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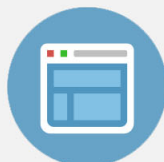
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## Ordered microdroplet formations of thin ferrofluid layer breakups

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The ordered breakup pattern of a thin layer of ferrofluid drop subjected to a uniform perpendicular field is experimentally investigated. The results confirm a universal pattern formation containing numerous breaking droplets of a uniform size, which is independent of the initial area of ferrofluid drop and the propagating directions of the formation waves. Two quantitative observations regarding the size and number of breaking droplets are concluded. Both the experiments and theoretical analysis show the correlation between the diameter of breaking droplets ( $d$ ) and magnetization strength ( $M$ ) can be characterized as  $d \propto 1/M^2$ . The uniform size of breaking droplets under a constant field strength results in a linear proportionality between the number of breaking droplets ( $N$ ) and the initial area of ferrofluid drop ( $A$ ) as  $N \propto A$ , which is verified by the experiments. © 2010 American Institute of Physics. [doi:10.1063/1.3298761]

### I. INTRODUCTION AND EXPERIMENTAL SETUP

The ferrofluids are colloidal suspensions in carrier fluids and can be affected by forces of magnetic polarization. Manipulated by an external magnetic field, the ferrofluids have been thoroughly investigated and widely used in various engineering applications.<sup>1-3</sup> In particular, interfacial instability on the free surface of a horizontal ferrofluid layer subjected to a uniform perpendicular magnetic field, which is referred to as Rosensweig instability as it was studied by Cowley and Rosensweig,<sup>4</sup> also interests researchers from both scientific and practical points of views. When the magnetic strength reaches a critical value, a sudden transition from an original flat surface into a hexagonal pattern of three-dimensional liquid crests, or the so-called Rosensweig crests, occurs. Patterns of these Rosensweig crests had been accurately demonstrated by means of radioscopy.<sup>5</sup> In addition, the dynamics of the crests has also been intensively studied both experimentally<sup>6-12</sup> and theoretically.<sup>13-16</sup> However, these researches focus mainly on the magnetic fluid layer at a relatively larger scale. With advances in microtechnology, it is interesting to further investigate the instability phenomena of an extremely thin layer, in which stronger effects by surface tension might lead to interesting behaviors.

Even though an interesting rupture in continuity of an extremely thin layer into individual droplets that preserve the hexagonal geometry had been reported near three decades ago,<sup>6</sup> only recently the Rosensweig instability of a thin film of a ferrofluid drop with diameter ranging at  $O(10^2-10^3)$   $\mu\text{m}$  on a dry plate was investigated in more detail.<sup>17,18</sup> The ferrodrops are observed breaking up into numerous subscale droplets with the breakup pattern dependent strongly on its initial diameter and field condition. This particularly simple phenomenon of drop rupture can possibly be applied as a noninvasive means for the partition of small drops as well as a tunable mechanism for orderly formation of microdroplets. Different patterns of striking rupture insta-

bilities are reported in a condition of dry plate.<sup>17,18</sup> To better understand the mechanisms acting on the ferrofluids as well as the important issues for practical applications, such as the number and volume of the breaking subscale droplets, a more comprehensive study<sup>19</sup> has been presented in which both the top and side views of breaking droplets are recorded and analyzed. Based on the fully three-dimensional observations, the film rupture mechanism is interpreted in terms of general dimensionless parameters. In addition, the uneven formation of breaking droplets on a dry plate is resolved by carrying similar experiments in a plate prewetted by a miscible nonmagnetic fluid. Furthermore, new interfacial instabilities in ferrofluids are reported<sup>20-22</sup> by immersing a ferrofluid drop in a miscible nonmagnetic fluid. Under such miscible circumstances in a similar perpendicular field,<sup>20,21</sup> a Rosensweig crest first grows rapidly, and then gradually decays, to ultimately reimmerse into the surrounding nonmagnetic layer. Associated with the decaying Rosensweig crest, a striking pattern of miscible labyrinthine instability<sup>23-27</sup> is triggered in the plane. The concurrence of a vertical Rosensweig crest and the in-plane labyrinthine pattern features new hybrid ferrofluid instability. If this immersed ferrofluid layer is placed in a radial field, visually striking patterns are obtained whose morphologies change from circular at zero field to complex starburstlike structures at a finite field.<sup>22</sup> The above findings indicate the initial contact to a miscible environment would alter the interfacial phenomena significantly. In the present study, breakup formation of a circular thin ferrofluid layer on a surface prewetted by a miscible fluid is investigated systematically. Quantitative conclusions regarding the numbers and sizes of breaking subscale droplets will be presented.

The experimental setup consists of a circular thin layer of a millimeter-sized ferrodrops, whose initial diameter is denoted as  $D$ , deposited on a glass plate prewetted by a miscible mineral oil and subjected to a uniform perpendicular magnetic field  $H$ , as the principle sketch shown in Fig. 1. The field strength is generated by a pair of coils in the Helmholtz configuration, so that a corresponding uniform magne-

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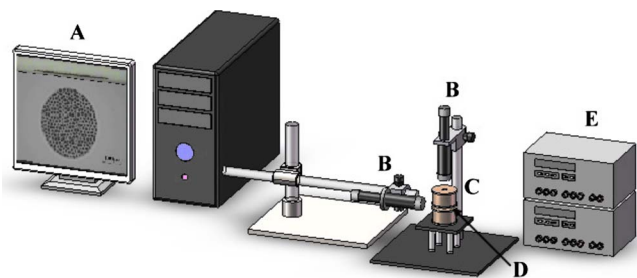


FIG. 1. (Color online) Principle sketch of experimental setup: the experimental setup consists of an extremely thin layer of ferrofluid drop placed on a glass plate (D) subjected to a uniform perpendicular field. The field strength is produced by a pair of coils in the Helmholtz configuration (C) powered by a computerized programmable power source (E). The top and side views are recorded by CCD cameras (B) and transmitted to a personal computer (A) for further analysis.

tization strength  $M$  is induced in ferrofluids. A light mineral oil-based ferrofluid (EMG901) produced by Ferrotec is used in the experiments. The saturation magnetization ( $M_s$ ), density ( $\rho$ ), and fluid/air surface tension ( $\gamma$ ) of this particular ferrofluid are  $M_s=600$  G,  $\rho=1530$  kg/m<sup>3</sup>, and  $\gamma=25$  mN/m, respectively. In each experiment, the power source is turned on instantaneously to generate a uniform field distribution and kept constant by fixing the current intensity. Three different field strengths of  $H=765$ , 676, and 588 Oe are experimented, whose correspondent strengths of magnetization are  $M=475$ , 466, and 454 G, respectively. To ensure uniformity of the magnetic field distribution, a SYPRIS 6010 Gaussmeter is used to measure the field strength in a  $7 \times 7$  mm<sup>2</sup> domain with a  $1 \times 1$  mm<sup>2</sup> resolution. We have observed that the maximum variation in the field strength is less than 1.5%, with strongest and weakest field strengths at the domain center and the edges, respectively. The time evolution of the interfacial morphology is directly recorded by charge-coupled device (CCD) cameras, providing the upper and side views of the surface. The cameras are connected to microscopes so that the pictures can be properly enlarged, and then transmitted to a computer for further analysis.

## II. RESULTS AND DISCUSSION

### A. A representative case

We first demonstrate the experimental results of a representative case for a drop with an initial diameters of  $D \approx 3.1$  mm under a uniform field strength of  $H=765$  Oe, as shown in Fig. 2. Both the snapshots of side views and top views are displayed in the top and bottom of each image, respectively. It is noticed that while the top view at the initial stage [bottom image in Fig. 2(a)] appears as a nice circular shape, the initial thickness of its side profile is extremely thin. As a result, the side view of the initial drop is not clearly recorded in the top image in Fig. 2(a). Nevertheless, if an initially flat profile is assumed, the corresponding thickness of the circular ferrofluid layer can be estimated by its volume and area. We have tested numerous cases within the drop sizes experimented, and consistent thicknesses of  $50 \pm 15$   $\mu$ m were obtained regardless of their initial diameters. As a result, all the drops can be analyzed assuming a

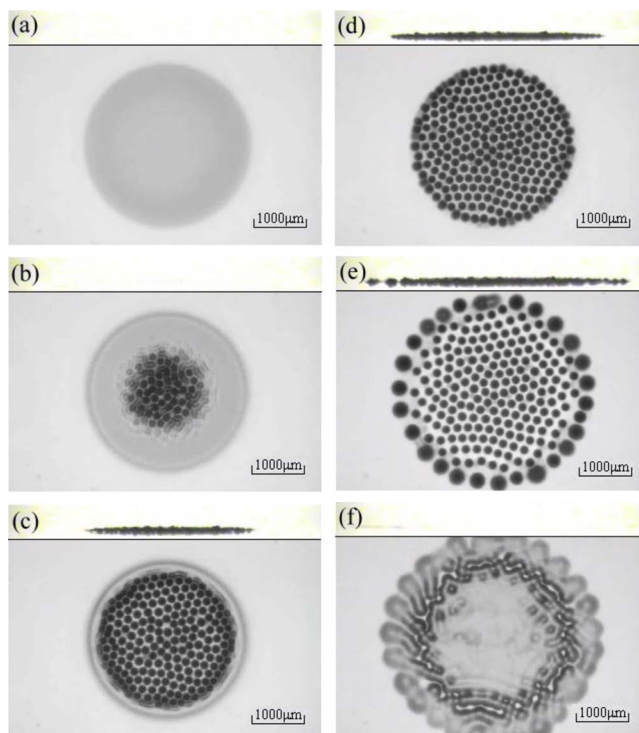


FIG. 2. (Color online) Snapshots of a drop with an initial diameter of  $D \approx 3.1$  mm under a field strength of  $H=765$  Oe at (a)  $t=0$  s, (b)  $t=0.2$  s, (c)  $t=0.6$  s, (d)  $t=0.8$  s, (e)  $t=1$  s, and (f)  $t=10$  s ( $H=0$  Oe). The side and top views of ferrofluids are displayed at the upper and lower portions of each image, respectively. For the situations without formation of Rosensweig crests at  $t=0$ , 0.2, and 10 s, the fluid layer is extremely thin. The side view images cannot be effectively captured by CCD camera at these times.

uniform thickness. Immediately after the presence of magnetic field, drop breakup starts to evolve in the central region at  $t=0.2$  s, as demonstrated in Fig. 2(b). The individual breaking droplets, or Rosensweig crests if observed from the side view, have not yet fully evolved at this early time stage, so that no distinguishable side images are recorded. As time proceeds, the pattern formation is propagated outward at  $t=0.6$  s and fully evolved at  $t=0.8$  s, as shown in Figs. 2(c) and 2(d), respectively. The fully evolved pattern appears as an orderly structure, which contains numerous breaking droplets of nearly the same size. Also observed in the side view image of fully evolved pattern is a slight height variation with a maximum at the center. Because of the magnetorepulsive force generated between the magnetized breaking droplets, the droplets apparently drift outwardly and result in coalescences of outer droplets at  $t=1$  s, as shown in Fig. 2(e). It should be noticed that the imperfect uniformity of field strength might induce motion of the droplets as well. Nevertheless, even if there does exist a slight field gradient, as described in Sec. I, the maximum field is located at the center of experimental domain. The induced motion by magnetic body force would be inward attraction, instead of outward expulsion. As a result, the present outward drift of the droplets should be mainly affected by the magnetorepulsive force. As time proceeds, similar behaviors of droplet drifts and coalescences keep occurring throughout the experiment until the removal of the magnetic field at  $t=10$  s, as shown in Fig. 2(f).

General features, such as the orderly pattern of breaking droplets and their outward drift, are consistent with the earlier results reported in Refs. 17 and 19. Additional interesting observations, which include the dynamical propagation of the pattern formation wave and coalescences of breaking droplets, are worthy of more detailed discussion. As discussed in Ref. 19, a weakest surface constraint occurs at the point of a maximum height. In the present situation, even though the initial side profile is nearly flat, a slight initial height variation is inevitable. Even the original image of the initial side profile cannot be obtained, the initial height variation can be confirmed from the consequences of slightly higher crests at the central region in the side view images of Figs. 2(c)–2(e). This explains the interfacial breakup would naturally start from the central region, where the surface constraints are the weakest, and consequently propagate outwardly. In addition, these breaking droplets at the circumferential region show significant diffusion with the surrounding prewetting fluids, and further weaken their strengths of magnetization. Lower magnetization strengths of the circumferential breaking droplets result in weaker local magnetorepulsive forces. As a result, the circumferential breaking droplets appear slower drifting speeds, compared with their inner counterparts. Since the outward drifting speeds are slower for these circumferential droplets, they are caught and merge with the inner droplets, which are expelled faster from behind by stronger repulsions. The coalescences at the rim increase the weights of merged droplets while the diffusion is enhanced by their outward motion, so that their drifts are further slowed down. The droplet coalescences proceed continuously after the full evolution of pattern formation.

## B. Influences of initial droplet sizes and field strengths

Effects of the initial size of the ferrodrops are demonstrated by a reference case of  $D \approx 1.8$  mm under the same field strength of  $H = 765$  Oe shown in Fig. 3. Similar pattern evolution to the previous larger drop (Fig. 2), which can be characterized qualitatively by an orderly structure and outward drifts of the breaking droplets associated with formation propagation from the center and droplet coalescences at the circumference, is observed. It should be noticed that this pattern is universally observed in various conditions of different drop sizes as well as field strengths. Nevertheless, quantitative influences, featured by the number of breaking droplets ( $N$ ) and their sizes ( $d$ ), might be altered significantly by initial drop sizes and field strengths, as shown in Figs. 4 and 5. Under a fixed field strength, a higher number of breaking droplets are generated for a larger initial drop. The correlation of numbers of breaking droplets and areas of the initial drop ( $A$ ) follows a linear proportionality, i.e.,  $N \propto A$ , as depicted in Fig. 4. Nevertheless, sizes of the breaking droplets are not affected significantly by the initial drop sizes. Diameters of the breaking droplets appear quite uniformly distributed regardless of the initial areas of ferrodrops, as shown in Fig. 5. On the other hand, both numbers and diameters of the breaking droplets depend strongly on the field strength for a constant size of initial ferrodrops. A higher number of breaking droplets are formed under a stronger

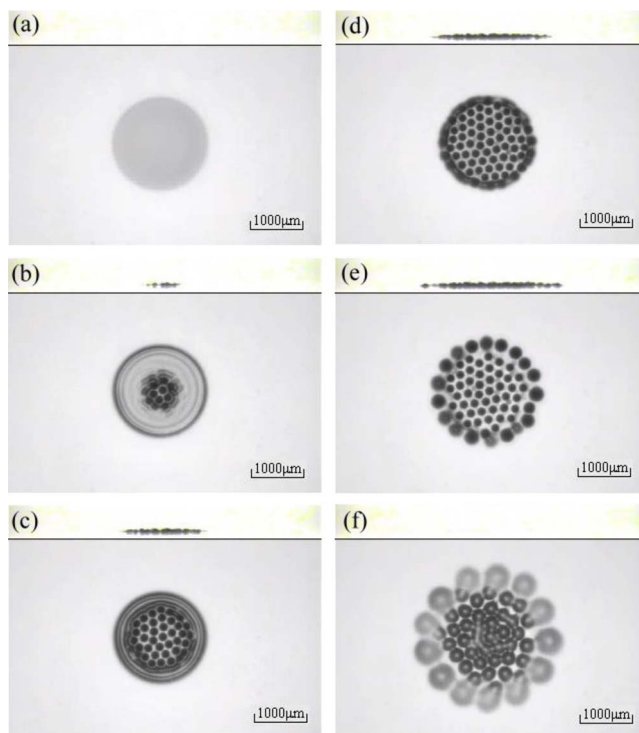


FIG. 3. (Color online) Snapshots of a drop with an initial diameter of  $D \approx 1.8$  mm under a field strength of  $H = 765$  Oe at (a)  $t = 0$  s, (b)  $t = 0.2$  s, (c)  $t = 0.6$  s, (d)  $t = 0.8$  s, (e)  $t = 1$  s, and (f)  $t = 13$  s ( $H = 0$  Oe). The side and top views of ferrofluids are displayed at the upper and lower portions of each image, respectively. No apparent side view images can be taken effectively at  $t = 0$  and 13 s, when no Rosensweig crests are formed.

field. On the contrary, a stronger field results in a smaller diameter of breaking droplets. The mean diameters of the breaking droplets under field strengths of  $H = 765$ , 676, and 588 Oe are  $d = 310$ , 328, and 350  $\mu\text{m}$ , respectively, as shown in Fig. 5.

The above quantitative observations can be explained by the forces acting on the ferrodrops. The governing mechanisms for the formation of breaking droplets can be determined by the balance of pressures as<sup>11</sup>

$$\gamma\kappa + \rho gz = \mu_0 \int_H M(H) dH + \frac{\mu_0}{2} (\mathbf{M} \cdot \mathbf{n})^2 - p_0, \quad (1)$$

where  $\kappa$  is the sum of the principal curvatures,  $\mu_0$  the permeability in vacuum, and  $\mathbf{n}$  the unit normal vector. The two terms on the left-hand side represent the surface and hydrostatic pressures, respectively. The first two terms on the right-hand side represent the induced pressures by the field and the pressure  $p_0$  is a constant reference pressure. For small droplets, the hydrostatic pressure and the reference pressure are negligible. The principal curvature can be roughly represented by the diameter of the breaking droplet as  $\kappa \propto 1/d$ , so that the above equation can be approximated as  $d \propto 1/M^2$  under the current condition of a uniform field strength. Considering the diameters of breaking droplets of  $d = 310$ , 328, and 350  $\mu\text{m}$  under the corresponding magnetization strengths of  $M = 475$ , 466, and 454 G, the proportionality of  $d \propto 1/M^2$  is well validated. It confirms that diameters of the breaking droplets are affected inversely by the mag-



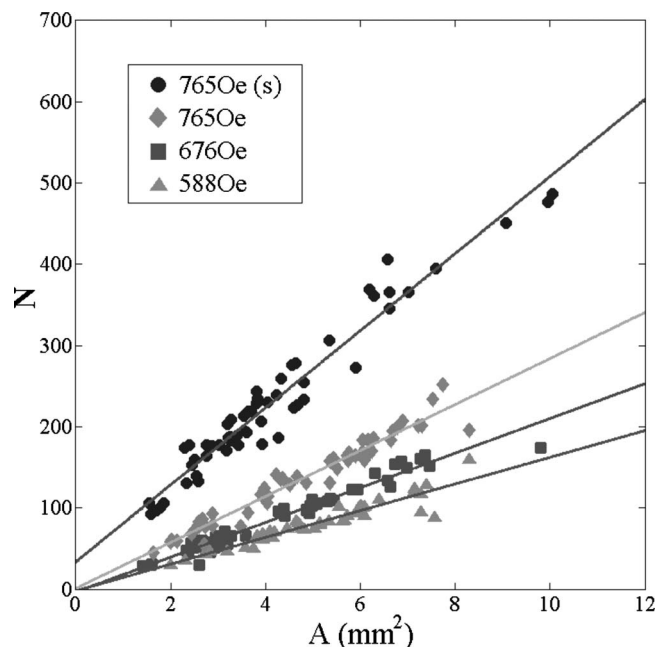


FIG. 4. Numbers of breaking subscale droplets  $N$  vs initial areas of ferrodroplets  $A$ : the correlation can be approximated by a linear proportionality of  $N \propto A$  for all the cases. The cases of oversaturated ferrofluid are indicated by (s) in the inset.

netized field strengths but independent of the initial areas of original ferrodrop, as shown in Fig. 5. Furthermore, the uniform size of breaking droplets under a constant field strength also explains the proportionality of  $N \propto A$ . By the same token, smaller breaking droplets formed under a stronger field naturally result in a higher number of droplets for the same size of initial ferrodrop, as shown in Fig. 4.

### C. Propagation of pattern formation wave

The propagation of pattern formation wave is an interesting issue and worthy of more discussion. In addition to the fascinating dynamics, it is also practical to realize if the

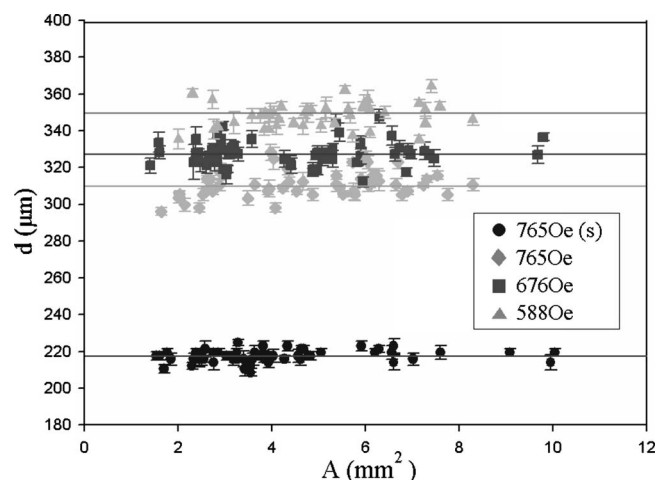


FIG. 5. Diameters of breaking subscale droplets  $d$  vs initial areas of ferrodroplets  $A$ : the sizes of breaking subscale droplets remain very close under the same field strength regardless of their initial areas. The cases of oversaturated ferrofluid are indicated by (s) in the inset.

propagating behavior affects the final pattern formation. As demonstrated in Figs. 2 and 3, the breakup of an initially well-deposited spherical cap always starts in the central region, where the constraints by surface tension are minimal because of smallest local curvature, and result in the outward propagations of pattern formation waves. To manipulate the starting location of breakup, a point defect on the initial surface is produced intentionally. It should be noticed that this initial defect might appear in practical situations should the present technique be massively applied in the formation of microscale orderly structures. To achieve this ill depositing condition, the ferrofluids are first exposed to the room temperature for 30 min before experiments. The exposure results in the evaporation of a slight amount of light mineral oils that serve as the based fluids of stable ferrofluids, and leads to an oversaturated condition with a higher concentration of ferroparticles in the exposed ferrodrop compared with its original state. Consequently, the concentration increase of ferroparticles for the oversaturated ferrofluid possesses a stronger magnetization strength as well as a higher viscosity. A similar process of exposing the ferrofluids to produce oversaturated ferrofluids had been applied to study the pattern formation of a microdrop.<sup>28</sup> One can take advantage of the higher viscosity to purposely produce a point defect on the initial profile, so that the breaking location can be manipulated as presented below. It is noted that the change in magnetization strength can also be used to validate the robustness of the quantitative correlation for the pattern formation in different magnetization situations.

Figure 6 demonstrates the pattern formation of a saturated ferrodrop with an initial diameter of  $D \approx 3.1$  mm under a field strength of  $H = 765$  Oe. The initial profile is still maintained as smooth as possible without apparent defects in this particular case, so it can be directly compared with the similar condition of its stable state without saturation, as shown in Fig. 2. A few obvious distinctions can be easily identified. The response time of breakup is significantly longer in the present saturated drop and the size of breaking droplets is also much smaller. In addition, the outward drifts of the breaking droplets no longer occur, so that no droplet coalescences are observed. As a result, the orderly pattern formation remains in a state of equilibrium until the removal of magnetic field. These distinctions are attributed by the change in fluid properties, such as a greater magnetization strength and the higher viscosity mentioned above. Because of the stronger viscous cohesion, it takes a longer time to break the ferrodrop. In addition, the higher viscosity is able to resist the magnetorepulsion, and prevents the breaking droplets from drifting outwardly. The smaller size of breaking droplets can be understood by the effects of a stronger magnetization from Eq. (1), and consequently leads to a higher number of breaking droplets as one would expect. Nevertheless, the overall pattern formation, such as orderly breakups as well as the propagation from the central region, is qualitatively similar to what had been observed in Fig. 2. The result indicates the orderly pattern formation can be universally preserved in different states of magnetization.

To manipulate the starting location of breakup, a point defect on the initial surface is intentionally produced, as the

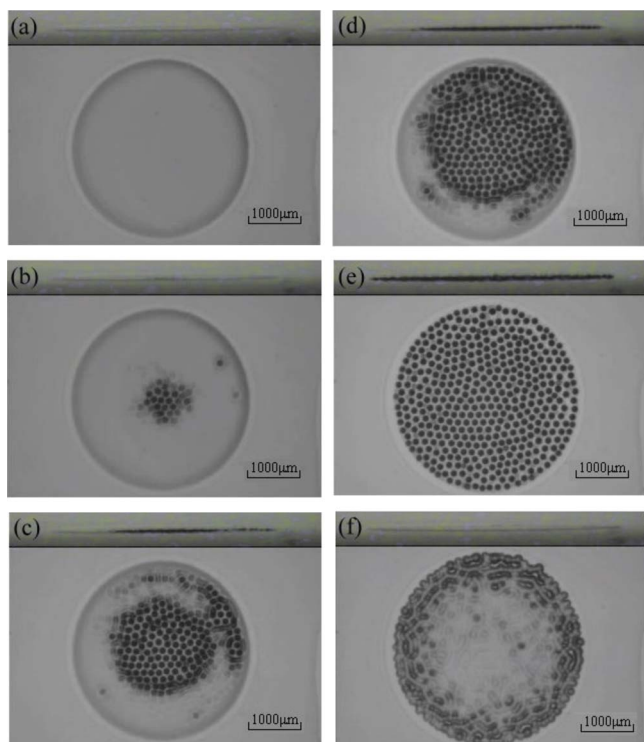


FIG. 6. (Color online) Snapshots of an oversaturated ferrofluid drop with an initial diameter of  $D \approx 3.1$  mm under a field strength of  $H = 765$  Oe at (a)  $t = 0$  s, (b)  $t = 5$  s, (c)  $t = 6$  s, (d)  $t = 7$  s, (e)  $t = 15$  s, and (f)  $t = 30$  s ( $H = 0$  Oe). The side and top views of ferrofluids are displayed at the upper and lower portions of each image, respectively.

cases shown in Figs. 7 and 8, whose initial diameters are  $D \approx 2.74$  and  $2.25$  mm, respectively. The point defect is introduced by using a needle to dip an insignificant amount of ferrofluids into the surface of the initial drop, so that smoothness of the surface is broken with a “point bump” appearing at the location of defect. The defects effectively cause the formation waves to start at the points desired and consequently propagate through the entire ferrofluid drop. It is interesting to note that the orderly patterns of breakups are still nicely preserved even with defects on the initial surfaces. In order to further study the influences of wave propagation, similar experiments for droplets of various sizes are carried and the quantitative measurements of interests, such as the number of the breaking droplets  $N$  and their diameters  $d$ , are displayed in Figs. 4 and 5. Again, the general arguments concluded in the previous stable conditions without oversaturation, i.e.,  $N \propto A$  and a uniform size of  $d \approx 217$   $\mu\text{m}$ , are well followed. Consistent with the reason stated in the previous paragraph, the saturated condition generally leads to higher numbers of breaking droplets with smaller diameters. These facts verify the universality of orderly breaking patterns regardless of the direction of their formation wave propagations.

### III. CONCLUDING REMARKS

In this study, the breakup patterns of a thin layer of ferrofluid drop subjected to a uniform perpendicular field are experimentally investigated. Orderly breakups into numerous droplets of a uniform size are obtained universally regardless

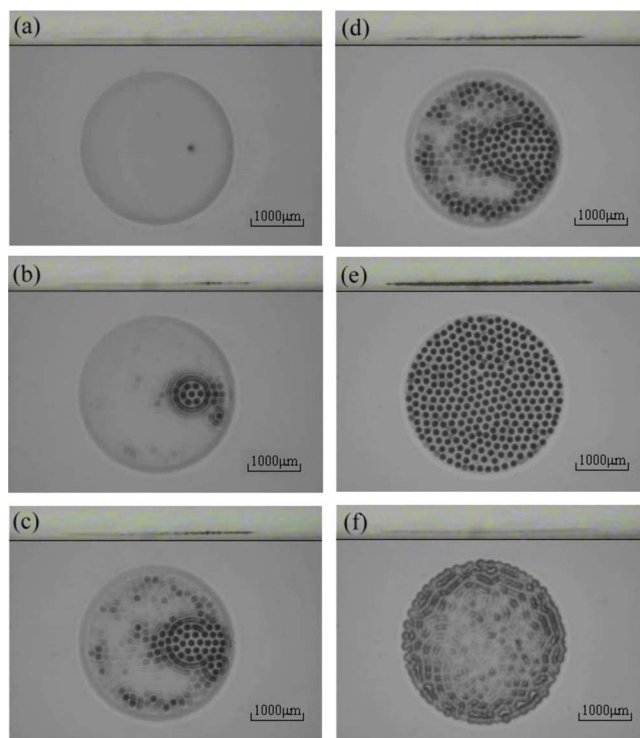


FIG. 7. (Color online) Snapshots of an oversaturated ferrofluid drop with an initial diameter of  $D \approx 2.74$  mm under a field strength of  $H = 765$  Oe at (a)  $t = 0$  s, (b)  $t = 6$  s, (c)  $t = 7$  s, (d)  $t = 9$  s, (e)  $t = 18$  s, and (f)  $t = 39$  s ( $H = 0$  Oe). The side and top views of ferrofluids are displayed at the upper and lower portions of each image, respectively. An intentional initial defect is produced at the right portion of the fluid surface, as shown in the top view of (a).

of their initial diameters and saturation states of the ferrofluid drops. For stable ferrofluid drops, whose concentrations of nanoparticles are not oversaturated, breaking droplets appear to drift in the radial direction because of the magnetorepulsion associated with weaker viscous resistances, and consequently proceed to continuous coalescences in the circumferential region. The pattern breakups always form from the central region, where the constraints by surface tension are the weakest, and formation wave propagates outwardly. On the other hand, if the ferrofluid drops are oversaturated by exposure to the air for 30 min before the experiments, the higher fluid viscosity prevents the breaking droplets from drifting and the orderly patterns remain in equilibrium. In addition, under the state of oversaturation it is possible to manipulate the starting locations of pattern breakups by intentionally producing point defects on the initial interfaces of ferrofluid drops. With the presence of point defects, the breakups are altered to start at the exact locations of the defects, instead of the central region, and propagate throughout the entire ferrofluid drop. Nevertheless, the results confirm that similar pattern formations are well preserved and not affected by the starting locations of the breakups and their propagating directions of the formation waves. This universal pattern formation is important for the practical situations, in which the drops might not be perfectly deposited initially.

The results also obtained two quantitative observations, such as the number of the breaking droplets ( $N$ ) and their

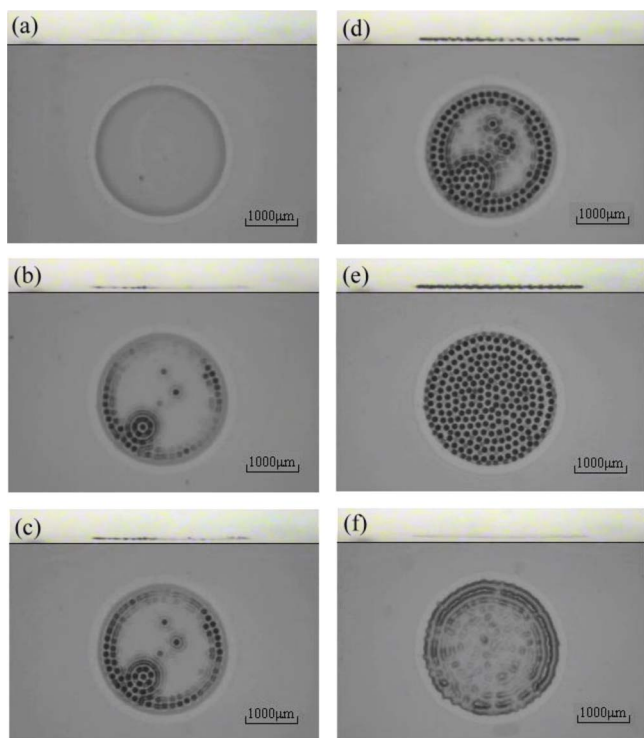


FIG. 8. (Color online) Snapshots of an oversaturated ferrofluid drop with an initial diameter of  $D \approx 2.25$  mm under a field strength of  $H=765$  Oe at (a)  $t=0$  s, (b)  $t=8$  s, (c)  $t=9$  s, (d)  $t=10$  s, (e)  $t=18$  s, and (f)  $t=30$  s ( $H=0$  Oe). The side and top views of ferrofluids are displayed at the upper and lower portions of each image, respectively. The initial defect is now produced at the lower left part of the fluid surface, as shown in (a).

diameter ( $d$ ), which can be concluded to describe the orderly breaking patterns. The diameters of breaking droplets are mainly determined by the magnetization strengths ( $M$ ) and are independent of the initial areas of the ferrofluid drops ( $A$ ). Both the experiments and theoretical analysis confirm the correlation of  $d \propto 1/M^2$ . The uniform size of breaking droplets under a constant field strength regardless of their initial areas would result in a linear proportionality of  $N \propto A$ , which is also verified by the experiments.

## ACKNOWLEDGMENTS

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