the downstream square horizontal channel, i.e. the increased local capillary number $\text{Ca}^{\text{eff}} = \mu 2U/\gamma = 2\text{Ca}$, as seen in figure 2 for $x_c/\ell_z = 7$.

The droplet dynamics along the front view reveals that on this plane the intersecting flows at the cross-junction act like a constriction, created by the streamlines of the vertical intersecting flows, and thus the droplet needs to be compressed inside and outside of this constriction. Observe that the droplet profiles shown in figure 2(c) can be used to imagine the constriction shape being parallel to the top and bottom sequence of the droplet profiles. Similar but axisymmetric shapes have been observed for pressure- and buoyancy-driven droplet motion inside tubes with axisymmetric constrictions, for the same physical reasons, e.g. [14, 25].

However, in our problem the asymmetric shape of the cross-junction gives rise to a different sequence of droplet shapes along the top view, as seen in figure 2(b). Excluding the axisymmetric-like bullet shape upstream or far downstream of the junction in the square horizontal channel, the droplet has obtained a clearly three-dimensional shape inside and close downstream of the cross-junction. Inside the junction, the droplet remains laterally extended along the top view with a pointed front and a flat rear profile, i.e. it remains bullet-shaped, as shown in figure 2(b) for $x_c/\ell_z = 0.63$. In addition, its rear profile remains flat or becomes slightly concave from top when the droplet rear exits the junction even though the droplet forms a very pointed convex rear in the front view, as seen figure 2(b) for $x_c/\ell_z = 2.45, 3.55$. Clearly, the droplet shows a rich three-dimensional deformation behavior as it flows through the microfluidic junction that cannot be described from single-view observations in microfluidic experimental systems.

A. Effects of flow rate

We present now the effects of the capillary number Ca (or flow rate) on the droplet deformation and motion inside the microfluidic device. In particular, we consider a droplet with size $a/\ell_z = 0.7$, viscosity ratio $\lambda = 0.2$, relative lateral flow rate $Q_v/Q = 0.5$, and investigate its transient dynamics for small and moderate capillary numbers, $\text{Ca} = 0.01, 0.1, 0.2, 0.4, 0.5$. We quantify our results by presenting the evolution of the droplet's lengths and the edge curvatures as a function of the droplet centroid x_c .

Figure 3(a) shows the evolution of the droplet's length L_x as the droplet shape changes significantly inside the micro-device, seen earlier in figure 2. Starting from the initial spherical shape, the droplet's length L_x increases only slightly upstream of the cross-junction where the droplet obtains a bullet shape. When the droplet enters the cross-junction, the stronger hydrodynamic forces owing to the intersecting vertical channels cause a significant droplet elongation. The droplet continues to elongate as it exits the cross-junction, achieving its highest length near $x_c/\ell_z = 2$ for the higher flow rates studied, i.e. when its tail exits the junction and the droplet has obtained an elongated inverse-bullet shape with a pointed rear, as seen in figure 2(a, c) for $x_c/\ell_z = 2.45$. Afterwards, the droplet length L_x is gradually reduced as the droplet obtains a bullet shape in the downstream square channel far from the cross-junction. The monotonic increase of the droplet's length L_x with the flow rate Ca (owing to the higher hydrodynamic forces) can be very significant especially for the higher flow rates studied which cause the droplet's transient length to double.

In a similar manner, the droplet's height L_z decreases significantly when the droplet is inside the cross-junction (to accommodate the strong local hydrodynamic forces), and gradually increases after the droplet has passed the junction, as shown in figure 3(b). In practice, the droplet needs to obtain a slender-like shape inside the junction (with a length ratio $L_x/L_z \approx 4$ for the higher flow rates studied) to pass the junction, as seen in figure 2 for $x_c/\ell_z = 2.45$. The strong hydrodynamic forces due to the cross-junction also cause a reduction of the droplet width L_y (which usually cannot be observed in microfluidic experimental systems). However, the width reduction is not as significant as the height reduction inside the junction. As shown in figure 3(b, c), even when the droplet is at the end of the cross-junction (i.e. when $x_c/\ell_z = 1$), its height has been reduced by nearly 50% but its width by only 15% for the two higher flow rates studied. Only when the droplet is sufficiently downstream of the junction, it starts to obtain a deformed axisymmetric-like bullet shape with $L_y \approx L_z$. Note that the sharp variation of both lengths, L_z and L_y , at their minimum value results from the sudden change of the lengths location from the tail area to the droplet front, as seen in figure 2.

We consider now the flow rate effects on the droplet velocity. Figure 4 shows that, by increasing the capil-

lary number Ca , the droplet velocity U_x is practically unchanged upstream of the cross-junction, then it shows a fast increase inside the junction, while after the cross-junction the more deformed droplets at the higher flow rates travel faster downstream.

Owing to the axisymmetric-like shape, the droplet velocity in the upstream and downstream square channels can be explained based on findings from droplet motion in cylindrical vessels. In particular, the droplet's velocity can be approximated by the average velocity of the inner fluid based on the laminar annular flow of two concentric fluids in a tube

$$\frac{U_x}{U} \approx \frac{2\lambda + (1 - 2\lambda)\delta^2}{\lambda + (1 - \lambda)\delta^4} \quad (4)$$

where δ is the dimensionless half-height of the droplet in the square channel, i.e. $\delta = L_z/2R$ [18]. It is of interest to note that the droplet velocity in the microfluidic junction as well as Eq.(4) result from the combined effects of the surrounding and inner fluids' shear stresses on the droplet interface.

Upstream of the cross-junction, increasing the flow rate Ca has little effect on the droplet height L_z and thus on its velocity U_x , as shown in figures 3(b) and 4. However, downstream of the junction, owing to the intersecting vertical flows, the increased flow rate causes a significant droplet deformation and thus a significant reduction of the droplet height as seen in figure 3(b). Observe that Eq.(4) predicts that for the parameters studied here, U_x increases monotonically with decreasing δ or L_z . Thus, in the downstream square channel, increasing the flow rate Ca results in an increase of the velocity of the more deformed droplets, as shown in figure 4.

It is important to observe in figure 4, that when the droplet is inside the cross-junction (which occupies the scaled x -region $[-1, 1]$), the droplet velocity is still much lower than its velocity downstream of the junction. This suggests that inside the junction, the droplet does not move as fast as the average surrounding fluid which has an increased velocity owing to the intersecting vertical flows that revert into the horizontal direction inside the junction. Therefore, in that location, and for an observer on the droplet centroid, the surrounding fluid flows towards the downstream channel direction, i.e. the positive x -direction. This justifies the droplet's inverse-bullet shape inside the junction for interfacial stability, as discussed earlier for figure 2.

We conclude this section by mentioning that we have also studied the effects of the droplet size a by considering small and moderate-sized droplets with $a/\ell_z = 0.5$ – 0.95 for viscosity ratio $\lambda = 0.2$ and capillary number $Ca = 0.1$. As the droplet size increases for a fixed flow rate, its deformation increases monotonically owing to the increased hydrodynamic forces in the narrower gap between the droplet interface and the channel walls. In addition, the narrower gap with increasing droplet size results in a reduction of the droplet velocity, in agreement with Eq.(4); thus, larger droplets take more time

to pass the cross-junction. The droplet deformation and motion for the different droplet sizes we studied is qualitatively similar to that presented in this section for size $a/\ell_z = 0.7$, and thus the corresponding figures are omitted.

B. Effects of viscosity ratio

In this section we investigate the effects of the viscosity ratio λ on the transient deformation of a droplet as it passes through the microfluidic cross-junction. For this, we consider a droplet with a fixed size and flow rates, while we vary the viscosity ratio in the range $\lambda = 0$ – 20 , i.e. we investigate the dynamics of inviscid to very viscous droplets.

Figure 5(a) presents the length evolution of a droplet with $a/\ell_z = 0.7$, $Ca = 0.1$, $Q_v/Q = 0.5$, and several values of the viscosity ratio. The transient dynamics of low-viscosity droplets (i.e. $\lambda \leq 0.01$) are independent of the viscosity ratio λ , and thus our results for $\lambda = 0.01$ also represent the limit of very low-viscosity droplets, $\lambda \ll 1$. In addition, starting from a low-viscosity droplet with $\lambda = 0.01$ and increasing the inner fluid viscosity, the droplet deformation increases until $\lambda \approx 1, 2$, and then decreases significantly with the viscosity ratio. Thus, three distinct deformation regimes are observed, i.e. for very low, moderate and high viscosity ratios λ .

In contrast to the complicated effects of the viscosity ratio on the droplet deformation, increasing the viscosity ratio λ results in a monotonic decrease of the droplet velocity along the entire micro-device as shown in figure 5(b). This is consistent with the combined effects of the surrounding and inner fluids' shear stresses on the droplet interface as also seen in Eq.(4) which predicts a monotonic decrease of the droplet velocity with the viscosity ratio for the parameters studied in this paper. It is of interest to note that high-viscosity droplets travel with about the same speed in the micro-geometry, as our results for $\lambda = 10, 20$ indicate.

To explain the rather complicated effects of the viscosity ratio on the transient droplet deformation, we need to consider that the droplet is deformed owing to the combined effects of the surrounding and inner fluids' normal stresses on the droplet interface. Therefore, the droplet's transient dynamics is characterized by the surface-tension time scale necessary to reach steady state, which at the small deformation regime was found to be

$$\tau_\gamma \sim (1 + \lambda) \frac{\mu a}{\gamma} \quad (5)$$

by the early analytical investigation of Taylor [33]. (See also section 4 in the review article by Rallison [27].) Observe that this time scale is formally valid for free-suspended droplets but also characterizes qualitatively the dynamics of small and moderate-sized droplets in duct flows as in the present study. Utilizing the dimensionless parameters valid for the current work, the

surface-tension time scale becomes

$$\tau_\gamma \sim (1 + \lambda) \text{Ca} \frac{a}{\ell_z} \tau_f \quad (6)$$

where $\tau_f = \ell_z/\mathcal{U}$ is the flow time scale and the capillary number $\text{Ca} = \mu\mathcal{U}/\gamma$ contains the surface tension γ . For non-small droplet deformations in microfluidic flows, such as the ones studied in the present paper, or for the flow-induced deformation of free-suspended droplets or droplets attached on solid plates, our computational results show that the droplet dynamics is well described by the surface-tension time scale given by Eq.(5) or Eq.(6). (See for example, figure 7 in our earlier investigation of droplets attached on solid surfaces [8].)

For low enough viscosity ratio, e.g. for $\lambda \leq 0.01$ in our problem, the inner fluid does not practically participate in the transient dynamics, and thus all low-viscosity droplets show identical evolution, in agreement with our computational results shown in figure 5.

As the viscosity ratio increases from small values until $\lambda \approx 1, 2$, both the inner and the surrounding fluids affect the droplet deformation and tend to cause a higher deformation. In this case, the time τ_γ necessary for the droplet to react to the flow changes imposed by the cross-junction is increased only moderately with λ while the droplet velocity is decreased slightly with λ . Thus, the combined hydrodynamic forces from the surrounding and inner fluids have the time to increase the transient deformation of the droplet as it moves through the junction, as seen in figure 5.

For highly viscous droplets (e.g. $\lambda \geq 5$), it is the inner fluid which mostly affects the interfacial deformation. For such droplets, as the viscosity ratio λ increases, the time τ_γ necessary for the droplet to react to the flow changes imposed by the cross-junction is increased considerably, being proportional to λ now as seen in Eq.(6). This makes the deformation rate much slower as the viscosity ratio increases while the droplet velocity is practically independent of λ as shown in figure 5(b). Thus, a high-viscosity droplet does not have enough time to deform as it flows through the cross-junction, and the droplet's transient deformation decreases with the viscosity ratio for very viscous droplets with $\lambda \geq 5$, as shown in figure 5(a). For the same reason, during the final relaxation stage towards the bullet shape far downstream from the junction, droplets with $\lambda = 10, 20$ need a significant time (or channel length) to reach equilibrium while droplets with smaller viscosity ratio relax faster, in agreement with Eq.(6) and our computational findings shown in figure 5. It is of interest to note that the steady-state droplet deformation far downstream of the junction increases with the viscosity ratio for highly viscous droplets owing to the increased inner hydrodynamic forces [18, 24, 40].

The effects of the viscosity ratio on the overall droplet deformation identified for capillary number $\text{Ca} = 0.1$ are also valid for higher flow rates, but now the increased hydrodynamic forces result in a dramatically different rear edge development for droplets of different viscosity ratios.

In particular, low-viscosity droplets develop a fully three-dimensional pointed tail at higher flow rates, as shown in figure 6 for $\lambda = 0.01$ and capillary number $\text{Ca} = 0.3$. This phenomenon is similar to the droplet's pointed rear shown in figure 2 for $\lambda = 0.2$ but now the low inner viscosity is accompanied with a more pointed rear tail which helps stabilizing the interfacial shape. The pointed tails observed here are similar to the pointed slender shapes low-viscosity droplets obtain in extensional flows so that they are able to withstand increased flow rates [4, 13, 34]. As the droplet exits the cross-junction, the rear tail is developed only along the front view while the droplet rear remains extended but rounded and flat along the top view, as seen in figure 6 for $x_c/\ell_z = 1.58, 2.07$. Only when the droplet is sufficiently downstream of the junction, its tail becomes axisymmetric-like, as shown in figure 6 for $x_c/\ell_z = 3.15$.

The transient development of the fully three-dimensional tail for low-viscosity droplets is shown quantitatively in figure 7(b) where we plot the evolution of the rear edge curvatures, $\mathcal{C}_{xz}^u$ and $\mathcal{C}_{xy}^u$, along the $y = 0$ and $z = 0$ planes, respectively. Observe that when the droplet is at the junction end (i.e. $x_c/\ell_z = 1$), its rear edge passes the junction middle and starts to obtain a pointed profile only along the front view. This edge profile becomes much more pointed when the droplet rear exits the junction, i.e. for droplet centroid $x_c/\ell_z = 2$, where still the droplet tail remains rounded along the top view. Only when the droplet tail has exited the junction, it begins to return to an axisymmetric-like shape, i.e. the rear curvature $\mathcal{C}_{xy}^u$ along the top view starts to increase but still remains smaller than the curvature $\mathcal{C}_{xz}^u$ along the front view. Far downstream of the junction (i.e. for $x_c/\ell_z \geq 3$), the tail becomes nearly axisymmetric, i.e. the two rear curvatures, $\mathcal{C}_{xz}^u$ and $\mathcal{C}_{xy}^u$, are nearly equal and the tail gradually disappears as the droplet obtains a bullet shape in the downstream square channel.

On the other hand, as the droplet passes through the cross-junction, its front edge remains axisymmetric-like, i.e. both front curvatures $\mathcal{C}_{xz}^d$ and $\mathcal{C}_{xy}^d$ start to increase similarly (even though $\mathcal{C}_{xz}^d > \mathcal{C}_{xy}^d$) when the droplet front is near the junction middle, while these curvatures also decrease similarly when the droplet front exits the junction, as shown in figure 7(c).

Increasing further the viscosity ratio to $\lambda = 1$, the circulation of the inner viscous fluid prohibits the development of a point rear edge when the droplet exits the junction, and thus the droplet obtains only an elongated profile along the front view. (See figures 3 and 4 of the supplementary material of this paper which present the deformation of equiviscous droplets.) Nevertheless, the droplet shape remains fully three-dimensional since the droplet profile is bullet-like along the top view. In direct contrast to our results for lower viscosity ratios, the downstream edge of equiviscous droplets remains more pointed than its upstream edge.

We emphasize that the overall droplet deformation, expressed in terms of the droplet lengths, is similar for

low and moderate viscosity ratios, as our results for $\lambda = 0.01, 0.2, 1$ and $\text{Ca} = 0.3$ indicate. However, the droplet's rear edge development is dramatically different. Along the front view, low-viscosity droplets are accompanied with the development of very pointed tails for interfacial stability, moderate viscosity droplets (e.g. $\lambda = 0.2$) develop elongated and pointed edges, while equiviscous droplets develop elongated and rounded edges. For both low and moderate viscosity ratios, the droplet remains laterally extended through the junction, obtaining a highly non-axisymmetric three-dimensional shape.

The deformation evolution of high-viscosity droplets is much simpler, though. The much slower deformation rate of these droplets, owing to the increased surface-tension time scale τ_γ , prohibits a significant transient deformation inside the junction. Thus, the entire passing of high-viscosity droplets through the micro-junction is axisymmetric-like, and these droplets smoothly transition to a more deformed bullet shape far downstream of the cross-junction, as our results for $\lambda = 10, 20$ reveal.

C. Effects of lateral flows

Having identified the effects of the capillary number Ca and the viscosity ratio λ on the droplet deformation, in this section we investigate the effects of different relative lateral flow rates Q_v/Q on the tail formation of low-viscosity droplets.

As shown in figures 8 and 9, by increasing the lateral flow and thus the hydrodynamic forces after the junction, the low-viscosity droplets develop a very pointed asymmetric tail which causes a significant elongation of the overall shape along the main flow direction, i.e. a significant increase of the droplet's length L_x . The sharp tail developed along the front view as the droplet exits the junction (i.e. during the deformation period) becomes axisymmetric-like during the restoration period quite downstream of the junction as the tail length is reduced, as seen in figures 8 and 9. Far downstream of the junction, the tail disappears and the droplet obtains a bullet-like shape.

The large tail curvatures, especially for the highest relative lateral flow rate studied, $Q_v/Q = 0.75$, seen in figure 8, reveals the transient appearance of small local length scales at the bubble tail, which makes the current investigation a multi-length interfacial dynamics problem. For $Q_v/Q = 0.75$ our implicit algorithm divides the spectral element at the droplet's tail into five smaller elements as the pointed tail is formed, to produce sufficient spatial discretization needed for the accurate determination of the interfacial shape. Element division is the adaptive mesh reconstruction technique of our spectral element algorithms so that they are able to produce a reasonable spectral element discretization, needed especially for local interfacial deformations such as tails and necks, as described in our earlier papers [8, 39].

4. CONCLUSIONS

In this paper we have investigated computationally the transient deformation of a droplet flowing along the centerline of a microfluidic cross-junction device. In particular, we have considered droplets with constant surface tension which are naturally buoyant in the surrounding fluid, and have size smaller than the cross-section of the square channels comprising the cross-junction. Our investigation involves low-to-strong flow rates for both the main and the lateral flows, and a wide range of viscosity ratio. We emphasize that in this work we have considered flow rates which do not result in interfacial breaking. This study is motivated by a wide range of industrial and biomedical applications including enhanced oil recovery, lubrication processes, design of fuel cells, drug delivery, micro-particle fabrication as well as lab-on-a-chip devices for clinical and environmental monitoring and analysis [2, 24, 35, 38].

Our investigation shows that the intersecting flows at the cross-junction act like a constriction, created by the streamlines of the intersecting flows, and thus the droplet needs to be compressed inside and outside of this constriction. Thus, the droplet shows a rich deformation behavior as it passes through the micro-junction. After obtaining a bullet-like shape in the square channel before the cross-junction, the droplet becomes slender inside the junction (to accommodate the intersecting flows), then it obtains an inverse-bullet shape as it exits the cross-junction which reverts to a more deformed bullet-like shape far downstream of the cross-junction (owing to the combined flow rates of the intersecting channels).

Increasing the viscosity ratio from small values up to $O(1)$, the droplet deformation increases monotonically owing to the higher inner hydrodynamic forces. However, the deformation of a highly viscous droplet decreases significantly with the viscosity ratio because the droplet does not have enough time to accommodate the much slower deformation rate as it moves inside the microfluidic device. In addition, the viscosity ratio has different effects on the droplet's rear edge development. Along the channel view, low-viscosity droplets develop very pointed tails for interfacial stability owing to the local extensional-type flow while higher viscosity droplets develop elongated and rounded edges. In both cases, the droplet remains laterally extended in the micro-junction, obtaining a fully three-dimensional shape that cannot be described from single-view observations in microfluidic experiments or based on axisymmetric or two-dimensional computations.

Therefore, our work highlights the three-dimensional effects of the asymmetric microfluidic geometry on the droplet deformation and the dramatically different effects of the fluids viscosity ratio on the droplet's overall length scales and the local length scales at the droplet edges. In addition, the large edge curvatures and thus the small local length scales developed transiently, especially at the tail of low-viscosity droplets, reveal that the current in-

vestigation is a multi-length interfacial dynamics problem.

The asymmetric deformation of the interfacial shape for droplets of any viscosity ratio and the tail formation for low-viscosity droplets suggest that fabrication of microparticles or microcapsules with various non-spherical shapes can be achieved. For this, one needs to utilize a high enough concentration of a cross-linking agent in the intersecting channels to achieve a fast cross-linking just after the cross-junction where the asymmetric droplet deformation occurs.

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Appendix A: Supplementary material

Supplementary data associated with this article can be found, in the online version.

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FIG. 1. Illustration of a neutral droplet flowing at the centerline of a microfluidic cross-junction.

FIG. 2. The shape of a droplet (plotted row-wise) with $a/\ell_z = 0.7$, $\lambda = 0.2$, $\text{Ca} = 0.4$, and $Q_v/Q = 0.5$, as seen from (a) the negative y -axis (i.e. front view) and (b) the positive z -axis (i.e. top view), at centroids $x_c/\ell_z = -1.50, -0.07, 0.63, 2.45, 3.55, 7$, (c) Droplet profile (i.e. droplet intersection with the plane $y = 0$) shown inside the microfluidic device at $x_c/\ell_z = -1.50, -0.07, 0.63, 2.45, 3.55, 5.73$.

FIG. 3. Droplet lengths as a function of the centroid x_c , for a droplet with $a/\ell_z = 0.7$, $\lambda = 0.2$, $Q_v/Q = 0.5$, and capillary number $\text{Ca} = 0.01, 0.1, 0.2, 0.3, 0.4, 0.5$. (a) Length L_x , (b) height L_z and (c) width L_y (scaled with the length $2a$ of the undisturbed spherical shape).

FIG. 4. Droplet velocity U_x (scaled with the average undisturbed velocity U of the upstream horizontal channel) as a function of the centroid x_c , for a droplet with $a/\ell_z = 0.7$, $\lambda = 0.2$, $Q_v/Q = 0.5$, and capillary number $\text{Ca} = 0.01, 0.1, 0.2, 0.3, 0.4, 0.5$.

FIG. 5. Evolution of droplet properties as a function of the centroid x_c , for a droplet with $a/\ell_z = 0.7$, $\text{Ca} = 0.1$, $Q_v/Q = 0.5$, and viscosity ratio $\lambda = 0.01, 0.1, 0.5, 1, 2, 10, 20$. (a) Droplet length L_x (scaled with the length $2a$ of the undisturbed spherical shape). The curve for $\lambda = 2$ is near the $\lambda = 1$ curve and has been omitted for clarity. (b) Droplet velocity U_x (scaled with the average undisturbed velocity $\mathcal{U}$ of the upstream horizontal channel). Our results for $\lambda = 0.01$ are identical to those for $\lambda = 0.001, 0$.

FIG. 6. The shape of a droplet (plotted row-wise) with $a/\ell_z = 0.7$, $\text{Ca} = 0.3$, $Q_v/Q = 0.5$, and viscosity ratio $\lambda = 0.01$ as seen from (a) the negative y -axis (i.e. front view) and (b) the positive z -axis (i.e. top view), at centroids $x_c/\ell_z = -0.03, 1.58, 2.07, 2.70, 3.15, 4.27$. (c) Droplet profile (i.e. droplet intersection with the plane $y = 0$) shown inside the microfluidic device at the same centroids x_c/ℓ_z as in (a, b).

FIG. 7. Evolution of droplet properties as a function of the centroid x_c , for a droplet with $a/\ell_z = 0.7$, $\text{Ca} = 0.3$, $Q_v/Q = 0.5$, and viscosity ratio $\lambda = 0.01$. (a) Droplet lengths, L_x , L_z and L_y (scaled with the length $2a$ of the undisturbed spherical shape). (b) Curvatures C_{xz}^u and C_{xy}^u at the droplet's upstream edge. (c) Curvatures C_{xz}^d and C_{xy}^d at the droplet's downstream edge. The curvatures are scaled with the curvature of the undisturbed spherical shape.

FIG. 8. Evolution of droplet properties as a function of the centroid x_c , for a droplet with $a/\ell_z = 0.7$, $\lambda = 0.01$ and $\text{Ca} = 0.3$. (a) Droplet length L_x for $Q_v/Q = 0.10, 0.25, 0.50, 0.75$. (b) Curvatures C_{xz}^u and C_{xy}^u at the upstream edge and (c) curvatures C_{xz}^d and C_{xy}^d at the downstream edge, for $Q_v/Q = 0.75$. The curvatures are scaled with the curvature of the undisturbed spherical shape.

FIG. 9. The shape of a droplet with $a/\ell_z = 0.7$, $\lambda = 0.01$, $\text{Ca} = 0.3$ and $Q_v/Q = 0.75$ at $x_c/\ell_z = 2.25, 3.16$, as seen from (a) the negative y -axis (i.e. front view) and (b) the positive z -axis (i.e. top view).

Graphical Abstract

We investigate computationally the transient deformation of a droplet flowing through a microfluidic cross-junction. Our work highlights the three-dimensional effects of the asymmetric microfluidic geometry on the droplet deformation, and the multi-length nature of the tail formation for low-viscosity droplets.