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Magnetically induced spreading and pattern selection in thin ferrofluid drops

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We report an experimental study of a fingering pattern formation which occurs during the spreading of an immiscible thin ferrofluid drop subjected to a radial magnetic field. Our results indicate that this ferrohydrodynamic system works as a magnetic analog of conventional spin coating, where centrifugal driving is replaced with a magnetic body force induced by the radial applied field. In this context, a magnetically tunable pattern selection mechanism is proposed in which the shape and number of the arising fingered structures can be properly controlled.

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Ferrofluids are a remarkable class of smart materials consisting of magnetizable nanometer-sized solid particles suspended in a nonmagnetic liquid carrier [1]. When a ferrofluid sample is subjected to an applied magnetic field, the magnetized liquid undergoes spontaneous deformation assuming a variety of shapes. Due to its visual appeal and prompt response to magnetic stimuli, this fluid material has become an archetypal dipolar system for the study of a number of pattern-forming processes in engineering and science [2], and even in art [3].

One emblematic example of pattern-formation phenomena in magnetic fluids is the Rosensweig instability [4,5]. It takes place when an initially flat free surface of a ferrofluid film is subjected to a perpendicular magnetic field generated by a pair of Helmholtz coils. When the magnetic surface force exceeds the stabilizing effects of gravity and surface tension, a stationary array of peaks is formed. Typically, the thickness of the ferrofluid layer is on the order of centimeters, being comparable to the wavelength of the unstable surface mode. Rosensweig peaks also arise in extremely thin ferrofluid films, whose characteristic thickness is considerably smaller [$O(10-10^3)$ μm]. Under such circumstances, an interesting film rupture process is displayed, where the peaks break up into individual droplets [6,7].

A small change in the conventional Helmholtz coil arrangement generates a completely different magnetic field geometry, resulting in still unexplored patterned structures. By reversing the direction of the current in one of the coils a purely radial magnetic field arises. A theoretical study of this situation [8], considering the response of confined immiscible magnetic droplets, revealed the development of polygon-shaped and starfishlike patterns. An experimental realization of such a radial field setup has also been studied [9], regarding the flow of miscible ferrofluid drops. The combination of negligible surface tension effects and radial field resulted in visually striking starburstlike structures, in which a great number of slim fingers grow radially outward.

An equally interesting aspect is the close connection between the radial magnetic body force and centrifugal forces [8,9]. This offers the promise of a magnetic analog for spin coating processes, in which the actual rotation of the system is not required. The phenomenon of spin coating—the flow of a thin liquid film spreading on a rotating horizontal surface—is a topic of considerable scientific and practical importance [10–12]. In this context, it is of fundamental importance to find ways to predict and control the shape of emergent patterns. In this paper, we introduce a control process for a system consisting of an immiscible thin ferrofluid drop subjected to a radial magnetic field. The focus of our study is twofold: first, we propose a simple addressable experimental method that serves as a magnetic equivalent to conventional spin coating; then, we present a magnetically assisted pattern selection mechanism that enables an improved control over the resulting pattern morphologies.

Our experimental setup is very simple. A ferrofluid drop is deposited on a flat glass plate and is placed in the midplane between a pair of Helmholtz coils. The magnetic fluid sample is a light mineral oil-based commercially available ferrofluid (EMG905) produced by Ferrotec. The viscosity and density of the ferrofluid are $\mu_d=9$ cp and $\rho_d=1.24$ g/ml, respectively. Its saturation magnetization is $M_s=400$ G. A radial magnetic field is applied $H_r=H_0 r/L$, where r denotes the radial distance. Because of the coils' body, the observing range is limited to the central hole of the coil whose radius is $L=15$ mm. H_0 is the averaged field strength at $r=L$. Three different electric currents $I=2.2$, 2.6, and 3.0 A are used, corresponding to magnetic field magnitudes $H_0=211$, 250, and 288 Oe, respectively. The time evolution of the drop is recorded from above by a charge-coupled device camera, providing an upper view of the situation. The camera is connected to a microscope, so that the pictures can be properly enlarged and recorded, and then transmitted to a computer for further analysis. We stress that, despite the relative simplicity of the setup, these experiments present quite challenging features. One major difficulty refers to the search for an exactly axisymmetric radial magnetic field distribution on the plane the ferrofluid drop. The almost unavoidable misalignment of the sample with respect to the midplane between the coils is an important practical

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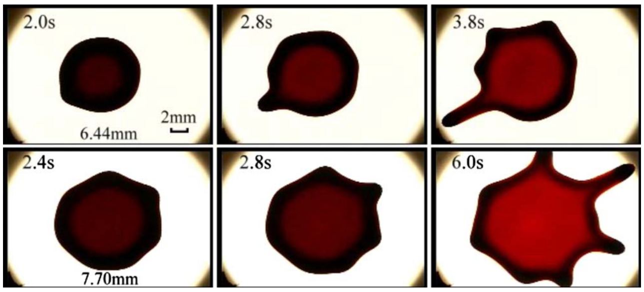

FIG. 1. (Color online) Time evolution of ferrofluid drops subjected to a purely radial magnetic field for two different initial drop diameters and $H_0=288$ Oe.

impediment. Another limitation is the imperfect manufacturing of the coils themselves. For a more detailed account about these aspects and the setup itself, we refer to Ref. [9].

Figure 1 shows a sequence of images from a run with radial field strength $H_0=288$ Oe and initial droplet diameters of $d=6.44$ mm (first row) and 7.70 mm (second row). The radial magnetic field produces a magnetic body force pointing radially outward, inducing the spreading of the ferrofluid drop along the radial direction. After the radial magnetic field is applied one observes a period latency of duration t_ℓ in which the droplet area does not change very significantly. At early stages $(t>t_\ell)$ one verifies the formation of small bumps at the border of the drop due to the destabilizing action of the applied field. This initial stage is followed by the growth of a simple dominant finger that tends to shield the development of other fingering structures. Only after the dominant finger reaches the inner edges of the coils the small fingers that have been left behind start to grow and compete. As expected, larger drops lead to more unstable interfaces. We point out to the great similarity between the patterns depicted in Fig. 1 and those appearing in spin coating experiments [10–12] with nonmagnetic fluids. There is in fact a more direct connection between the centrifugal force in rotating fluid systems and the magnetic force produced by a radial magnetic field: both point outward and increase linearly with the radial distance r [8,9]. In this sense, the situation illustrated in Fig. 1 can be seen as a magnetic counterpart of the usual spin coating problem.

We carried out various experimental runs for the radial field situation and performed quantitative measurements to investigate how the dimensionless drop area scales with dimensionless time $A' \sim t'^{\gamma}$, where area is rescaled using $A_\ell = A(t=t_\ell)$ and time is rescaled by $\tau=\pi\mu_d L^2/4\mu_0 A_\ell H_0^2$, with μ_0 denoting the magnetic permeability of empty space. By extracting data from these experiments we find the mean value of the exponent $\gamma=0.4897$, with standard deviation equal to 0.0310. This is in very good agreement with equivalent measurements executed for conventional spin coating experiments [10–12] in which the drop radius scales with time with exponent 1/4. This direct connection introduces the potential for a magnetically tunable coating technique.

In many fluid dynamics systems it is desirable to be able to prescribe the shape and symmetry properties of the resulting patterns, creating the possibility of designing materials uniquely targeted to specific technologically important applications [13,14]. However, as illustrated in Fig. 1 it is extremely hard to produce nice symmetric patterned structures partly due to the significant shielding effects, and inherent imperfections on the substrate, or in the magnetic field configuration itself.

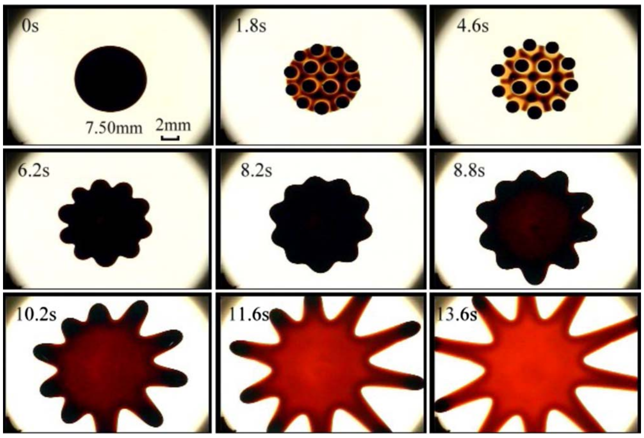

FIG. 2. (Color online) Time evolution of a ferrofluid drop subjected first to a perpendicular magnetic field, and then to a purely radial field ($d=7.5$ mm and $H_0=211$ Oe).

To overcome these difficulties, we propose a control process which enables one to set a given initial condition through magnetic means, and then use the radial field to induce the growth and spreading of a pattern with desired symmetry and shape. Instead of applying a radial field from the beginning, we first apply a perpendicular magnetic field and use it to prescribe an initial condition of interest. This innovative selection protocol has been inspired by the recent results of Refs. [6,7]. In these experimental studies an initially circular thin ferrofluid drop breaks up into numerous subscale droplets, which form various tiny three-dimensional crests, keeping a nearly circular shape for the perimeter of the resulting structure. This constitutes a thin-film version of the usual Rosensweig instability [4,5]. Our goal is to be able to manipulate the number of subdroplets located at the rim of the array.

It turns out that it is very simple to incorporate a perpendicular field by using our experimental setup. A magnetic field applied normal to the ferrofluid sample is produced by passing an electric current only through the bottom coil. As a result, the subscale droplets arise and tend to drift outwardly due to magnetic repulsion. After that, the perpendicular field is turned off and immediately replaced with a radial field by applying opposing currents in both coils. As the perpendicular field is removed, the various small peaks collapse and the droplets coalesce. This forms a new continuous structure presenting a well-defined number of small fingers, originated from the outermost peaks originally produced by the perpendicular field. The radial applied field acts on these fingers creating a symmetric radially growing fingering pattern.

This magnetically induced selection mechanism is illustrated in Fig. 2 which depicts a typical time evolution of a drop of initial diameter $d=7.5$ mm, subjected to a magnetic field strength $H_0=211$ Oe. Due to surface tension, in the absence of applied field, the drop is nearly circular ($t=0$ s). First, a perpendicular field is applied generating an array of

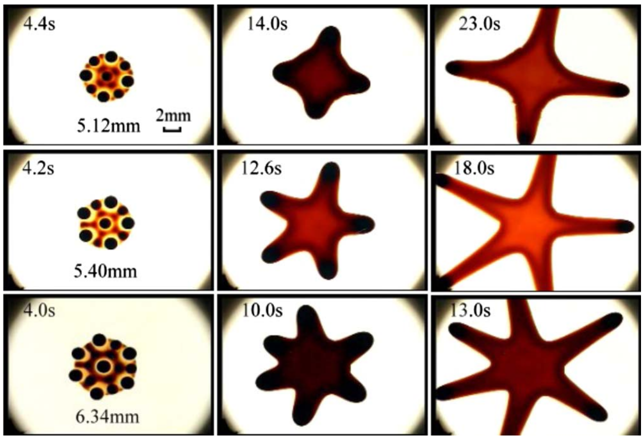

FIG. 3. (Color online) Illustration of the magnetic-field-induced pattern selection mechanism for $H_0=211$ Oe and three different initial drop diameters.

numerous subscale droplets ($t=1.8$ s). A total number of ten roughly equal-size droplets emerge along the outer circular rim. At time $t=4.6$ s the ten droplets have drifted away. At time $t=6.2$ s, the perpendicular field is turned off, and the radial field is immediately switched on. This leads to the collapse of the peaks, followed by the coalescence of the subscaled droplets, creating a tenfold symmetric perturbed drop reminiscent of the limiting ten droplets previously formed ($t=8.2$ s). Subsequently, ten fingers start to spread out due to the action of the radial magnetic force as shown at times $t=8.8$, 10.2, and 11.6 s. All the fingers are fully evolved at $t=13.6$ s. This reference case clearly demonstrates the capability of tuning the ultimate number of growing fingers by prescribing a proper initial perturbation with the assistance of a perpendicular field. We denote the number of subscale droplets at the outermost boundary by N_p and the final number of evolving fingers by N_f. In Fig. 2 we have that $N_p=N_f=10$.

We proceed by briefly discussing an important difference between the pattern-formation processes depicted in Figs. 1 and 2. In the purely radial applied field in Fig. 1, as well as in spin coating situations [10–12], there is a significant fluid accumulation at the perimeter of the drop during the entire experiment. Although dominant fingers in Fig. 1 evolve carrying a significant amount of fluid material at their fingertips, a considerable amount of ferrofluid can be still observed at the rim. On the other hand, the initial perturbations induced by the perpendicular field in Fig. 2 create pathways for the transport of ferrofluid already at earlier times. For instance, at $t=11.6$ s almost all ferrofluid is accumulated at the fingertips. After this, only these fingers tend to evolve, while the main body of the droplet spreads quite insignificantly. This accumulation at the fingertips explains the favorable development of the initial perturbations throughout the growth process and results in a larger growth rate for the situations illustrated in Fig. 2.

To obtain a desired pattern, it is essential to manipulate the number of initial perturbations N_p. Two major contributions influence the determination of N_p: the magnetic field magnitude and the size of the initial drop [6,7]. Figure 3 displays the time evolution of fingering patterns for three different initial drop sizes $d=5.12$ (first row), 5.40 (second row), and 6.34 (third row) mm keeping $H_0=211$ Oe. In Fig. 3 it is evident that larger drop sizes lead to the rising number of resulting subscale droplets. Moreover, in contrast to what has been observed in Fig. 2 the subscale droplets shown in Fig. 3 are not completely uniform in size, but smaller secondary subscale droplets appear between the primary ones located at outer circular perimeter. However, only the primary subscale droplets show significant outward drift and result in effective initial perturbations.

The uniformity in size of the subscale droplets is determined by the flatness of the initial ferrofluid drop. For smaller droplets whose surface tension is more prominent, the height variation along the vertical direction (normal to the plate) is significant. This favors a drop breakup into subscale droplets of different sizes. On the other hand, due to gravitational effects larger drops present a more uniform vertical profile resulting in the formation subscale droplets of similar sizes [6,7]. Figure 3 illustrates that by tuning the initial drop size we can make N_p coincide with the final number N_f, so that $N_p=N_f=4$, 5, and 6 for $d=5.12$, 5.40, and 6.34 mm, respectively. This constitutes a clean pattern selection mechanism involving the interplay of drop size and magnetic effects. Complementary information about the effect of initial ferrofluid drop sizes in selecting the ultimate pattern morphologies is summarized in Fig. 4(a). For $H_0=211$ Oe and also for $H_0=250$ Oe, we see that $N_p \approx N_f$, both increasing nearly linearly with increasingly larger initial drop diameter d. Notice that N_p increases as the field strength is raised, leading consequently to higher values of N_f. Nevertheless, as illustrated in Fig. 4(b) discrepancies between N_p

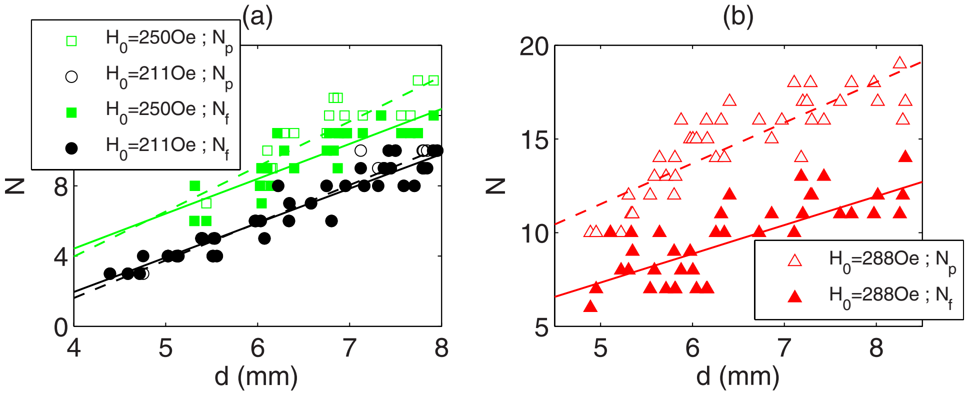

FIG. 4. (Color online) The number of initial interface perturbations N_p and the final number of fingered structures N_f as functions of the drop diameter d for (a) $H_0=211$ and 250 Oe; (b) $H_0=288$ Oe. Note that in (a) $N_p \approx N_f$, and in (b) $N_p > N_f$.

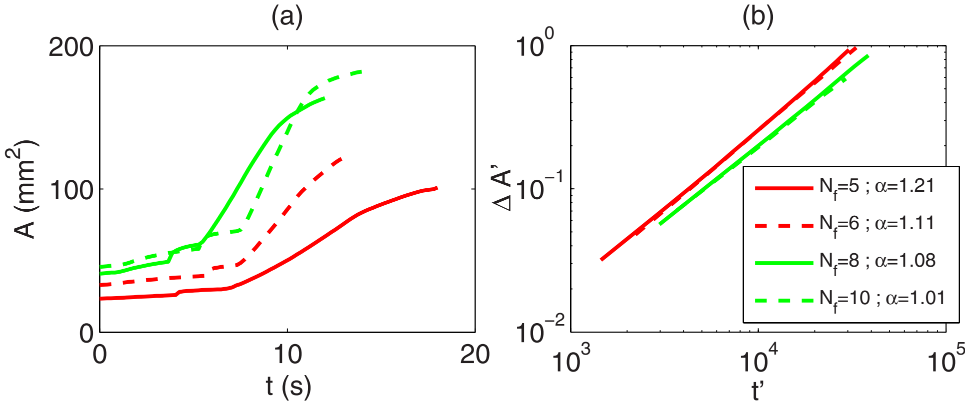

FIG. 5. (Color online) (a) Time evolution of the drop expanding area in dimensional form and (b) time evolution of the dimensionless area increment $\Delta A'$. Curves correspond to a few illustrative values of N_f and α.

and N_f begin to emerge for higher field magnitudes: $N_f < N_p$ for the case of $H_0 = 288$ Oe. The discrepancy becomes more apparent for droplets of larger sizes. This indicates that the selection mechanism works better for smaller drops, and not so high magnetic fields.

Figure 5(a) plots the time evolution of the expanding ferrofluid drop area for a few typical situations and $H_0 = 211$ Oe. For the reference case of Fig. 2 ($N_f = 10$), the area increases modestly up to $t = 5.28$ s. This period of low increase corresponds to the outward drifting of the subscale droplets. After that, a sudden jump is observed when the perpendicular field is removed and replaced with the radial field, so that the breaking crests collapse. Following a latent period of insignificant growth, the area grows consistently at $t = 7.5$ s due to the development of outward moving fingers. As in the purely radial field case, the starting time of this uniform growth period is denoted by t_ℓ. Finally, the growth rate starts to decline at $t = 10.5$ s when some of the fingers have reached the inner edges of the coils. A similar behavior is observed for $N_f = 5$, 6, and 8. Figure 5(b) focuses on this period of uniform growth and shows a log-log plot of the dimensionless area increment $\Delta A' = A' - 1$ as a function of dimensionless time t'. The area has been scaled by the drop area A_ℓ at $t = t_\ell$, and the time is rescaled by τ. For more than 100 experiments we have performed for various values of H_0, we have verified the scaling behavior $\Delta A' \sim t'^{\alpha}$ with mean value $\alpha = 0.999\,59$ and standard deviation of 0.0693. This clearly indicates a larger growth rate behavior in comparison to purely radial magnetic field case and regular spin coating.

In conclusion, our experimental results suggest an alternative to regular spin coating for thin ferrofluid film spreading, where centrifugal force is replaced with a radial magnetic body force. They also demonstrate the feasibility of a magnetically tunable pattern selection mechanism which provides a predicable way to obtain symmetric shapes with a specific number of fingers. This is achieved by adjusting the strength of applied perpendicular and radial magnetic fields and the diameter of the initial circular ferrofluid drop. These results highlight research prospects of a truly multidisciplinary topic that merits further investigation.

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