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Shrinking instability of toroidal droplets

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Toroidal droplets are inherently unstable due to surface tension. They can break up, similar to cylindrical jets, but also exhibit a shrinking instability, which is inherent to the toroidal shape. We investigate the evolution of shrinking toroidal droplets using particle image velocimetry. We obtain the flow field inside the droplets and show that as the torus evolves, its cross-section significantly deviates from circular. We then use the experimentally obtained velocities at the torus interface to theoretically reconstruct the internal flow field. Our calculation correctly describes the experimental results and elucidates the role of those modes that, among the many possible ones, are required to capture all of the relevant experimental features.

Experimental Preliminaries

To determine the flow field, we use particle image velocimetry (PIV). We introduce optical heterogeneities in the liquid by adding tracer particles; these are polystyrene beads with an average diameter of 16.2 μm and a density of 1.06 g/mL. The method relies on video recording the time evolution of the liquid and spatially correlating the optical heterogeneities between consecutive frames, which allows assigning instantaneous velocities to different regions within an image. We use existent software (PIVLab (11)) and confirm the accuracy of the experimental setup and computer programs by video recording the motion of water with tracer particles rotating at a constant angular speed. We image along the rotation axis and confirm that the measured angular speed agrees with that imposed in the experiment.

To further test the experimental setup and data analysis, we perform experiments with sinking spherical droplets. In this case, the expected flow field is well known for any inner and outer liquid viscosities (13–15). We note that we choose the radius of these droplets so that they match the typical tube radius of our toroidal droplets. Because the liquid–liquid interface in our experiments is curved, light refraction could be significant, depending on the optical contrast between the liquids. Hence, these experiments also allow testing whether these effects play a dominant role in our experiments with toroidal droplets. The outer liquid is 1,000 centistokes (cSt) silicone oil, whereas the spherical droplets are made of deionized water with tracer particles. Because the respective densities are $\rho_\text{o} \approx 0.97$ g/mL and $\rho_\text{t} \approx 0.99$ g/mL, the droplets sink. We visualize the flow by illuminating with a laser sheet through the center of the droplet, as shown in Fig. L4, and find that the flow has the typical

Significance

Liquid doughnuts or tori exhibit an inherent instability consisting in the shrinking of their “hole.” For sufficiently thick tori, this behavior is the only route for the torus to become spherical and thus minimize the area for a given volume. Of note, this shrinking instability is not completely understood. In this work, we experimentally determine the flow field of toroidal droplets as they shrink and account for our observations by solving the relevant equations of motion in toroidal coordinates. We find that four modes, out of the many possible ones, are needed to successfully account for all of the relevant features in the experiment. These results highlight the rich fluid mechanics that result in the presence of interfaces with nonconstant mean curvature.

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2The genus of a compact boundaryless surface is equal to the number of handles. A sphere has no handles, whereas a torus or a cup of coffee has one handle. The handle of the torus is then a global, topological property of the surface.
circulation profile in the reference frame of the drop, expected for these situations, as shown in Fig. 1B. We note that the color code corresponds to the measured speeds, normalized with the sinking speed, $v_{sink}$, for these situations, as shown in Fig. 1B. We note that the color scale corresponds to the measured speed divided by the shrinking speed, $v_{sh}$, which we obtain by averaging the $x$-component of the velocity through the whole cross-section of the torus. The lack of up–down symmetry in the velocity field indicates that the observed flow field is not purely shrinking but that it also reflects the sinking of the toroidal droplet due to the density mismatch of the inner and outer liquids.

To isolate the flow associated with shrinking, we recall that the Reynolds number in our experiments is $Re \sim O(10^{-6})$, implying that we are well within the Stokes regime, where

$$\mu \Delta u = V p,$$

with velocity $u$ and pressure $p$. Because the governing equations in our problem are linear, we can use the superposition principle and think of the flow in Fig. 3C as the sum of the flow associated with sinking and the flow associated with shrinking. Interestingly, these flows have differentiating up–down symmetries. Consider sinking first. In this case, $v_i$ is antisymmetric under a $z \to -z$ operation $[v_i(z) = -v_i(-z)]$, whereas $v_\theta$ is symmetric under a $z \to -z$ operation $[v_\theta(z) = v_\theta(-z)]$, as clearly seen in the schematic in Fig. 4A, where, for the sake of simplicity, we illustrate the situation for a thin toroidal droplet; the up–down symmetry reflected here, however, is general and maintained irrespective of the aspect ratio of the torus. For a shrinking droplet, however, $v_i(z) = v_i(-z)$ and $v_\theta(z) = -v_\theta(-z)$, as clearly seen in the schematic in Fig. 4B, where again we illustrate the situation for a thin toroidal droplet. We then obtain the symmetric and antisymmetric components of each of the velocity components, $v_i = [v_i(z) + v_i(-z)]/2$ and $v_\theta = [v_i(z) - v_i(-z)]/2$, with $i = x, z$, and take the symmetric part of $v_i$ and the antisymmetric part of $v_\theta$ to obtain the flow field associated with shrinking. The result is shown in Fig. 3D. Note that the flow now has the expected up–down symmetry. In addition, we see that the velocity is higher near the inside of the torus, reflective of the shrinking process. By subtracting $v_{sh}$, we obtain the flow field in the drop frame, as shown in Fig. 3E. It is now evident that the flow field is not radial at the interface but, instead, that there are both radial and tangential components of the velocity, which, in turn, causes a circulation that results from the viscous shear stresses exerted at the interface. In this frame of reference, we also see that there is a source near the outer part of the torus, which results from volume conservation and implies that as the torus shrinks, the tube radius must increase.

Shrinking Toroidal Droplets

We generate toroidal droplets with controllable central-circle radius, $R_0$, and tube radius, $a_0$, as defined in Fig. 2, by injecting an inner liquid through a tip into a rotating continuous phase (5). The inner liquid is a 0.1 wt% solution of PEG in water containing tracer particles at a volume fraction of 0.001. The continuous phase is a 60,000 cSt silicone oil that rotates at a constant angular speed, $\omega$, as schematically shown in Fig. 3A. For sufficiently high $\omega$, the shear exerted by the outside liquid onto the liquid exiting the tip induces formation of a jet, which follows the imposed rotation and closes up into a torus (5). The presence of PEG lowers the interfacial tension from $\gamma = (40 \pm 2)$ mN/m to $\gamma = (26 \pm 2)$ mN/m, hence slowing down the subsequent dynamics to allow enough time for any flow field due to the generation process to dissipate. As for a sinking spherical droplet, the flow inside a shrinking torus has azimuthal symmetry. It is thus enough to image the flow in a 2D cross-section through the center of the torus, which we do by using a laser sheet, as also shown in Fig. 3A.

A typical snapshot of a toroidal droplet during the shrinking process is shown in Fig. 3B (also see Movie S3); this droplet has an aspect ratio of $R_0/a_0 \approx 1.2$, which is below the threshold where Rayleigh–Plateau modes can cause breakup. Hence, these droplets can only evolve via shrinking. From the image, we realize that the cross-section of the torus does not remain circular. Instead, we observe that the initial circular cross-section elongates vertically and is slightly flatter near the inside of the torus.
To test the flow decomposition used to isolate the shrinking component, we perform computer simulations. We use the level set method for two phase flows in COMSOL Multiphysics (16) and simulate the experimental situation in the absence of gravity. We find that the overall drop deformation and flow field agrees well with our experimental findings, as shown in Fig. 3F. Also note that the flow field obtained in the simulations exhibits the up–down symmetries expected for shrinking. This result confirms the procedure to isolate the shrinking flow from the experimental flow field.

We then theoretically address the shrinking problem to understand the origin of the various features associated with the shrinking flow. We use the solution for the stream function, \( \Psi \), in the Stokes regime in the set of toroidal coordinates \((\eta, \chi, \phi)\) (9, 17). This coordinate system makes use of a focal circle with radius \( c = \sqrt{R_0^2 - \eta_0^2} \lt R_0 \). The coordinate \( \phi \) is just the azimuthal angle. The coordinate \( \chi \) is the angle defined with respect to the focal circle, as shown in Fig. 2; it can take values \( \chi \in (-\pi, \pi) \). Finally, \( \eta \), which is related to the aspect ratio of the torus, defines a given toroidal surface. Specifically, we take \( \eta_0 \) to be the value corresponding to our interface. As a result, \( \cosh (\eta) = R_0 / \eta_0 \). The inner part of the torus then corresponds to \( \eta > \eta_0 \), as shown in Fig. 2. In this coordinate system (9):

\[
\Psi = \frac{c \sinh (\eta)}{(\cosh (\eta) - \cos (\chi))^{3/2}} \sum_{n=-\infty}^{\infty} C_n \sin(n\chi) Q_n^{1/2} (\cosh (\eta)),
\]

where \( Q_n^m \) is the associated Legendre polynomial of the second kind, and \( C_n \) are the amplitudes of each mode, which need to be determined. We make this determination by considering the experimental velocity at the interface and obtaining the radial, \( v_r \), and tangential, \( v_\theta \), components as a function of the polar angle \( \theta \) (defined in Fig. 3E). We find that \( v_\theta \) is not constant but that, instead, it peaks at \( \theta \approx 50^\circ \) and \( \theta \approx -50^\circ \), as shown by the blue line in Fig. 5A and consistent with the polar angle in the inside region of the torus below which the cross-section of the toroidal droplet is most deformed from the circular shape (Fig. 3B). Similarly, \( v_r \) also has extrema at the same angle, as shown by the blue line in Fig. 5B.

Fig. 3. (A) Schematic of the experimental setup for the generation of toroidal droplets together with the PIV setup (not drawn to scale). The cuvette is a parallelepiped with a square base of side 6 cm; the flat walls enable illuminating and imaging without refraction. Typical injection flow rates are \( \approx 50-60 \text{ mL/h} \). We note that as soon as the torus is formed, we remove the needle and stop the rotation. (B) Typical snapshot of a shrinking toroidal droplet with \( R_0 / \eta_0 \approx 1.2 \). (C) Flow field obtained using PIV. (D and E) Flow field associated with shrinking in the laboratory frame (D) and in the drop frame (E). (F) Computer simulation of shrinking toroidal droplets in the drop frame. The color scales in C–F correspond to the measured speed normalized with \( v_{sh} \).
From the stream function, we can calculate the velocity (18),

\[ v_\eta = \frac{(cosh(\eta) - cos(\chi))^2}{c sinh(\eta)} \frac{\partial \Psi}{\partial \chi}, \]

\[ v_\chi = -\frac{(cosh(\eta) - cos(\chi))^2}{c sinh(\eta)} \frac{\partial \Psi}{\partial \eta}, \]

and use it to fit the experimental velocities at the interface to obtain the constants \( C_n \). We find that by only considering the \( n = -2, -1, 1, 2 \) terms, we are able to capture all experimental features. Note that the \( n = 0 \) mode does not contribute because \( \Psi = 0 \) for \( n = 0 \). We emphasize that we perform a single simultaneous fit for both \( v_\eta \) and \( v_\chi \) while constraining the value of the constants \( C_n \) by forcing the resultant velocity field to have the same shrinking speed as in the experiment. The result, shown in Fig. 5A and B with dashed lines, correctly reproduces the experimental boundary velocities.

We obtain \( C_{-2} = (0.2 \pm 0.1)v_{sh}, \) \( C_{-1} = (0.50 \pm 0.06)v_{sh}, \)
\( C_1 = (-1.33 \pm 0.06)v_{sh}, \) and \( C_2 = (-0.4 \pm 0.1)v_{sh}. \) Using these constants, we obtain the flow field inside the toroidal droplet. The result, shown in Fig. 5C in the drop frame, captures all relevant features seen in the experiments (Fig. 3E), including the circulation due to viscous stresses, the higher velocities in the inside region of the torus compared with those in the outside of the torus, and the presence of a source in the flow field near the outer part of the torus. We now consider each mode separately;
these modes are shown in Fig. 6 in the drop frame. The \( n = 1 \) mode is radial at the interface, as shown in Fig. 6A, and has the largest weight of all four modes. This was the only mode considered in ref. 9 and is responsible for the increase in the tube radius as the torus shrinks. Note that in the presence of this mode only, the cross-section of the torus would remain approximately constant throughout the shrinking process. The \( n = -1 \) mode is tangential at the interface, as shown in Fig. 6B, and is responsible for the circulation seen experimentally. Finally, the \( n = \pm 2 \) modes can account for the shape we observe. The \( n = 2 \) mode produces a flow field that slows down the interface in the inside region of the torus, at \( \theta \approx 0^\circ \), as shown in Fig. 6C, hence promoting the experimental flattening in this region. The \( n = -2 \) mode, shown in Fig. 6D, is very similar to the \( n = 2 \) mode, but with inverted velocities. Note the difference in the absolute values in the velocities, however, reflecting that the \( n = 2 \) mode dominates over the \( n = -2 \). Note also that the flow fields for each mode are all consistent with the expected up–down symmetries.

Conclusions

We have experimentally obtained the flow field inside shrinking toroidal droplets using PIV and the linearity and symmetry properties of the flow in the problem. We observe that the cross-section of the torus does not remain circular over time but, rather, that it flattens in the inside region of the torus. Furthermore, the velocity at the interface has both radial and tangential components. We account for our observations by theoretically solving the Stokes equations using the stream function in toroidal coordinates. Four modes are enough to account for all experimental features. Of all modes, the \( n = 1 \) mode is the most significant and accounts for the flow field due to the expansion of the tube of the torus. The additional modes are needed to account for the observed circulation and deformation of the drop during shrinking. Overall, our results illustrate the behavior brought about by having interfaces with nonconstant mean curvature.

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