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Three-lobe-shaped equilibrium states in magnetic liquid bridges

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Stable 3-lobed cross sectional equilibrium shapes, invariant under rotations of 120°, are reported in magnetic liquid drops confined between two horizontal plates in the presence of a vertical dc magnetic field. Their connectivity with other 4-, 3- or 2-lobed shapes as magnetic field strength varies is studied experimentally. Although 3-lobed shapes are found to evolve into bent 2-lobed ones by slowly decreasing the field strength, the reverse transition is not observed when the field strength increases. Moreover, 2-lobed shapes suffer a bending transition, by increasing the field strength, which is found to be hysteretic.

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The cross section of a drop of magnetic liquid (commonly called ferrofluid), which bridges the gap between two horizontal solid plates, evolves into a plethora of static patterns in a dc magnetic field applied normal to the plates [1–3]. Each cross section of the liquid bridge remains circular as long as the field strength does not exceed a critical value. Beyond this threshold, the circular symmetry breaks and the bridge equilibrates to a 2-lobed (dumbbell) shape in its cross section. In stronger fields it is observed that the dumbbell elongates or bends forming a horseshoe shape, or that the initially circular cross section becomes multibranched and eventually a labyrinth [3,4]. In this Rapid Communication, the existence of 3-lobed equilibrium cross sectional shapes in magnetic liquid bridges is investigated experimentally. Special attention is given to 3-lobed shapes that are invariant under rotations of 120°; these shapes, the lobes of which are of equal size, are hereafter referred to as symmetric 3-lobed, in contrast to other 3-lobed shapes that lack the invariance under 120° rotations. Their stability is examined by means of studying their transitions to other 3- or 2-lobed shape families as the applied magnetic field strength varies.

The stability of 3-lobed configurations has recently attracted the interest of experimental studies in rigidly rotating drops. Ohsaka and Trinh [5] report a novel technique for cutting down 2-lobed shape perturbations in an acoustically levitated and rigidly rotating drop in the Earth's environment. By applying an acoustically driven oscillation, they depress the 2-lobed perturbations and they drive the axisymmetric drop to the angular velocity where the drops become unstable to 3-lobed perturbations. This way, they obtain 3-lobed shapes, which, however, evolve to 2-lobed ones when the forced oscillation is turned off.

Force competition determines the static equilibrium of a ferrofluid bridge. The forces acting on the liquid, namely magnetic force, gravity, and capillary force (the resultant of surface tension acting in a curved interface), are conservative. Thus, the equilibrium is governed by the minimization of an appropriate potential energy functional. In the absence of magnetic field and gravity, the bridge shape is determined by capillary force that tends to minimize the free surface in the face of the incompressibility of the liquid and of the wetting properties of the liquid/solid system, as reflected in the apparent contact angle at the liquid/plates/air contact line. This results in the formation of a neck in a circular—in each cross section—meniscus, when the liquid wets the solid plates, as is the case here. The neck is located halfway between the plates, and is shifted up by gravity, which also broadens the bridge at the lower plate. In the presence of a vertical magnetic field, the ferrofluid, which is a colloidal dispersion of ferrite particles in a liquid carrier, polarizes and tends to align in the direction perpendicular to the plates [6]. Raising the field strength strengthens the alignment of the magnetic dipoles of the ferrite particles in the vertical direction; thus, the meniscus attains an almost cylindrical shape except in the vicinity of the plates, due to their wetting by the liquid. The parallel oriented dipoles repel each other and the corresponding energy decreases as the mean distance between them increases, i.e., as they approach the free surface. Hence, magnetic force tends to increase the free surface area. When any of the ever-present infinitesimal noncircular disturbances causes a decrease of the dipole energy that surpasses the corresponding increase of the free surface energy, instability occurs, which leads to noncircular shapes. Briefly, the cross sectional instabilities are caused by a long-range repulsive interaction (dipole-dipole interaction) competing against a short-range attractive interaction, due to surface tension. For a cylindrical and uniformly magnetized liquid column, perturbation analysis gives the critical magnetization at the onset of the circular symmetry breaking instability [4,6].

The equilibrium deformation of the initially circular bridge strongly depends on its mean diameter as well as on the rate of change of the field strength [2,7]. By slowly increasing the field strength, small drops are driven in 2-lobed or bent dumbbell shapes, whereas large drops become multibranched when the field increases rapidly [7]. Langer *et al.* [8] and Jackson *et al.* [9] studied the shape evolution of a uniformly magnetized liquid bridge as a dissipative dynamical process. At fixed applied field strength, the initially circular cross section is perturbed by random noise and its shape evolution is computed numerically. Many complicated patterns are captured, exhibiting qualitative agreement with experiments, but important realities, such as the wetting ef-

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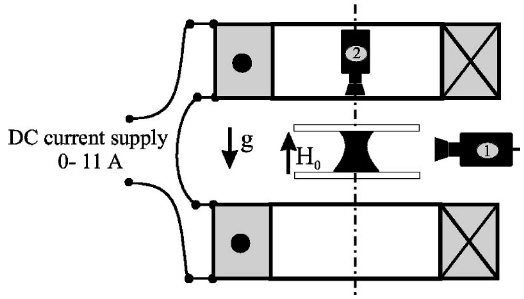

FIG. 1. Sketch of the main parts of the experimental apparatus.

fects and the nonuniform demagnetizing (or fringing) field that is created by the ferrofluid, are ignored. Performing realistic calculations of the magnetohydrostatic equilibrium paths as well as experiments, mechanisms of prelabyrinthine cross sectional instabilities are illuminated by the present authors [10]. Accounting for the effects of the wetting, of the fringing field, and of the nonlinear magnetization of the liquid, as well as of the gravity, the three-dimensional, nonlinear and free boundary problem that arises is solved by the Galerkin finite element method. Results show that circular cross sections become 2-lobed at a critical field strength, which corresponds to a pitchfork bifurcation point in the solution space. The 2-lobed solution family bifurcates subcritically from the circular solution family, which turns unstable to 2-lobed perturbations. The 2-lobed solutions are unstable near the bifurcation point but they regain stability past a turning point. The solution connectivity, as briefly outlined above, suggests a hysteretic transition from circular to 2-lobed shapes, a mechanism that is also confirmed experimentally [11]. Symmetric 3-lobed solutions are also found to bifurcate subcritically from the circular solutions at higher field strength than the 2-lobed ones. Their connectivity with nonsymmetric 3-lobed or 2-lobed shapes is under investigation. The aim of the experimental study that follows is to determine whether symmetric 3-lobed cross sections are stable and, moreover, to examine the relative stability of 3- and 2-lobed shapes by observing equilibrium shape transitions as magnetic field strength varies quasistatically. Simple techniques are also reported for the shape control of the cross section, partially inferred from the solution space diagrams available from the existing theoretical studies [10,11].

The experimental setup is sketched in Fig. 1. A pair of Helmholtz coils placed 4 cm apart and supplied by a controlled dc current is used for the production of the magnetic field. The inner and outer diameters of each coil are 5.5 cm and 13 cm, respectively. The ferrofluid bridge is placed halfway between the coils, centered at their common axis. Its size does not exceed the boundaries of a cylindrical region, 2 mm in height and 1 cm in diameter, where the field is vertical and almost uniform, with a variation less than 0.5%. The top and the side view of the bridge are recorded by two CCD microcameras suitably mounted to the coils' arrangement (see Fig. 1). The ferrofluid used in the experiments is based on light mineral oil and was used as received by Ferrofluidics Corporation with the following properties: magnetization at saturation $M_s = 400$ G, surface tension $\sigma = 28$ dyn/cm, initial susceptibility $\chi_i = 2.2$, density $\rho = 1.19$ g/ml, viscosity 9 cp at 27°C. Two Plexiglas plates 1.6 ± 0.05 mm apart are used to trap the sample drops that vary in size between 10 and 30 ± 0.25 mm^3. The drop shape is measured from the digitized close view photographs captured by the cameras and the volume is estimated numerically by its vertical profile shape. The apparent contact angle is measured to be $20° \pm 3°$ in any drop deformation. The field strength is measured by a Phywe teslameter equipped with an axial Hall probe. Length and field strength measurements are reported in normalized form. The radius of a sphere of volume equal to that of the drop is chosen as a characteristic length; as characteristic field strength, H_c is chosen that at the onset of the circular symmetry breaking instability [11].

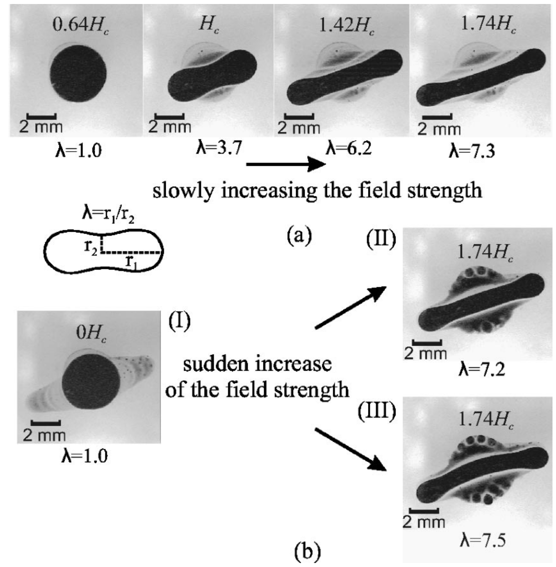

FIG. 2. Top-view photographs of equilibrium states of the captive drop in magnetic field. (a) Slow increase of the field strength. (b) The same drop with a sudden increase of the field strength. In all the photographs, except the first one in (a), a thin visible ferrofluid film is left on the plates as the drop contact line recedes.

Experiments show that the 2-lobed shape cross section is the most preferred nonaxisymmetric deformation, especially when the drop size is small. In Fig. 2 the field strength increases in a stepwise manner and in each step the drop of volume 10.1 ± 0.25 mm^3 is left to reach an equilibrium shape (no more than 5 s are required). The drop undergoes a well known shape evolution. The circular symmetry breaks at a critical strength H_c ($H_c = 235$ G), with the formation of a 2-lobed shape, which keeps elongating as the field strength is increased up to $1.74H_c$. To investigate whether the final equilibrium state is independent of the rate of variation of the field strength, the strength is decreased in a stepwise manner down to zero and then is suddenly increased up to $1.74H_c$. This procedure is repeated once more and, as photographs (II) and (III) in Fig. 2(b) show, the resulting states do not notably differ from the one that arises by slowly increasing the field strength [cf. the corresponding photograph in Fig. 2(a)]. This is also confirmed by comparing the aspect ratios, λ, of the cross sections, shown in Fig. 2. The latter observations imply that the noncircular equilibrium states lie on the 2-lobed shape family, which is followed even if the field is

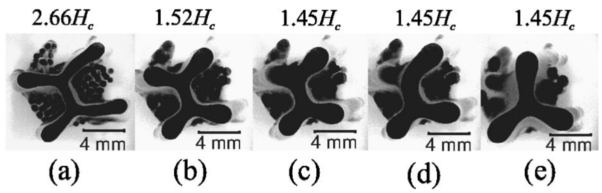

FIG. 3. The transition from 4- to 3-lobed cross sectional shapes.

raised suddenly. What is required for obtaining higher harmonic shapes, namely 3- or 4-lobed ones, is to pass over the 2-lobed shape family. This is efficiently attained, as experiment showed, when larger drop samples are used and especially when the field jumps at high strengths. In Fig. 3(a), a stable multibranched cross section with two nodes and four lobes evolves almost instantly from a circular one by switching the field from zero to 2.66H_c. In each node, three liquid branches emanate almost every 120°. As magnetic field strength quasistatically decreases, the four lobes as well as the branch that connects the two nodes become smaller. At approximately 1.52H_c, all the lobes become roughly the same size [see Fig. 3(b)] while at a critical strength ($\approx$1.45H_c) one of them is sharply incorporated into the main drop body. Then one of the nodes disappears and the 4-lobed cross section gradually evolves into a symmetric and stable 3-lobed shape [see Fig. 3(e)]. The measured difference between the lobe lengths in the 3-lobed configuration in Fig. 3(e) is less than 10%. The transition from 4- to 3-lobed shapes is clearly seen in the transient state photographs in Figs. 3(c) and 3(d). Three-lobed shapes were not only obtained by 4-lobed ones by slowly decreasing the field strength but also directly from circular shapes by a jump in the field strength. The latter procedure was not always successful and the circular cross section sometimes evolved into 2-lobed shapes.

Equilibrium states of a drop of volume 27.27 $\pm$0.25 mm^3, obtained as the magnetic field strength is varied in a stepwise manner, are shown in Fig. 4. The dimensionless sum of the lobe lengths is plotted versus the strength. In the case of the circular cross section this magnitude is the dimensionless diameter. Three successive routes are followed in order to elucidate the connectivity between noncircular, namely 3- and 2-lobed, and circular shape families. The first route starts from a symmetric 3-lobed shape at point *A* [photo (a)] and is marked with triangles; the field decreases slowly until the drop attains a circular cross section [point *B* and photo (e)]. The second route starts from the circular drop at point *B* and is marked with squares; the field strength increases slowly up to 1.62H_c, at point *C*. From point *C* starts the third route, again marked with squares, and is followed by slowly decreasing the strength until the drop regains a circular cross section (point *B*). Although the second and third routes are both marked with squares, notice that the directions of the guiding arrows are opposite. It is shown in the first route that when the field strength is decreased the sizes of the three lobes of the symmetric 3-lobed shape decrease but they remain almost equal until a critical strength is reached. This is seen better in Fig. 5 where the length of each lobe is plotted separately. The three lobes keep roughly the same size down to 1.31H_c where one of them sharply becomes smaller than the other two. Further decrease of the strength causes the smallest lobe to become even smaller without affecting the angle between them. Hence, although invariance under 120° rotations is broken, reflective symmetry remains. The latter configuration [photo (b) in Fig. 4] is stable down to 1.25H_c. At this critical strength the smallest lobe is rapidly incorporated into the main drop body and the cross section evolves quickly into a bent 2-lobed shape [photo (c)]. The bending subsides as the field strength decreases. At 1.12H_c the 2-lobed cross section is no longer bent and is becoming almost straight. Finally it evolves into a circular shape at 0.95H_c [photo (e)]. In the second route, the 2-lobed shape shows up at a higher strength—the critical one for this case—namely at H_c=165 G, verifying the hysteretic transition circle—dumbbell (2-lobed)—circle [11]. Further increase of the strength elongates the 2-lobed cross section [photo (f)] without causing any bending or branching up to 1.55H_c. Beyond this threshold, the elongated 2-lobed shape evolves into a markedly bent shape [photo (g)]. The bending subsides smoothly as the field strength decreases along the

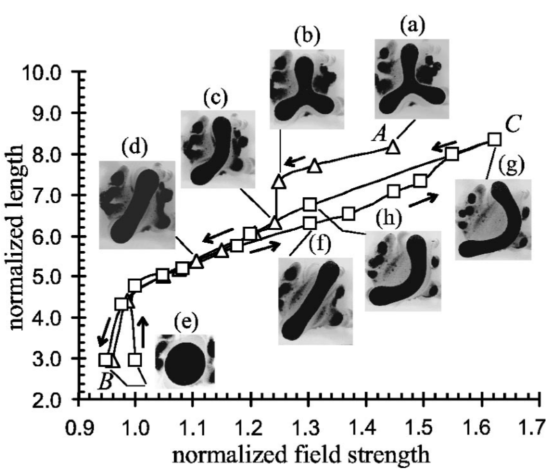

FIG. 4. Measurements of the normalized length of the cross section are plotted as a function of the applied field strength. The open triangles represent the evolution of 3-lobed shapes whereas the open squares the evolution of 2-lobed ones. Lines are drawn as a guide to the eye.

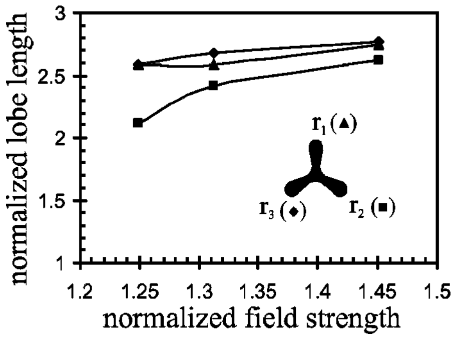

FIG. 5. Measurements of the normalized lobe lengths of the 3-lobed cross section as the magnetic field strength varies along a part of the first route in Fig. 4. Lines are drawn as a guide to the eye.

third route [photo (h)]. The third route meets the first one at $\approx 1.24H_c$ along which transitions from 3- to 2-lobed cross sections were observed. Namely, the straight 2-lobed shapes reappear at $1.12H_c$, where all three routes give almost the same equilibrium shapes. Thus, as the experiment shows, the bending transition is hysteretic. The corresponding hysteresis strength is $1.55H_c$-$1.12H_c = 0.43H_c$. In all the photographs shown, the black shadows are attributed to the liquid film left behind when the drop contact line recedes on the plates. To assure that the hysteresis is not due to the hysteresis on the advancing-receding equilibrium contact angles, all the experiments are carried out on already wet, with ferrofluid, plates.

In our experiments, almost any multibranched cross section can be driven into a symmetric 3-lobed configuration by slowly decreasing the field strength and successively incorporating a certain number of lobes in the main drop body. Symmetric 3-lobed shapes keep their symmetry of equal lobes down to a critical field where one of them (the slightly smaller one) ‘‘enters’’ the main drop body; a bent 2-lobed cross section then arises. However, starting from a circular cross section and slowly increasing the field strength no multibranched shapes are reached even if the field exceeds the value where stable 3-lobed shapes exist. Instead, 2-lobed shapes arise at a critical strength. Then, at higher field strength, a secondary transition occurs accompanied by the bending of the 2-lobed shape. The bending subsides hysteretically as the field strength decreases. It is thus suggested that no stable equilibrium path connects the 2- and the 3-lobed shape families, since the 2-lobed drops never become branched by slowly increasing the field. Since in all the experiments done, circular drops evolved into 3-lobed ones only by rapidly increasing the field strength passing over the 2-lobed solution family, no experimental evidence has been obtained regarding the emergence of the 3-lobed shape family through a primary bifurcation from the circular family. What is required is to drive the circular drop by slowly increasing the field (thus following equilibrium states) to the 3-lobed bifurcation. Suppressing, in some way, the 2-lobed perturbations, which lead to 2-lobed shapes, could only allow this. Suppression of 2-lobed disturbances is implemented by Ohsaka and Trinh [5] in the system of rigidly rotating liquid drops towards promoting the 3-lobed bifurcation. The unstable nature of the 3-lobed rigidly rotating drops [12] does not allow the appearance of gyrostatic equilibrium shapes; thus constantly changing shapes are observed as the drop rotates [5]. In contrast to the rotating drop in which 2- and 3-lobed shapes are unstable, in the ferrofluid bridge the later configurations are stable and static. Thus, shape control is more convenient. Since the corresponding experiments are simple and well controlled, the ferrofluid bridge offers convenience for testing experimental techniques so designed as to suppress or promote selected harmonic shape bifurcations; tests are underway and are promising.

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