Relaxation of patterns in 2D modulated phases

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Abstract. Competition between repulsive long-range and attractive short-range interactions leads to modulated phases and structure formation on mesoscopic length scales in a variety of different systems. The patterns may be trapped in different metastable configurations, and the experimentally observed pattern generally depends on the history of preparation. In the present paper, we simulate the relaxation of non-equilibrium patterns towards equilibrium. We derive general equations for the driving force of the motion at the phase boundaries and for the corresponding flow field. As an example, we apply these equations to a dipolar long-range potential, as in Langmuir monolayers or ferrofluid films. We simulate the two-fold shape instability of an isolated domain and the formation of labyrinth patterns—unconfined and confined in a circular trap. Our simulations are in qualitative agreement with the experiments.
1. Introduction

Competing interactions between spacially separated elements of a system can cause patterns on characteristic length scales. Technologically, such self-assembled patterning is of special interest for the bottom-up design of devices. Modulated phases are one example for the formation of equilibrium structures on meso- or macroscopic scales arising from competing interactions. Static and dynamic modulated phases are observed in a wide diversity of physical systems [1]—which can be two-dimensional (2D) or 3D. The geometry of patterns is diverse; in general, simple morphologies with some degree of regularity are formed. These include tubes, sheets and spherical droplets in 3D systems, and stripe and droplet patterns in 2D systems. Systems exhibiting modulated phases range from type 1 superconductors [2, 3] via Langmuir monolayers [4, 5], chemical reaction-diffusion systems [6, 7], Rayleigh–Benard instabilities [8, 9] to ferromagnetic garnet films [10] or films of ferrofluids [11, 12]. All of these systems share the fact that their patterns can be understood in terms of competing forces on different length scales [1]. On the one hand, short-range attractions lead to the formation of coexisting phases that phase-separate because the boundary between two phases costs interfacial energy. On the other—due to long-range repulsion—macroscopic phase separation is energetically unfavourable. The patterns formed are therefore a compromise between interfacial energy and the tendency of a phase to be as far apart from itself as possible. Generally, several metastable equilibria exist, and the actual equilibrium achieved, even its morphology, depends on the history of the system. To understand which equilibrium structure is finally established, one has to study the dynamics of the system. This is a demanding task since several dynamic processes, such as phase transitions and shape equilibrations, often take place in parallel. The current work addresses those systems in which one has separation of time scales. We restrict ourselves to systems where shape equilibration is fast compared to the phase equilibration and to those where the topology of the phases does not change. As a result of the different time scales, the area fraction of the phases can be assumed to be constant during the processes studied. We present general equations for the driving forces and the
dissipative dynamics that form a closed system of 1D equations for the phase boundaries. We then specialize our equations to a dipolar long-range interaction encountered in, for example, magnetic garnet films or Langmuir monolayers. As a dissipative mechanism, we simulate patterns that relax towards equilibrium due to hydrodynamic flow dominated by an underlying fluid subphase of infinite depth.

2. General model

2.1. Statics

Assume a 2D system of two coexisting non-interacting phases. The order parameter \( \mu(\mathbf{r}) \) therefore can attain the two equilibrium values \( \mu_1 \) or \( \mu_2 \). We assume the phase boundaries, where the order parameter jumps from \( \mu_1 \) to \( \mu_2 \), to be much smaller in width than the typical size of the patterns, and approximate the boundaries as infinitely sharp with an interfacial energy proportional to its length and characterized by a line tension \( \lambda \). The area fraction between both phases is assumed to be fixed during the shape equilibration, such that we can treat both phases as incompressible. Long-range interactions are expressed explicitly by the pair interaction potential \( \mu_i \mu_j V(|\mathbf{r} - \mathbf{r}'|) \), where \( \mathbf{r} (\mathbf{r}') \) points to phase \( i (j) \). The total free energy of the system is the sum of particular contributions:

\[
F = F_{sr} + F_{lr} + F_0.
\]

where the short-range energy \( F_{sr} = \lambda \int_P ds \) is proportional to the total length of interface \( P \). The 2D bulk energy \( F_0 = \sum_i F_0(A_i) \) depends only on the areas of the phases—not on their shapes. Integration of the long-range interaction over all interacting pairs of area elements yields

\[
F_{lr} = \frac{1}{2} \int_A \int_A \mu(\mathbf{r}) \mu(\mathbf{r}') V(|\mathbf{r} - \mathbf{r}'|) dA dA', \quad = \frac{\mu^2}{2} \int_P \int_P \mathbf{n} \cdot \mathbf{n}' \Phi(|\mathbf{r}(s) - \mathbf{r}(s')|) ds ds' + F_0'.
\]

In the first line of equation (2), the integration is done over the total area \( A \). In the second line, the integration is performed over the domain boundaries parametrized by the arc length \( s (s') \). \( \mathbf{n} \) and \( \mathbf{n}' \) denote the normal vectors to the boundary (pointing into phase 1), and \( \mu = \mu_1 - \mu_2 \). The shape-dependent contributions of the long-range energy depend only on the difference in order parameter, while the shape-independent contribution \( F_0' \) can be included in the bulk energy. The line integral expression of the long-range free energy (equation (2)) is obtained by twice applying Green’s theorem and the potential \( \Phi \) satisfies \(-\nabla^2 \Phi = V\).

2.2. Driving force

If the pattern is not in equilibrium, then forces occur at the phase boundaries. These forces cause the motion of the phase boundaries and the evolution of the system towards local equilibrium. The force density at the phase boundary is given as a functional derivative of the free energy with respect to the shape:

\[
f_{ij}(\mathbf{r}(s)) = \left( \frac{\delta F}{\delta \mathbf{r}(s)} \right)_{A_i, A_j} = (p_{ij} - \lambda \kappa(\mathbf{r}(s)) + p_{li}(\mathbf{r})) \mathbf{n}(\mathbf{r}).
\]
The force density acts perpendicular to the interface and can be understood as a result of a pressure difference across the interface; \( p_{ij} \) denotes the difference of the static pressures (constant for each domain), \( \kappa(\mathbf{r}(s)) \) is the curvature of the phase boundary, and \( p_{\text{lr}}(\mathbf{r}) \) is the pressure caused by the long-range interaction:

\[
p_{\text{lr}}(\mathbf{r}) = -\mu^2 \int_{\text{phase}_2} V(|\mathbf{r} - \mathbf{r}'|) \, dA, \quad = -\mu^2 \int_{p} \mathbf{n}' \cdot \nabla \Phi(|\mathbf{r} - \mathbf{r}'(s')|) \, ds'.
\] (4)

In equilibrium, the force density vanishes and equation (3) represents a generalized 2D Young–Laplace equation for the equilibrium shape.

2.3. Relaxation mechanism

The forces discussed in section 2.2 will cause the domain boundaries to move with velocity \( \mathbf{u} \) that is proportional to the force:

\[
\mathbf{u}(\mathbf{r}) = \int O(\mathbf{r}, \mathbf{r}') \cdot \mathbf{f}(s') \, ds',
\] (5)

The physics of the dissipation mechanism is expressed in the particular form of the non-local second rank tensor \( O \). In general, \( O \) will also depend on the instantaneous non-equilibrium pattern formed by the two phases and therefore changes with time and space as the pattern evolves. However, if the dissipation mechanisms of phases 1 and 2 are the same, then \( O \) will depend on the separation \( \mathbf{r} - \mathbf{r}' \) only. Such a situation arises, for example, for magnetic garnet films (due to symmetry reasons) or for Langmuir monolayers—if the surface shear viscosities of both phases are equal or negligible compared to the product of the subphase viscosity and the length scale of the pattern. \( O \) assumes an entirely local form, \( O \propto \delta(\mathbf{r} - \mathbf{r}') \mathbf{1} \), for magnetic garnet films and Langmuir monolayers on a shallow water subphase. On the other hand, for highly viscous Langmuir monolayers, \( O \)—in the context of hydrodynamics termed Oseen tensor—accounts for long-range logarithmic hydrodynamic interactions. If the monolayer viscosities are negligible compared to the viscous dissipation in the subphase, the Oseen tensor transmits only intermediate-range hydrodynamic interactions. Explicit expressions for \( O \) exist for special geometries and relations between surface and bulk dissipative mechanisms. For these cases, equations (3)–(5) supply a closed system of first-order integro-differential equations for the time evolution of the phase boundaries. In the rest of this work, we will numerically solve some special cases for equations (3)–(5) and qualitatively compare them with experiments on Langmuir monolayers and magnetic garnet films.

3. Simulation

For our simulations, we consider a dipolar pair interaction between electric dipoles (in Langmuir monolayers) or between nanomagnets (in ferrofluid films or magnetic garnet films):

\[
V_{\text{dip}} = \frac{1}{\rho^3}, \quad \text{with} \quad \rho = \sqrt{(\mathbf{r} - \mathbf{r}')^2 + \Delta^2}.
\] (6)

We use the line integral expression of the long-range interaction in equation (4) to reduce the computation time of the numerical integration. Inserting the dipole potential (6) into (4), the line integral expression of the dipole pressure reads [13]

\[
p_{\text{dip}}(\mathbf{r}) = -\mu^2 \int_{p} \frac{\mathbf{n} \cdot (\mathbf{r} - \mathbf{r}')}{\Delta(\rho + \Delta) \rho} \, ds.
\] (7)
The parameter $\Delta$ prevents the energy (2) and pressure (7) from diverging. Its physical meaning is that of the film thickness in ferrofluids [11, 14] and the thickness of the phase boundaries in the case of magnetic garnet films and Langmuir monolayers [13]. In the numerical treatment, $\Delta$ can be chosen arbitrarily as long as it is smaller than the radius of curvature of the patterns. The entire dynamic is invariant under the transformation [15]–[17]:

$$(\lambda, p_{ij}, \Delta) \rightarrow (\lambda^*, p_{ij}^*, \Delta^*)$$

with

$$\lambda^* = \lambda - \mu^2 \ln \frac{\Delta^*}{\Delta} \quad \text{and} \quad p_{ij}^* = p_{ij} - \frac{\pi}{\Delta} + \frac{\pi}{\Delta^*}.$$  

In the numerics, we chose $\Delta$ slightly larger than our integration step width, but smaller than the radius of curvature of the patterns.

For the simulation, we use the Oseen tensor of a Langmuir monolayer of vanishing surface viscosity on a subphase with the viscosity $\eta$ and infinite depth

$$O(\mathbf{r}(s), \mathbf{r}(s')) = \frac{(\mathbf{r}(s) - \mathbf{r}(s'))(\mathbf{r}(s) - \mathbf{r}(s'))}{2\pi\eta|\mathbf{r}(s) - \mathbf{r}(s')|^3} + \delta(s - s') \frac{1}{2\pi\eta},$$

where $\delta(s - s')$ denotes the Dirac delta function and $\mathbf{1}$ is the unit tensor in two dimensions. Experiments in the liquid-expanded phase and gaseous phase of the Langmuir monolayers [18]–[21] show that the surface shear viscosities of both phases can indeed be neglected. Analytical expressions for the Oseen tensor for higher surface shear viscosities and simple geometries have been supplied by Fischer [22].

3.1. Numerics

The domain boundaries were discretized by $n = 1, \ldots, 1000$ sampling points $\mathbf{r}_n$ per domain. The force density and velocity density were calculated using equations (3), (7) and (5). The time was discretized such that the sampling points in the next time step $t + \Delta t$ are given by

$$\mathbf{r}_n(t + \Delta t) = \mathbf{r}_n(t) + \mathbf{u}(\mathbf{r}_n) \Delta t.$$  

(10)

After each time step, the sampling points were additionally moved in a tangential direction equalizing the distance between neighbouring sampling points to ensure the homogeneity of the boundary discretization over the whole simulated time period.

The long-range pressure was calculated using (7). The numerical length scale $\Delta^*$ was chosen such that it fulfils

$$|\mathbf{r}_{n+1} - \mathbf{r}_n|_{\text{max}} \ll \Delta^* \ll \frac{1}{\kappa_{\text{max}}}.$$  

(11)

Theoretically, equation (5) conserves the area of each domain independent of the static pressure $p_{ij}$. However, due to the discretization, numerical errors occur and it is necessary to introduce a stabilizing numerical compressibility $k$. The static pressure is expressed by the equation

$$p_{ij} = p_0 \left(1 + \frac{A_0 - A}{A_0} k\right).$$  

(12)

where $A$ is the actual area of a domain and $A_0$ the initial area. The parameter $k$ is chosen in such a way that $A$ deviates from $A_0$ by less than 5% during the whole simulation.
Figure 1. Circle to dog-bone shape transition. (a)–(d) (and movie 1a) Experimental observation of the transition in a magnetized bismuth-substituted ferrite garnet film (Y$_{2.4}$Bi$_{0.6}$Fe$_4$GaO$_{12}$) grown by liquid-phase epitaxy on a (111) surface subject to a jump in the external magnetic field $H \approx 2500$ A m$^{-1}$, $\Delta H/H \approx 10\%$. (a) $t = 0$, (b) $t = 120$ ms, (c) $t = 180$ ms and (d) $t = 320$ ms. Scale bar, 4 $\mu$m. (e)–(h) (and movie 1b) Simulation. The numerical parameters for the simulation are: $\mu = 1$, $\Delta_{\text{dipole}} = 20$ pixels and $A = 40115$ pixels$^2$ (for circular shape, $R = 113$ pixels). The transition occurs at $\lambda = 1.6$ as shown in (f)–(h), whereas at larger line tensions the domain remains circular as in (e) ($\lambda = 1.61$). The theoretical critical radius (13) for this parameter is $R_2 = 128$ pixels—about 10% larger than that for the simulation.

4. Results

4.1. Circle to dog-bone shape transition

Firstly, we present the simulation of a rather simple geometry—the transition from a circular domain to an elongated dog-bone. If the radius of a circular domain exceeds a critical size and the dipolar repulsion overcomes the line tension, then the domain becomes instable with respect to thermal fluctuations and shape transitions occur. At the threshold, the interface line elongates to lower the dipolar energy of the droplet. For a two-fold deformation, the critical radius is given by the equation [23]

$$R_2 > \frac{\Delta_{\text{dipole}}}{8} \exp \left( \frac{\lambda}{\mu^2} + \frac{7}{3} \right).$$

(13)

A general solution for $n$-fold deformations is given by Deutch and Low [24] and Khattari and Fischer [25]. Figures 1(a)–(d) (and movie 1a) show a circle to dog-bone shape transition in a magnetized bismuth-substituted ferrite garnet film (Y$_{2.4}$Bi$_{0.6}$Fe$_4$GaO$_{12}$) grown by liquid-phase epitaxy on a (111) surface of an approximately 0.5 mm-thick gadolinium gallium garnet (GGG) substrate. The anisotropy and exchange energy are not important for driving such transitions (as opposed to the creation of phases and domain walls, for example, where these energies play a major role). Figure 1(e)–(h) (and movie 1b) show our corresponding simulations. Our simulations reproduce the threshold value (13) within 10%, giving an estimate of the accuracy of the numerics.
Figure 2. Labyrinth pattern observed in (a) a magnetized bismuth-substituted ferrite garnet film \( (Y_{2.4}Bi_{0.6}Fe_4GaO_{12}) \) grown by liquid-phase epitaxy on a (111) surface, and (b) in a Langmuir monolayer of methyl-octadecanoate at three-phase coexistence. The liquid-expanded phase (bright) and gaseous phase (black) form a labyrinth within a liquid-condensed matrix phase. The average thickness of the stripes is (a) 8.3 \( \mu \)m and (b) 3.3 \( \mu \)m.

Figure 3. Simulated formation of a labyrinth pattern. The boundary conditions are periodic with hexagonal symmetry—lattice constant \( a = 500 \) pixels (\( \approx 100 \) \( \mu \)m), total size of one image: \( 640 \times 480 \) pixels\(^2\) (130 \( \times \) 97 \( \mu \)m). The numerical parameters are: \( \Delta = 20 \) pixels (1.5 \( \mu \)m), \( \lambda = 0.9 \) (0.05 pN), \( \mu = 1 \) (0.24 pN) and \( \eta = 10^{-6} \) (10\(^{-3}\) Pa s). The area fraction is about 50%. The initial shape (a) and the structure (b) after 100 time steps (\( \approx \) 0.14 s), (c) after 500 time steps (\( \approx \) 0.7 s) and (d) after 25 000 time steps (\( \approx \) 35 s) are shown. The structure in (d) is very close to equilibrium and the motion has almost stopped. The average stripe width is \( \approx 47 \) pixels (3.3 \( \mu \)m). The entire dynamic simulation can be viewed in movie 3.

4.2. Labyrinths

Stripe patterns, or labyrinths, are observed in Langmuir monolayers, ferrofluids and garnet films (figure 2). Kwok and Vanderbilt showed that, for almost all area fractions, stripe phases minimize the total free energy of the system. However, which morphology is observed in a particular experiment strongly depends on the history of the system—which may be trapped in various metastable equilibria above the ground state [26].

We simulated the formation of a labyrinth out of an arbitrary initial structure. Figure 3 (and movie 3) shows the time evolution of a periodic non-equilibrium two-phase pattern to a labyrinth.
Figure 4. Fluorescence microscopy images of a liquid-expanded droplet (bright) in a gaseous cavity (dark) confined in a liquid-condensed matrix (grey) of a methyl-octadecanoate Langmuir monolayer at 24 °C (reproduced with permission from [25]). The shape deformation of the liquid expanded (LE) droplet occurs upon compression of the monolayer. The dynamics of the shape transformation can be seen in more detail in movie 4.

The boundary conditions are periodic with a hexagonal symmetry. After ≈35 s simulated time (>25 000 time steps), the pattern is close to equilibrium. Comparing the average stripe width with figure 2(b) (3.3 µm), the numerical parameters correspond to $\lambda = 0.05$ pN and $\Delta = 1.5$ µm, while $\mu^2 = 0.06$ pN was measured previously [27].

4.3. Confined droplets

As the last scenario we chose a droplet confined in a circle formed by an embedding solid surrounding of order parameter $\mu_3$. Figure 4 (and movie 4) shows the shape instability in the three-phase coexistence region of a methyl-octadecanoate Langmuir monolayer observed by Khattari and Fischer [25]. The liquid-expanded droplet (bright) confined within a gas bubble (black) immersed in liquid-condensed phase (grey) becomes unstable upon compression. Khattari and Fischer [25] found analytically that the confinement should stabilize the droplet and that the instability can only be understood in terms of non-constant materials parameters.

For this geometry, the electrostatic pressure has an additional contribution from the LC region

$$p_{LC}(r) = -\mu \mu_{LC} \int_{p_{LC}} \frac{n \cdot (r - r')}{\Delta(\rho + \Delta)\rho} ds. \tag{14}$$

We performed several simulations at constant dipole densities ($\mu$ and $\mu_{LC}$ were set to the values measured by Heinig et al [27]) but at varying line tension $\lambda$. All simulations start with the initial shape (figure 5, $t = 0$) in the upper left corner of figure 4. Figure 5(b) ($t = 10$ s, 20 s and 3 min) and movie 5b show that the confined droplet keeps its circular shape for situations in which it would deform when unconfined (figure 5(a) and movie 5a). A decrease of the line tension destabilizes the confined droplet and leads to the formation of a labyrinth pattern shown in figures 5(c) and (d) and movie 5c and movie 5d. The length scale of the patterns (i.e. the stripe width) is a characteristic quantity in each figure and depends on the line tension. The length scale decreases from 4 µm in figure 5(c) to 3 µm in figure 5(d). The experimental patterns in
The dipole densities for methyl-octadecanoate Langmuir monolayer phases measured by Heinig et al [27] were used for the simulation: $\mu^2 = 0.06 \text{ pN}$ and $\mu^2_{\text{LC/G}} = 0.2 \text{ pN}$; $t = 0$, initial shape directly taken from the experimental image. (a) (and movie 5a) Relaxation of unconfined droplet for $\lambda_5 = 0.431 \text{ pN}$. (For a better comparison to the line tension values in literature we rescaled $\lambda$ to $\Delta = 5 \text{ Å}$. ) The circle remains on the position of the LC interface. (b) (and movie 5b) Time evolution of the confined droplet with the same parameters. As described by Khattari and Fischer, the confinement stabilizes the droplet. (c) (and movie 5c) $\lambda_5 = 0.42 \text{ pN}$; equilibrium stripe thickness, $\approx 4 \mu\text{m}$. (d) (and movie 5d) $\lambda_5 = 0.416 \text{ pN}$; equilibrium stripe thickness, $\approx 3 \mu\text{m}$. Only a decrease in the line tension during the experiment in figure 3 could have caused the observed structures.

Figure 5. Simulation of the time evolution of the initial shape in figure 4 (upper left corner) for different line tensions. The width of one image is $\approx 56 \mu\text{m}$. figure 4 can be explained within the model by a systematic decrease of the line tension upon compression. Finally, in the lower right corner of figure 4, the line tension completely vanishes and the interface line finalizes its existence. This behaviour might be explained as a mixing transition of methyl-octadecanoate with the fluorescent label or another unknown impurity. Close to the critical point, the line tension vanishes and the length scale on which deformations occur approaches zero. Mixing transitions of lipid monolayers and cholesterol were investigated by Seul [28] and Radhakrishnan and McConnell [29]. The structures they observed show a similar behaviour close to the critical point.

5. Summary

An equation for the driving force density (3) acting on domain boundaries in non-equilibrium modulated phases is derived for the case where the phase transition kinetics are fast compared to
the shape equilibration. The force density depends on the long-range pair interaction potentials and the short-range interactions which are all represented by line tensions. The force acts perpendicular to the phase boundaries and transforms to a generalized Young–Laplace equation at equilibrium. The dissipative time evolution is described by a second rank tensor linearly relating the domain boundary velocity to the force (equation (5)). The tensor is local for relaxation dominated by the friction of the domain wall and non-local for a Stokes flow dominated by viscosities. We have simulated the time evolution of non-equilibrium patterns towards equilibrium for the special dissipation mechanism of a non-viscous Langmuir monolayer on a deep subphase interacting through long-range dipolar interactions. The simulated patterns qualitatively agree with those studied in the experiments.

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References

[1] Seul M and Andelman D 1995 Science 267 476
[2] Huebener R P 1979 Magnetic Flux Structures in Superconductors (Berlin: Springer)
[3] Jedy V and Gourdon C 2004 Phys. Rev. Lett. 92 147001
[4] McConnell H M 1991 Annu. Rev. Phys. Chem. 42 171
[5] Kaganer V M, Möhwald H and Dutta P 1999 Rev. Mod. Phys. 71 779
[6] Castets V, Dulos E, Boissonade J and De Kepper P 1990 Phys. Rev. Lett. 64 2953
[7] Quyang Q and Swinney H L 1991 Nature 352 610
[8] Behringer R P 1985 Rev. Mod. Phys. 57 657
[9] Cross M C and Hohenberg P C 1993 Rev. Mod. Phys. 65 851
[10] Seul M and Wolfe R 1992 Phys. Rev. Lett. 68 2460
[11] Wang H, Zhu Y, Boyd C, Luo W, Cebers A and Rosensweig R E 1994 Phys. Rev. Lett. 72 1929
[12] Liu J, Wu A, Ivey M L, Flores G A, Javier K, Bibette J and Richard J 1995 Phys. Rev. Lett. 74 2828
[13] Heinig P, Wurltizer S, John Th and Fischer Th M 2002 J. Phys. Chem. 106 11951
[14] Richardi J, Ingent D and Pileni M P 2002 Phys. Rev. E 66 046306
[15] Riviére S, Henon S, Meunier J, Albrecht G, Boissonade M M and Baszkin A 1995 Phys. Rev. Lett. 75 2506
[16] Heinig P, Steffen P, Wurlitzer S and Fischer Th M 2001 Langmuir 17 6633
[17] de Koker R and McConnell H M 1993 J. Phys. Chem. 97 13419
[18] Heckl W M, Miller A and Möhwald H 1988 Thin Solid Films 159 125
[19] Schwartz D, Knobler C M and Bruinsma R 1994 Phys. Rev. Lett. 73 2841
[20] Klingler J F and McConnell H M 1993 J. Phys. Chem. 97 6096
[21] Steffen P, Heinig P, Wurlitzer S, Khattari Z and Fischer Th M 2001 J. Chem. Phys. 115 994
[22] Fischer Th M 2004 J. Fluid Mech. 498 123
[23] McConnell H M and Moy V T 1988 J. Phys. Chem. 92 4520
[24] Deutch F M and Low F E 1992 J. Phys. Chem. 96 7097
[25] Khattari Z and Fischer Th M 2002 J. Phys. Chem. B 106 1677
[26] de Koker R and McConnell H M 1996 J. Phys. Chem. 100 7722
[27] Heinig P, Wurlitzer S, Steffen P, Kremer F and Fischer Th M 2000 Langmuir 16 10254
[28] Seul M 1990 Physica A 168 198
[29] Radhakrishnan A and McConnell H M 1999 Biophys. J. 77 1507
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The authors regret not having cited the following papers on labyrinthine patterns in magnetic fluids.

References

Cebers A and Maiorov M M 1980 Magnitnaya Gidrodinamika 1 27
Cebers A and Drikis I 1996 Magnitnaya Gidrodinamika 1 11
Dikris I, Bacri J-C and Cebers A 1999 Magnitnaya Gidrodinamika 3 203