PHYSICAL REVIEW E **68**, 035301(R) (2003)

Orientational preference and predictability in a symmetric arrangement of magnetic drops

David P. Jackson
Department of Physics and Astronomy, Dickinson College, Carlisle, Pennsylvania 17013, USA
(Received 30 May 2003; published 8 September 2003)

An investigation of a symmetrical arrangement of N quasi-two-dimensional magnetic domains in an external field is carried out. By minimizing the linearized interaction energy for this arrangement using a nearest-neighbor approximation, an orientationally preferred state of the system is found. This orientational preference leads to a large degree of predictability in the final patterns as demonstrated by some experiments using ferrofluids. The final state patterns are also investigated by carrying out a series of numerical simulations. These simulations exhibit a similar predictability and the final patterns bear a strong resemblance to those obtained experimentally.

DOI: 10.1103/PhysRevE.68.035301 PACS number(s): 47.54.+r, 47.20.−k, 75.50.Mm, 75.70.Ak

When a ferrofluid drop is placed in a Hele-Shaw cell and subjected to a perpendicular magnetic field, it undergoes a fingering instability that can lead to a complex, labyrinthine pattern [1,2]. This same kind of pattern has been observed in a number of different systems, including amphiphilic monolayers, garnet films, chemical reaction-diffusion systems, and type-I superconductors [3]. To understand this process, a number of researchers have undertaken analytic and numerical studies of a single domain system [4–8], and the results have been applied to both monolayers [9–11] and superconductors [12]. In all of these studies, no effort was made to incorporate the effects of multiple domains. Since there are typically *many* interacting domains in these pattern forming systems, it is of interest to study how the multidomain interactions can affect the pattern formation process.

Two basic approaches have been used to study these interactive effects. In one, a highly disordered array of ferrofluid drops was studied numerically as the system evolved into the labyrinthine phase [13]. In the other, a two-domain configuration was used to analytically determine the orientational preference of the domains early in the pattern formation process [14]. In this paper, the analytic two-domain calculations are extended to include an N-domain configuration in which the drops are arranged symmetrically at the vertices of a regular polygon. As in Ref. [14], the linearized interaction energy for the system is minimized with respect to droplet rotations to find the energetically preferred orientations. In addition, a series of numerical experiments are performed to explore this system far into the nonlinear regime and the results are compared to experiments.

The physical system to be investigated consists of N equal-sized ferrofluid drops with initial radii R_0 contained in a Hele-Shaw cell consisting of two closely spaced glass plates separated by a distance h. These drops are symmetrically arranged at the vertices of a regular N-sided polygon as shown in Fig. 1. When a magnetic field is applied perpendicular to the Hele-Shaw cell, each drop undergoes a fingering instability that leads to a branched structure. If these structures are close enough together, the interactions between them can affect the pattern formation process.

As is common in these systems, the magnetization M is taken to be collinear with the applied field and uniform throughout each of the domains. If the ribbon-shaped boundary of the ith domain is parametrized by s_i, then a point on this boundary can be described by a two-dimensional vector $\mathbf{r}_i=(x(s_i),y(s_i))$ that gives the location along the ribbon, and a point z_i that gives the height. When considering a multidomain system, each drop will contribute a self-energy term, but there will also be interaction terms of the form

$$\mathcal{E}_{ij}=-M^2h\oint ds_i\oint ds_j\hat{\mathbf{t}}_i\cdot\hat{\mathbf{t}}_j\Phi(R_{ij}/h), \tag{1}$$

where $i\neq j$. Here, $R_{ij}=|\mathbf{r}_i-\mathbf{r}_j|$ is the *in-plane* distance between a point on the ith contour and a point on the jth contour, $\hat{\mathbf{t}}_i$ and $\hat{\mathbf{t}}_j$ are unit tangent vectors at these two points, and

$$\Phi(\xi)=\sinh^{-1}(1/\xi)+\xi-\sqrt{1+\xi^2} \tag{2}$$

is a potential function that gives the strength of the interaction between these points. Since we are only interested in the interactive effects of the pattern formation process, the self-energy terms need not be included in the analysis. This

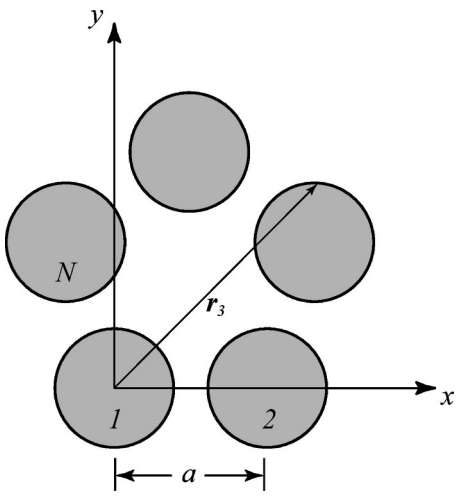

FIG. 1. Sketch showing the geometric arrangement of, in this case, five ferrofluid drops. Drop 1 is located at the origin, drop 2 is located a distance a along the x axis, and successive drops are located at the vertices of a regular N-sided polygon with edge length a.

means that surface tension plays no (direct) role in determining how these domains interact [15].

The total interaction energy of the system is obtained by summing the interaction energy between each pair of domains [16], which can be written

$$\mathcal{E}_{\rm int}=\sum_{j=2}^{N}\mathcal{E}_{1j}+\sum_{j=3}^{N}\mathcal{E}_{2j}+\sum_{j=4}^{N}\mathcal{E}_{3j}+\cdots. \tag{3}$$

When pure mode perturbations are introduced on the drops of the form $\zeta_i\cos[n_i(\theta+\alpha_i)]$, then two drops separated by a distance a have a linearized interaction energy proportional to (see Ref. [14])

$$\mathcal{E}_{ij}=I_{ij}^{(0)}+\zeta_i A_{ij}\cos n_i\alpha_{ij}+\zeta_j A_{ji}\cos n_j\alpha_{ji}. \tag{4}$$

Here, ζ_i is the amplitude of the perturbation on the ith drop, n_i is the mode number (number of protuberances) on the ith drop, α_{ij} is the (clockwise) rotation angle of drop i with respect to drop j, and A_{ij} is the *amplitude coefficient*, which depends on the mode number of the ith drop n_i and the distance between drops a. Also in Eq. (4), $I_{ij}^{(0)}$ represents the interaction between two perfectly circular domains.

Due to the symmetric arrangement of the drops, we can, without loss of generality, focus our attention on a single drop. Having found the preferred rotational angle for this drop, the preferred angles for the other $N-1$ drops can then be determined by symmetry. Inspection of Eq. (4) reveals that the only pieces of Eq. (3) that depend on the rotation of drop 1 come from the $\mathcal{E}_{1j}$ terms. Therefore, we can turn our attention to drop 1 and write down the portion of the interaction energy that depends on the orientation of drop 1, namely,

$$\mathcal{E}_{\rm int}^{(1)}=\sum_{j=2}^{N}\zeta_1 A_{1j}\cos n_1\alpha_{1j}. \tag{5}$$

To minimize this energy with respect to rotations, the rotation angles α_{1j} must be replaced with a single angle that can be used as the minimization parameter. Using the geometry shown in Fig. 1 gives the relation $\alpha_{1j}=\alpha_{12}+(j-2)\pi/N$. Plugging this into Eq. (5) and minimizing with respect to α_{12} will then lead to the preferred angles α_{12}^*. Unfortunately, the complexity of the amplitude coefficients precludes us from finding a closed form expression for α_{12}^*. Nevertheless, we can make analytic progress by including only nearest-neighbor interactions in the calculations. Since it is experimentally verified that the interactions do not play a prominent role in the pattern formation process when $a\gtrsim 4R_0$, this is expected to be an excellent approximation.

By including only nearest-neighbor interactions, only two terms survive from the sum in Eq. (5), $j=2$ and $j=N$. Moreover, because the distance between drops 1 and 2 is the same as the distance between drops 1 and N, the two amplitude coefficients are equal, $A_{12}=A_{1N}$. Differentiating $\mathcal{E}_{\rm int}$ with respect to α_{12} and setting it equal to zero leads to extreme angles α_{12}^* given by

$$\tan n_1\alpha_{12}^*=-\frac{\sin\left(\frac{N-2}{N}n_1\pi\right)}{1+\cos\left(\frac{N-2}{N}n_1\pi\right)}, \tag{6}$$

which is valid as long as the denominator is nonzero. To find the *minimum* energy states, we require

$$\left.\frac{\partial^2\mathcal{E}_{\rm int}^{(1)}}{\partial\alpha_{12}^2}\right|_{\alpha_{12}^*}>0, \tag{7}$$

which leads to

$$\alpha_{12}^*=-\frac{N-2}{2N}\pi+\frac{l\pi}{n_1}, \tag{8a}$$

where l is an integer that satisfies

$$2l+1<\frac{N-2}{N}n_1<2l+3. \tag{8b}$$

It is worth noting that when $(N-2)n_1/N$ is an odd integer, which corresponds to the denominator in Eq. (6) being equal to zero, the interaction energy does not have a minimum. This is due to a symmetry between the mode number n_1 and the number of drops in the system N. In these situations, the pattern selection mechanism may be due to next-nearest-neighbor interactions or, perhaps more likely, due to nonlinear effects. This interesting situation is currently under further investigation. For the remainder of this paper, we will simply assume that we are dealing with an energy minimum.

Although we have focused our attention on a single drop, the situation is entirely symmetric so that every drop will behave in a similar manner. Furthermore, if each drop has the same initial radius, one should expect that the same mode number will develop on each of the drops [7]. Thus, let us consider situations in which the mode numbers $n_i\equiv n$ are the same for all drops in the system.

First, consider a two-drop system in which each drop has a mode $n=2$ perturbation. This will be referred to as an $N=2$, $n=2$ configuration. In this situation, Eqs. (8) give $l=-1$ and thus a preferred angle for drop 1 of $\alpha_{12}=-\pi/2$. The rotation angle for drop 2 is found by symmetry. A similar procedure for an $N=5$, $n=3$ configuration leads to a preferred angle of $\alpha_{12}=-3\pi/10$. Again, the rotation angles for the other drops are found by symmetry. These two situations are shown in Fig. 2 along with some typical experimental results obtained using ferrofluids.

In the ferrofluid experiments, the plate spacing was about 1 mm and the drop radius was about 1 cm. The drops were formed by using a small hand magnet and ''tearing off'' a small amount of ferrofluid from a large ''reservoir.'' This process was not very accurate but it did allow us to obtain drops that were approximately the same size, within about 10%. Placing the drops in a symmetric array was easily done using the hand magnet. The drops would surround the magnet without fusing together and when the magnet was slowly lifted, the drops would relax onto the vertices of a polygon

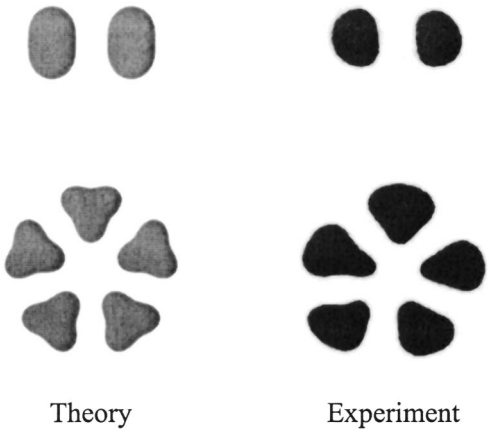

FIG. 2. The minimum energy configuration for an $N=2$, $n=2$ configuration (top) and an $N=5$, $n=3$ configuration (bottom) as determined by Eqs. (8), along with some typical experimental results.

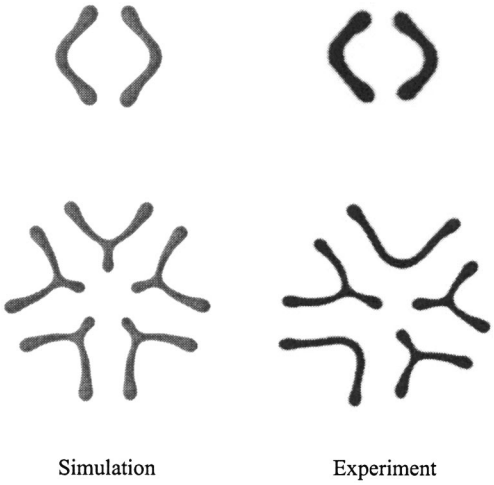

FIG. 3. Simulation and experiment for an $N=2$, $n=2$ configuration (top) and an $N=5$, $n=3$ configuration (bottom). The experimental results are the late stage of the evolution shown in Fig. 2.

with an edge length of approximately $2.5R_0$. A perpendicular magnetic field was ramped up by hand from zero to a maximum of about 200 G. Varying the rate at which the magnetic field was applied allowed us to select the mode number (to some degree) introduced on the drops as described in Ref. [7]. The evolution of the drops was captured with a video camera.

To study this system far into the nonlinear regime, it is important to include single domain interactions as well as interactions between the domains. The Navier-Stokes equation for a ferrofluid in a Hele-Shaw cell is

$$\mathbf{v}=-\frac{h^2}{12\eta}\nabla\Pi, \tag{9}$$

where η is the ferrofluid viscosity and Π is a generalized pressure that includes the magnetic effects. Incompressibility of the ferrofluid leads to Laplace's equation inside the domains. Assuming the pressure outside the ferrofluid is constant renders the domains hydrodynamically isolated. The only interactive effects are then due to the magnetization of the domains and appear through the generalized pressure at the boundary of a domain,

$$\Pi(\gamma_i)=\sigma\kappa(\gamma_i)+\frac{2M^2}{h}I(\gamma_i). \tag{10}$$

Here, σ is the surface tension, κ is the interface curvature, γ_i is an arbitrary parametrization of the ith interface, and I is the magnetic contribution. This magnetic contribution is an integral that depends on the shapes of *all* the domains and therefore represents a highly nonlocal interaction. The evolution equation for the interface is derived using the conformal mapping techniques as described in Ref. [17] and implemented in Ref. [7].

In the experiments, a water/tween mixture was used outside the ferrofluid to prevent sticking with the glass plates as much as possible. The addition of this outer liquid could lead to some hydrodynamic effects that are not accounted for in the simulations. In the simulations, sticking with the glass plates is modeled simply by stopping the calculations when the motion has reached some reasonably stable configuration.

Figure 3 shows some typical simulation and experimental patterns for the $N=2$, $n=2$ and $N=5$, $n=3$ configurations. The similarity between the experiments and simulations is quite striking. The experimental patterns in Fig. 3 are the late stages of the evolutions shown in Fig. 2. The numerical experiments were performed using circular initial states with a small amount (less than 1% of the circle radius) of random noise distributed in the first eight azimuthal modes. The number of points on each circle is increased as the evolution develops to preserve accuracy, but as few points as possible are used in the interest of speed.

The results shown here are fairly robust and appear virtually every time the experiment or the simulation is performed in these configurations. One obvious difference in the $N=5$, $n=3$ configuration is that two of the domains in the experiment have lost their third ''center-pointing'' finger. This was a common occurrence in the experiments. Typically, all five drops would begin with three fingers and appeared almost identical to the simulation. However, at some point, some of the drops would lose their center-pointing fingers. In Fig. 3, we see that two of the drops have lost this finger. This effect appears to be related to the initial positions of the drops which is not terribly accurate in these simple experiments. In addition, the simulations have been stopped when the evolution has slowed down considerably to represent the small amount of sticking with the glass plates. In fact, if we allow the simulation to continue to run, all five drops will lose their center-pointing fingers, similar to what happens in the experiments.

The numerical results show excellent agreement with the experiments, even though the outer fluid in the experiments is not air. This suggests that these domains behave as if they were hydrodynamically isolated. One can see some small differences between the experiments and the simulations, but by and large the agreement is surprisingly good.

The present work has concentrated on a particular sym-

metric arrangement of magnetic fluid drops and has led to some understanding of how multidomain interactions affect the pattern formation process in these systems. Much of the behavior seen here can also be seen in more complex scenarios, such as the disordered array of drops studied numerically by Drikis *et al.* in Ref. [13]. In addition, this study provides some insight into how the early stages of the pattern formation process can have a dramatic impact on the final state patterns that form. In almost every case studied, the ''backbone structure'' of the entire pattern appears to be ''frozen in'' very early in the pattern formation process.

It is also interesting to be able to make a direct comparison between the theoretical equations and the actual experiments. Normally in these systems, there is a large amount of randomness that precludes making direct physical comparisons between specific final state patterns. But in this symmetric situation, the final state patterns are very predictable and we can therefore compare specific patterns since they repeatedly arise. The striking similarity between the numerical results and the experimental results suggests that the dynamics of the domains are essentially hydrodynamically independent.

One can imagine extending the analysis here to a symmetric arrangement of drops which covers the plane. There should be no problem accomplishing this task for square or hexagonal lattices. In these cases, one would expect nearest-neighbor interactions to play a role on multiple sides of each drop. This would likely lead to increased frustration in the system and therefore less predictability in the final state patterns. For example, in a square lattice, if all drops had an $n=2$ mode perturbation, it is unlikely that they would all evolve parallel to each other during the initial stages of the pattern formation process. That would lead to fingers growing directly toward each other. Instead, one might see a pattern whereby each neighboring drop is oriented 90° with respect to each nearest neighbor.

In addition to square and hexagonal lattices, one can imagine trying to cover the plane in a quasiperiodic manner. In this case, there would be even more frustration caused by the nonperiodic nature of the lattice in addition to that caused by an increase in nearest neighbors. Although it is difficult to imagine what effects this might have, it seems likely that it would result in even less predictability of the final state patterns.

I would like to acknowledge some previous students whose contributions set the groundwork for much of what is presented here. They are Matthew Welker, Russell LaMantia, Brennan Gantner, and Matthew Grivich. This work was supported in part by Research Corporation Grant No. CC4693.

[1] R.E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, Cambridge, 1985).
[2] A.J. Dickstein *et al.*, Science **261**, 1012 (1993).
[3] M. Seul and D. Andelman, Science **267**, 476 (1995).
[4] A.O. Cebers and M.M. Mairov, Magnetohydrodynamics (N.Y.) **16**, 21 (1980).
[5] D.J. Keller, J.P. Korb, and H.M. McConnell, J. Phys. Chem. **91**, 6417 (1987).
[6] S.A. Langer, R.E. Goldstein, and D.P. Jackson, Phys. Rev. A **46**, 4894 (1992).
[7] D.P. Jackson, R.E. Goldstein, and A.O. Cebers, Phys. Rev. E **50**, 298 (1994).
[8] J.A. Miranda and M. Widom, Phys. Rev. E **55**, 3758 (1997).
[9] H.M. McConnell, P.A. Rice, and D.J. Benvegnu, J. Phys. Chem. **94**, 8965 (1990).
[10] M. Seul, J. Phys. Chem. **97**, 2941 (1993).
[11] R.E. Goldstein and D.P. Jackson, J. Phys. Chem. **98**, 9626 (1994).
[12] R.E. Goldstein, D.P. Jackson, and A.T. Dorsey, Phys. Rev. Lett. **76**, 3818 (1996).
[13] I. Drikis, J.C. Bacri, and A. Cebers, Magnetohydrodynamics (N.Y.) **35**, 157 (1999).
[14] D.P. Jackson and B. Gantner, Phys. Rev. E **64**, 056230 (2001).
[15] Surface tension does play an *indirect* role in how these domains interact. This is because surface tension is an important factor in determining the number of fingers that evolve on a single magnetic drop.
[16] L.D. Landau, E.M. Lifshitz, and L.P. Pitaevskii, *Electrodynamics of Continuous Media* (Pergamon Press, New York, 1984), pp. 118–119.
[17] D. Bensimon *et al.*, Rev. Mod. Phys. **58**, 977 (1986).