Discontinuous Transition in a Laminar Fluid Flow: A Change of Flow Topology inside a Droplet Moving in a Micron-Size Channel

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Even at moderate values of Reynolds number [e.g., Re = O(1)] a curved interface between liquids can induce an abrupt transition between topologically different configurations of laminar flow. Here we show for the first time direct evidence of a sharp transition in the speed of flow of a droplet upon a small increase of the value of the capillary number above a threshold and the associated change of topology of flow. The quantitative results on the dependence of the threshold capillary number on the contrast of viscosities and on the direction of transition cannot be explained by any of the existing theories and call for a new description.

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The applied interest in microfluidics is, to some extent, orthogonal to the interest of physicists. Low or moderate values of the Reynolds number generate nontrivial laminar flows and the useful smooth one-to-one correspondence between the input parameters and the flow on chip [1]. Conversely, nonlinear dynamics in, e.g., the flow of droplets through networks of microchannels can generate grasping effects of coding or decoding [2] or information, logic operations [3], or dynamic “memory” [4]. These effects are potentially useful in applications including (i) characterization of kinetics of reactions in droplets; (ii) synchronization, merging, and incubation of droplets; or (iii) amplification of small physicochemical changes of the content of droplets into detectable change in distribution of flow. Progress in this area demands the characterization and fundamental understanding of the flow of drops through microchannels.

Here we study an apparently simple problem—the speed of a droplet in a square cross-section (A) capillary filled with a wetting liquid (viscosity \( \mu_w \)). The immiscible droplet (\( \mu_d = \lambda \mu_w \)) does not wet the walls of the channel being separated from them by a thin film of the matrix liquid. Flow of this outer liquid at a volumetric flow rate \( Q_v \) (at a superficial speed \( u_s = Q_v/A \)) induces translation of the drop at speed \( u_d \). Questions are the following: what is the mobility \( \beta = u_d/u_s \) of the droplet, and what is the topology of flow inside the droplet?

In 1935 Fairbrother and Stubbs [5] observed that a bubble in a circular capillary translates faster than \( u_s \) because the wetting film is stationary with respect to the wall. Taylor [6] considered replacement of liquid from a capillary by gas and identified the fraction \( m \) of the cross section of the capillary occupied by the liquid film to be proportional to the square root of the capillary number (\( Ca = \mu_d u_b/\gamma \), with \( \gamma \) being the interfacial tension), corresponding to \( \beta = (1 - m)^{-1} \) for \( u_d = u_b \), Bretherton [7] studied a finite bubble in a circular capillary and found both the pressure drop along the bubble and \( m \) to be proportional to \( Ca^{2/3} \). Schwartz [8] confirmed these results for long bubbles and slightly corrected them for short ones.

Hodges [9] first modeled a viscous droplet in a circular capillary and proposed a topological transition of the flow in the droplet upon a change of \( \lambda \), which has not been observed before. For \( \lambda = O(Ca^{-1/3}) \) a single toroidal convection roll inside the droplet with two counterrotating rolls in the continuous liquid, at the front and rear of the droplet. For \( \lambda < Ca^{-1/3} \) these counterrotating vortices were predicted [9] to enter the drop creating pairs of stagnation lines at the fore and rear caps. Since the threshold value of the contrast of viscosities scales with the inverse cube root of the capillary number, for a fixed value of \( \lambda \) this translates into a prediction of three convection rolls inside the droplet below a threshold value of \( Ca \) and a single roll above the threshold, a transition observed recently in simulations of flow in circular capillaries [10].

Flow in circular capillaries differs qualitatively from the flow in rectangular channels that are typically used in microfluidic systems. Wong et al. [11,12] were first to treat analytically flow of bubbles in polygonal capillaries and showed that the continuous liquid can flow by the bubble, through the corners of the channel (gutters). Wong postulated domination of gutter flow for \( Ca < 10^{-6} \) (with pressure drop \( \propto Ca \)) and plug flow for \( Ca > 10^{-6} \) with \( \Delta p \propto Ca^{2/3} \). In contrast to the circular capillaries, the complexity of the nontrivial shape of the cross section of the droplet, and of the field of flow in the wetting film in the rectangular microchannels make it impossible to postulate a simple relation between the shape of the droplet and its mobility.

In spite of the contemporary importance of two-phase microfluidics [13,14] experimental reports on speed of drops in microchannels are few and cover narrow ranges of parameters. Fuerstman [15] monitored the motion of bubbles and found that the concentration of surfactant...
discriminates between different sources of dissipation: i.e., vortices at the caps of bubbles and flow in films at the walls of the channel. Vanapalli [16] measured $u_\star$ in a $(200 \times 120 \ \mu m^2)$ channel containing a droplet to measure its resistance and speed. They found $\beta = 1.28$ for drops and 1.04 for bubbles, independently of their length and of $Ca \in (10^{-3}, 10^{-2})$. Labrot [17] measured $\beta = 1.6$ for $Ca \in (10^{-3}, 4 \times 10^{-3})$, $\lambda \in (1/20, 70)$ in a $500 \times 300 \ \mu m^2$ channel. Sessoms [18] studied small drops and found $\beta > 1$ for drops smaller than the width of the capillary and $\beta < 1$ for larger ones.

Here we demonstrate the first systematic measurements of the speed of droplets translating in a square capillary over wide ranges of the Capillary number ($10^{-4}, 10^{-1}$) and the length $l$ of the droplets (between $l \sim 0.8 w$ and $16 w$, where $w$ is the width of the channel). We used a $T$-junction system fabricated in polycarbonate [19] draped with dodecylamine [20] to ensure wetting by hexadecane. A cross section of all channels was $A = w^2 = 360(\pm 7) \times 360(\pm 7) \ \mu m^2$ (see Supplemental Material, Fig. S1 [21]). We delivered the dispersed phase from a pressurized reservoir (pressure $p_{cr}$, Rexroth PR1-RGP) through an electromagnetic valve (EMV$_d$ in Fig. 1) and a capillary [22]. The droplet liquid comprised either distilled water ($\mu_d = 0.890 \ \text{mPa} \cdot \text{s}$, interfacial tension with hexadecane $\gamma = 46.1 \pm 1.6 \ \text{mN/m}$ and density $\rho_d = 997 \ \text{kg/m}^3$ at $25 \ ^\circ\text{C}$) or one of three different aqueous solutions of glycerine: [38.2% ($w/w$) of viscosity $\mu_d = 3.03 \pm 0.1 \ \text{mPa} \cdot \text{s}$, $\gamma = 27.1 \pm 0.7 \ \text{mN/m}$, and $\rho_d = 1092 \ \text{kg/m}^3$], [61.5% ($w/w$) $\mu_d = 10.1 \pm 0.3 \ \text{mPa} \cdot \text{s}$, $\gamma = 25.4 \pm 0.6 \ \text{mN/m}$, and $\rho_d = 1155 \ \text{kg/m}^3$], or [86.3% ($w/w$) $\mu_d = 99.6 \pm 0.8 \ \text{mPa} \cdot \text{s}$, $\gamma = 30.2 \pm 0.8 \ \text{mN/m}$, and $\rho_d = 1222 \ \text{kg/m}^3$]. Continuous phase (hexadecane, AlfaAesar) was also supplied from a pressurized container with the difference that $p_c$ was set by an electronic proportional regulator (Parker, MPT40) controlled by computer (CU) via LABVIEW. Pressures were monitored with digital manometers and the dependence $Q_c(p_c)$ was calibrated on a balance (WLC-C/2, Radwag). An insulating box enclosed the chip and a stereoscope (Nikon SMZ1500); CU recorded temperature on the chip, stabilized to $25(\pm 0.6) \ ^\circ\text{C}$. A linear camera (AViVA IIEM4CL2014) acquired images at 70 kHz and reported in real time to CU.

Liquids did not contain any surface active additives and we washed the microfluidic system thoroughly by passing hexadecane for several hours to ensure that no remains after modification could diffuse to the interface. This is important because the presence of surfactant at the interface and the dynamic surface tension effects can significantly alter the dynamics of flow and blur the already complicated problem.

Our automated setup allowed us to generate droplets of volume between $\sim 0.8 w^3$ and $16 w^3$, and to screen the speed of flow of the continuous phase $Q_c \in (1, 800 \ \text{ml/h})$ corresponding to the values of the Reynolds number $Re = Q_c \rho_c / w \mu_c \in (0.6, 535)$. The Bond number $Bo = (\rho_d - \rho_c) g A / \gamma = 0.006$ indicates that gravitational acceleration ($g$) does not influence our experiments. For each series of data for a given value of $Ca = Q_c \mu_c / A \gamma$, with $\mu_c = 3.032 \ \text{mPa} \cdot \text{s}$ and $\gamma = 53.3 \ \text{mN/m}$ [23] CU automatically performed a typical number of 10 experiments for each length of the droplet. In each experiment CU formed a single droplet to avoid any hydrodynamic interactions between subsequent droplets. After formation the droplet was advanced forward with given $Q_c$, and we monitored $u_d$ far downstream ($> 100 w$) of the $T$ junction to ensure stationary flow. At the moment of measurement $Q_c$, the total rate of flow in the channel and thus $\beta = u_d / u_\star$, is the same as $\beta = u_d / u_{total}$ with $u_{total} = (Q_c + Q_d) / A$ in more complicated flow protocols. We repeated each series at least once on a different day to ensure reproducibility. We measured the length of the droplets with accuracy better than 2%, and the speed with accuracy better than 0.5%, and checked reproducibility of the measurements on a separately prepared copy of the microfluidic system.

Figure 2 graphs $\beta = u_d / u_\star$ as a function of the length $l$ of the droplet for a range of values of $Ca$. Small droplets of $l \sim 1$ travel faster than $u_\star$. For longer drops we observe either a direct, asymptotic approach $\beta \to 1$, or decrease to $\beta_{\text{min}} < 1$ and then an asymptotic $\beta \to 1$ from below.

Our measurements uncover a discontinuity of $\beta$ in $Ca$. At the lowest value of $Ca = 1.5 \times 10^{-4}$ $\beta_{\text{min}} = 0.98$ at $l_{\text{min}} = 2.0$. As $Ca$ increases the minimum in $\beta$ becomes
discontinuity in the mobility allowing us to hypothesize given in the graphs. The contrast of viscosities is the minimum shallower (Fig. 2). Screens of mobility of reappears deeper and wider shifted towards longer drop-

The speed of droplets abruptly changes. The minimum is absent. With a further increase of \( \lambda > \lambda_{TR} \), the speed of droplets for \( l = 2w \) to \( 5w \), and then decreases to 0.6–0.65 for \( l = 16w \) (with the characteristic values of \( \beta \) and \( l \) increasing with increasing \( \lambda \)). For \( \lambda = 30 \), \( \beta \) is a linearly decreasing function of \( l \), without any dependence on \( \lambda \).

Lead by the notion that the abrupt crossover in speed that we observe for \( \lambda < 1 \) can only be caused by a change of topology of flow, we traced the flow using particles (\( \sim 1.0 \times 10^3 \) particles per ml, 5 \( \mu \)m YG-Microspheres, Polyscience, without surfactant). We performed particle image velocimetry [25–27] on high-speed (10^4 frames/s) videos (Photron1024) recorded at different focal depths (20, 80, and 180 \( \mu \)m from the ceiling of the channel). For \( \lambda < \lambda_{TR} \) we observed (Fig. 4 and Figs. S2-7 in the Supplemental Material [21]) convection rolls that spanned the whole length of the droplet. This flow field was observed previously [25] and—in difference to the case of circular capillary—instead of a single torus comprises a set of four rolls reflecting the fourfold rotational symmetry of the channel. For \( \lambda > \lambda_{TR} \) the original rolls shrunk to the middle section of the droplet, while at the caps new—counterrotating—rolls appeared (a qualitatively similar pattern has been observed previously in square capillaries [28]).

Interestingly, the transition that we observe undergoes in the opposite direction to that proposed [9] and observed numerically [10] for cylindrical capillaries. The criterion proposed by Hodges [9]: \( \lambda = O(\lambda_{TR}^{-1/3}) \) with three rolls in the droplets for small, and one roll for large value of \( \lambda \), predicts that the additional eddies should enter the droplet at \( \lambda < \lambda_{TR} \) smaller than a threshold value, while we observe this for \( \lambda > \lambda_{TR} \). Our observation thus contrasts also with the qualitative prediction [17] made for rectangular

\[ \frac{\beta_{\min}}{\beta_{\min}} = \lambda = \frac{\omega}{\omega_{\min}} \]

\[ \beta_{\min} \] exhibits a discontinuity at the transition for \( \lambda < 1 \). A diagram illustrating the transition from the simple to more complicated pattern of flow in droplets in square capillaries. (c) The relation between the threshold values \( \lambda \) and \( \lambda_{TR} \) as well as the orientation of the transition is different than in the case of circular capillaries [9].

FIG. 2. \( \beta \) plotted against \( l \) for \( \lambda < \lambda_{TR} \) (top) and for \( \lambda > \lambda_{TR} \) (bottom). The values of the capillary numbers are given in the graphs. The contrast of viscosities is \( \lambda = 0.33 \).

FIG. 3. (a) Crossover in the speed of the droplets upon an increase of the capillary number above the threshold: the value of \( \beta_{\min} \) exhibits a discontinuity at the transition for \( \lambda < 1 \). (b) A diagram illustrating the transition from the simple to more complicated pattern of flow in droplets in square capillaries. (c) The relation between the threshold values \( \lambda \) and \( \lambda_{TR} \) as well as the orientation of the transition is different than in the case of circular capillaries [9].

FIG. 4 (color online). Diagrams of the flow field in the droplet: (top) \( \lambda = 5 \times 10^{-4} < \lambda_{TR} \) and (bottom) \( \lambda = 2 \times 10^{-3} > \lambda_{TR} \). Left panels show the cross section through the center of the droplet with the dots marking the flow coinciding with the orientation of \( \mu \) and crosses the counterflow. Center and right panels show the velocity fields obtained at the middle of the channel (180 \( \mu \)m from the top wall) and the corresponding schematic representations. The bar codes the speed in mm/s in a frame moving with the droplet.
channels, that expected that the counterrotating rolls will transfer from the droplet to the continuous phase upon an increase of \( \lambda \), just as proposed by Hodges [9] for circular capillaries. Also the value of the critical capillary number is very different for circular capillaries [9] \( \text{Ca}_{\text{TR}} \sim O(27) \) as compared to \( \text{Ca}_{\text{TR}} \sim 10^{-3} \) in our experiments.

The abruptness of the change of the mobility of droplets upon transition has neither been observed nor predicted to date. Lozar [29] predicted numerically a transition of the flow field in front of a semi-infinite bubble upon an increase of \( \text{Ca} \) above \( \text{Ca}_{\text{TR}} \in (0,1) \), depending on the aspect ratio of a rectangular channel. Yet, he did not associate the transition with any discontinuity in the wetting fraction \( m \) (and, consequently, in the speed \( \dot{m} \)).

Radke expected \( \text{Ca}_{\text{TR}} \) to increase monotonically with \( \text{Ca} \), while Lozar [29] hypothesized a continuous transition between topologically distinct fields of flow in the droplet upon a change of the viscosity contrast. For similar reasons our findings cannot be matched with the theory [6,7] for the deposition of liquid at the walls of circular capillaries, as they expect \( m \) and, consequently, \( \dot{m} \) to increase monotonically with \( \text{Ca} \). The sharp crossover that we observe is also difficult to be associated with the plug- to corner-flow transition [11,12]: in the plug mode (\( \text{Ca} < 10^{-6} \)) Radke expected \( \beta > 1 \) while above the transition the value of \( \beta \) can be either larger or smaller than unity, as the flow through the corners is not a priori restricted to any orientation. In our experiments, we observe that—at the transition—the flow in the gutters is always oriented forward (Fig. 4).

In summary, we observed a sharp topological transition of the field of flow inside droplets translating in a microchannel of a square cross section. These results cannot be explained by existing models and call for a new theory that will be applicable to rectangular channels used in microfluidic chips. Development of such understanding is important for the design of lab-on-chip systems. For example, the precision of microfluidic analyses [31] of kinetics of reactions depends on the precision of prediction of the speed of droplets. In automated systems [22] the mobility of droplets conditions corrects synchronization. Precise characterization of finite changes of the mobility of drops may allow for the construction of systems that amplify a small change of properties of the liquid inside the drop (e.g., its viscosity) to a “macroscopic” flow pattern [3,4].

Our automated system allows for issuing individual drops of prescribed volume for measurements noninterfered by other droplets in the channel. The use of pressurized containers and resistive capillaries [32] provides for a stable rate of flow and the rapid screening of the value of \( \text{Ca} \). We hope that the automated microfluidic systems that can rapidly screen experimental parameters and the data they generate will boost the theoretical description of the problem and development of guidelines for the design of microfluidic systems.