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Macro-Organized Patterns in Ferrofluid Layer: Experimental Studies

Florence Elias (1,2), Cyrille Flament (1,2,*), Jean-Claude Bacri (1,2) and S. Neveu (3)

(1) Université Paris 7 - Denis Diderot, Unité de Formation et de Recherche de Physique, case 70.08, 2 place Jussieu, 75251 Paris Cedex 05, France
(2) Laboratoire d’Acoustique et d’Optique de la Matière Condensée, case 78 (**), 4 place Jussieu, 75252 Paris Cedex 05, France
(3) Laboratoire de Physico-Chimie Inorganique, case 63 (**), 4 place Jussieu, 75252 Paris Cedex 05, France

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Abstract. — We present some experimental results concerning the conformational instabilities observed in a thin layer made of an immiscible mixture of a magnetic fluid and another liquid submitted to an external magnetic field. We observed macroscopic domains with stripe or bubble shape. The transition between the two morphologies is observed and a coexistence of the two phases is visualized in a variable thickness layer. Furthermore, we give other results about the instabilities of the parallel stripe pattern: by increasing the external field the stripe pattern becomes a chevron structure, which also becomes unstable for higher values of the magnetic field and gives a hairpin pattern. Finally, we present a new system: a magnetic liquid froth in two dimensions which is analogous to soap froths with an additional external control parameter, the magnetic field.

1. Introduction

1.1. Dipolar Systems and Modulated Phases. — Thin layers of dipolar systems show very similar patterns where the same morphologies are present: bubbles and stripes. The stripes can be parallel or intricated in a labyrinthine-like structure. A first example where stripes and bubbles are observed are the garnet films or the bubble materials. At low temperature and for zero external field, stripe domains of opposite magnetization pre-exist. If a magnetic field is applied one can observe for a given temperature below the Curie point, the transition from the stripe phase to the bubble phase and for higher field the transition towards a uniform magnetized phase [1]. Another interesting system in which many morphologies are present is the Langmuir films. A Langmuir film is a monolayer at the air-water interface built

(*) Author for correspondence (e-mail: flament@omec.jussieu.fr)
(**) URA 800 associée au CNRS et à l'Université Paris 6
(***) URA associée au CNRS et à l'Université Paris 6

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with amphiphilic molecules [2, 3]. Each molecule carries an electric dipole; this permanent dipole can be perpendicular to the layer or tilted. First order transitions between liquid phase and condensed liquid phases (which are mesomorphic phases) or between two-dimensional gas phase and liquid phase are observed below the critical temperature. The coexistence of domains of the condensed phase in the expanded phase is a signature of the order of the transition. By increasing the surface pressure at constant temperature one can observe shape transitions of the domains.

In all these cases, the morphologies of the domains have been attributed to competition between two antagonistic energies: the surface energy tends to minimize the domain interface whereas the repulsive long range dipole-dipole interaction favours an extended interface. Since the surface density \( \Phi(x, y) \) of the system is non-homogeneous and periodic in space, one call this pattern a modulated phase. For instance the order parameter of the stripe system can be written in a single-mode analysis [1]:

\[
\Phi(x, y) = \Phi_0 [1 + \cos(qx)],
\]

where \( x \) is the direction perpendicular to the stripes and the spatial average \( \langle \Phi(x, y) \rangle = \Phi_0 \). For the hexagonal bubble lattice, one has:

\[
\Phi(x, y) = \Phi_0 \left[ 1 + \sum_{i=1}^{3} \cos(q_i \cdot r) \right] \text{ with } \sum_{i=1}^{3} q_i = 0 \text{ and } |q_i| = q.
\]

A recent and complete review about the modulated phases has been given by Seul and Andelman [4]. The authors list a number of various chemical and physical systems where ubiquitous stripes and bubbles are present, and they give a phenomenological description of these patterns.

The morphological transition from the stripes to the bubbles has been studied for the different systems. In the Langmuir films, the theoretical pioneer work of Andelman et al. [5] has been theoretically and experimentally continued by McConnell [6], and extended to more complicate domain shapes (e.g. transition from circular domain to dog-bone shape, or circular domain to ring shape) [7–10]. We can also refer to the works of Seul [11, 12] and Hasley [13]. Sornette has established the analogy between the magnetic striped phase and the lyotropic lamellar phase [14, 15]. All these studies concern equilibrium shapes, a recent paper of McConnell et al. [16] treats of the deformations of these equilibrium shapes. Several other authors had already studied instabilities of the stripe pattern in the garnet film system [17–20] or in smectic liquid-crystal [21]. A bending instability of the parallel stripes occurs leading to chevron pattern [22] for higher external strain. The creation of disorder in the stripe phase, pointed by Seul et al. [19], involves the nucleation and the unbinding of disclination pairs.

1.2. Thin Layer of Ferrofluid. — Our system consists in a mixture of a magnetic fluid (or MF, also called ferrofluid) and another immiscible liquid (typically an oil). The mixture is confined into a quasi-bidimensional geometry. Let us define the control parameters of this system: \( H_{\text{ext}} \) is the strength of the homogeneous magnetic field which is perpendicular to the layer, \( \Phi \) is the volume fraction of the MF, and \( h \) the thickness of the layer. Since the parameter \( h \) is small compared to the lateral extension of the layer, we have made the basic assumption that the system is invariant in the field direction. The layer is confined between two horizontal flat walls in order to build a Hele-Shaw system without gravity influence. Here we focus on static instabilities i.e. spatial conformation of the MF with the other immiscible liquid. We observe stripes or bubbles on a centimetric scale, while domains in other systems are mesoscopic.
Garnet films or Langmuir monolayers have a main feature in common: the average value of the order parameter is a non-conserved quantity and varies with the external strain like an internal variable. For the first one, the magnetization can vary continuously by a flip of the magnetization of a domain. For the Langmuir monolayers, the density of the fluid or the average area per molecule can vary continuously on the plateau coexistence by transformation of one phase into the other one. Our system is a little different since the average surface fraction is conserved \((\Phi(x, y)) = \Phi_0\) for any value of the applied field \(H_{\text{ext}}\) (as a matter of fact it is impossible to transform a liquid into the other one).

In a particular case it is possible to obtain a MF system similar to the first ones by using a MF prepared near the demixtion point. We obtain bubbles of a concentrated MF phase in a matrix of a diluted MF phase. This phase separation can be induced by altering the colloidal stability (by increasing the ionic strength, or by decreasing the temperature of the solution). In this case the typical size of the domains is micrometric as a consequence of the very low value of the surface tension between the both phases [23].

Finally we present a new system: the magnetic liquid froth. It is obtained with a perpendicular alternating magnetic field. In this case an instability occurs which consists in the nucleation of hole in a MF area. Since we obtain bubbles of oil in a MF matrix, we get the dual situation of the MF bubble phase. We also show experimentally that it is possible to get a transition from the froth phase to the bubble phase.

1.3. EXPERIMENTAL SET-UP. — A MF is a colloidal suspension of magnetic particles. Here we use an ionic MF (water based) with cobalt ferrite particles \((\text{CoFe}_2\text{O}_4)\) of mean size roughly 10 nm [24]. The magnetic response of the MF is given by a Langevin type law

\[
M = M_s L(\xi) = M_s [\text{coth}(\xi) - \xi^{-1}]; \quad \text{with} \quad \xi = \frac{\mu_0 \mu H_{\text{ext}}}{k_B T},
\]

\(\mu\) is the magnetic momentum of a particle, and the MF saturation magnetization is equal to \(M_s = 40 \text{ kA m}^{-1}\)

In the Hele-Shaw cell, the MF is in contact with an immiscible organic liquid in order to avoid wetting phenomena: the MF does not wet the cell because a microscopic thin film of oil always exists between the MF and the walls. The value of the surface tension, \(\sigma\), between the two liquids is obtained \textit{in situ} by measuring the critical wavelength at the threshold of the peak instability, or by measuring the deformation of a MF droplet induced by an external magnetic field [25]. We obtain \(\sigma \approx 10 \text{ mN m}^{-1}\) at room temperature; we proved that this quantity is independent of the magnetic field [25]. The density of the liquids is \(\rho_{\text{MF}} \approx 1600 \text{ kg m}^{-3}\) and \(\rho_{\text{oil}} \approx 800 \text{ kg m}^{-3}\)

We use two kinds of Hele-Shaw cell, the simplest one has two parallel plates separated by a fixed distance, \(h\). The thickness is a millimetric quantity, \(h = 0.7 \text{ mm}\), while the length of the cell is on a centimetric scale \(L \approx 10 \text{ cm}\). Another new experimental apparatus is obtained using two planar walls with a variable thickness: \(h = 0.5\) to 5 mm on the total length \(L\). This new type of cell allows the observation of a morphological transition which is discussed in the following chapter.

A cell is located between two Helmholtz coils as sketched in Figure 1. An image processing is required to get the local two-dimensional density \(\Phi(x, y)\) of the mixture. The images are then recorded by a CCD (Couple Charge Device) camera and digitized in a computer.
2. Stripe and Bubble Domains

The variable thickness layer permits the measurements of the geometrical variables of the domains as a function of $h$ (for a fixed value of the magnetic field) with a single experiment. Starting from a metastable situation where only intricated stripes are present, the system is perturbed by the approach of a little magnet near the cell, such as to break the metastable stripes. Therefore the final state is more stable than before the perturbation, but it can be still a metastable configuration (in fact, we do not know precisely how to reach the lowest energy state). In all the experiments, the same perturbation is used to obtain the final state, which appears to be the more stable. Figure 2 shows such an experiment for a volume fraction of MF $\Phi_0 = 0.45$. Domains with bubbles shapes are visible in the higher thickness, and intricated stripes are present in the lower thickness.

2.1. Intricated Stripe Phase. — An experiment performed with $\Phi_0 = 0.40$ at high field $H_{\text{ext}} = 33 \text{ kA m}^{-1}$ gives intricated stripes of variable width as shown in Figure 3a. Let us consider the intricated stripes as infinite parallel stripes; this simplification allows the calculation to be more convenient. The computation is performed considering a fixed thickness $h$ (see Fig. 4a). In this case, $\Phi$ is also the surface fraction of the MF. In a periodic lattice, it is related to the width of a stripe, $d$, by the ratio: $\Phi = d/l$, where $l$ is the distance between two stripes. The passage to experimental data will be improved considering that the variable thickness cell is a juxtaposition of fixed thickness cells, each one filled with a volume fraction $\Phi$ of the MF. This is justified by the fact that in the variable thickness cell, $\Phi$ does not depend on $h$, except at the edges. An energetic approach gives the internal variable $d$ (or $l$ since $\Phi$ is fixed) as a function of the control parameters: $H_{\text{ext}}$, $\Phi$, and $h$. The equilibrium state of the system is the result of the competition of two antagonistic phenomena: surface energy leads to minimal interface between MF and oil, whereas magnetic dipolar-dipolar repulsions favour an extended interface. The interfacial energy, $E_s$, of one stripe is:

$$E_s = 2\sigma L(h + d),$$  \hspace{1cm} (4)
Fig. 2. — Photograph of the coexistence of the two phases in the variable thickness layer for $\Phi = 0.45$ and high value of the magnetic field. The thickness, $h$, varies from 0.5 to 5 mm. The MF appears in black. Bubble shape domains are present in the higher thickness, while intricated stripes are visible in the lower thickness.

where $L$ is the length of the MF stripe. A more realistic expression has to infer two surface tensions between the oil and the magnetic fluid: $\sigma_1$ for the interfaces perpendicular to the layer, and at the top and the bottom of the cell, $\sigma_2$, in order to take into account the disjoining pressure due to the interaction of the closer interfaces cell-oil and oil-MF. In this case, we would have had $E_s = 2\sigma_1 Lh + 2\sigma_2 Ld$. Since it is impossible to measure directly $\sigma_2$, we take $\sigma_2 = \sigma_1 = \sigma$. The magnetic energy includes two contributions: the interaction between the dipoles and the external field, and a demagnetizing effect taking account the interactions between dipoles. As usually, we reduce to dipolar pairs interaction, and we calculate the demagnetizing factor for infinite stripes (i.e. $L \to \infty$). The magnetic energy, $E_m$, of one stripe can be written [26]:

$$E_m = -\mu_0 \int_{\text{stripe}} d^3r \mathbf{M} \cdot \mathbf{H}_{\text{ext}}.$$  \hspace{1cm} (5)

The magnetization, $\mathbf{M}$, is supposed to be parallel to the external field and includes the demagnetizing field, $\mathbf{H}_d$: $\mathbf{M} = \chi (\mathbf{H}_{\text{ext}} + \mathbf{H}_d)$, where $\chi$ is the magnetic susceptibility. We have measured $\chi(\mathbf{H}_{\text{ext}}) = 1.4$. Let us introduce the demagnetizing factor, $D$, defined by the relation $H_d = -DM$, where $D = D(\mathbf{r})$. We made the strong hypothesis that $D$ is constant and equals to its value at the centre of the stripe, $D = D(0)$; this assumption has been already improved in another calculation [25] and seems to be reasonable since it gives good results. In this case $E_m$ is written:

$$E_m = -\mu_0 \int_{\text{stripe}} \frac{\chi H_{\text{ext}}^2}{1 + \chi D} V,$$  \hspace{1cm} (6)

where $V = Lhd$ is the volume of one stripe. The total energy of one stripe is $E_{\text{stripe}} = E_s + E_m$. The dipole-dipole interaction inside a MF stripe is accounted by the demagnetizing
Fig. 3. — a) Photograph of a stripe pattern in the variable thickness cell, \( h: 0.5 \rightarrow 5 \text{ mm} \), with \( \Phi = 0.40 \) and \( H_{\text{ext}} = 33 \text{ kA m}^{-1} \). The more the cell thickness is high, the more the stripe width is high. b) Photograph of a bubble phase for \( \Phi = 0.20, H_{\text{ext}} = 13.2 \text{ kA m}^{-1} \), and \( h: 1.2 \rightarrow 5 \text{ mm} \). The more the cell thickness is high, the more the bubble diameter is high.

field, \( H_{d0} \), [26]:

\[
H_{d0}(r) = -\frac{1}{4\pi} \int_{\text{stripe}} \frac{\Delta^3 r'}{|r - r'|^3} \left( \mu - 3(\mu \cdot e_r)e_r \right),
\]

where \( \mu \) is the magnetic momentum of a particle, \( n(r) \) is the volume density of magnetic particles in the MF, \( e_r = \frac{r - r'}{|r - r'|} \) and \( \mu n = M \) is the MF magnetization. With a homogeneous
Fig. 4. — Notations used for the calculation of the geometrical variables, in the case of a periodic MF stripes system (a), and of a hexagonal lattice of MF bubbles (b). The computations are performed considering a fixed value of the cell thickness $h$.

Magnetization parallel to $H_{\text{ext}}$, we get in the centre of the stripe:

$$D_0 = \frac{1}{4\pi} \int dx \int dy \int dz \left( \frac{1}{(x^2 + y^2 + z^2)^{3/2}} - \frac{3z^2}{(x^2 + y^2 + z^2)^{5/2}} \right),$$

where the integration is carried from $-d/2$ to $d/2$ in the $x$-axis, from $-L \rightarrow -\infty$ to $L \rightarrow \infty$ in the $y$-axis, and from $-h/2$ to $h/2$ in the $z$-axis (see Fig. 4a). First, integrating by parts along the $z$-axis the second term of the integrant, we obtain the following formula:

$$D_0 = \frac{h}{4\pi} \int_{d/2}^{-d/2} dx \int_{-\infty}^{+\infty} dy \frac{1}{(x^2 + y^2 + h^2/4)^{3/2}}.$$

This leads to the final result: $D_0 = \frac{2}{3} \arctan(x)$, with $x = d/h$.

We have also to take into account two other ingredients: the interaction between stripes and the fact that the total amount of magnetic fluid is fixed, a contrario of the Langmuir monolayer and the garnet film experiments. More precisely the average of the surface fraction is fixed $\langle \Phi \rangle = \Phi_0$, but the local variable $\Phi(x, y)$ is an internal variable of the system. First, we evaluate the interactions between stripes by considering the pair interaction between one-dimensional stripes with an uniform dipolar moment (parallel to the external field) whose total value for one stripe is $M = MV = MLdh$; the system is now a two-dimensional pattern of stripes, sketched in Figure 4a, where one stripe is considered as a line of dipoles. This simpler analysis of the interactions between the domains is justified by the fact that the distance between two bubbles is equals to the one between two stripes (for a given value of the control parameters), so the amplitude of the interactions between domains does not depend of their particular shape. The demagnetizing field, $H_{d1}$, induced by the stripe-stripe interaction is expressed similarly as below:

$$H_{d1}(r) = -\frac{1}{4\pi} \int_{\text{total cell}} d^2r' \frac{1}{|r - r'|^3} \mathcal{M}n(r'),$$

where $n$ is now the two-dimensional stripe density: $n(r) = 1/L \sum_i \delta(x - il)$, where $x$ is the perpendicular direction to the stripes. The demagnetizing factor, $D_1$, at the centre of the system in the limit of infinite stripes is given, after straightforward calculation, by:

$$D_1 = \frac{dh}{2\pi l^2} \sum_i \frac{1}{l^2}.$$
The summation is given elsewhere [13]: \( \sum \text{stripes} \frac{1}{i^2} \approx 3.289 \). We have now to introduce that the average surface fraction is imposed, i.e. \( d/l = \Phi_0 \). We get the following result:

\[
D_1 = \frac{(3.289/2\pi)\Phi_0^2}{x}
\]

where the internal dimensionless variable equals \( x = d/h \). Using the dimensionless magnetic Bond number, \( N_B = \mu_0 H_{\text{ext}}^2 h/2\sigma \), the total energy of a stripe interacting with other stripes in a confined system, is:

\[
\frac{E_{\text{stripe}}(x)}{\sigma L h} = 2(1 + x) - \frac{\chi N_B x}{1 + \chi \left( \frac{3}{\pi} \arctan(x) + \frac{a}{x} \right)}
\]

where \( \alpha = 3.289\Phi_0^2/2\pi \) is the coupling stripe interaction parameter. A minimization of the stripe energy with respect to \( x \) gives the equilibrium magnetic Bond number, \( N_B^0 \) as a function of \( x_0 = d_0/h \), the equilibrium dimensionless width of the stripe:

\[
N_B^0(x_0) = \frac{2(1 + x_0^2)/\chi}{1 - \frac{\chi x_0}{1 + \chi x_0} \left( \frac{dD}{dx} \right)_{x=x_0}}
\]

where \( D = D_0 + D_1(\Phi) \).

The formula (13) gives implicitly the dependence of the stripes width \( d \) with respect to \( h \). Figure 5a shows the best interpolation calculated of \( d \) versus \( h \) obtained with a value of the stripe coupling parameter, \( \alpha \), equals to \( \alpha \approx 30 \), instead of the predicted one: 0.1. It means that the calculation of the stripe-stripe interaction has to be improved. Nevertheless, the comparison between calculations and measurements is not so bad, we have reported in Figure 6a the calculated quantity \( N_B^0(x) \) with the measured quantity \( x = x_0 \), versus the experimental parameter \( N_B = \mu_0 H_{\text{ext}}^2 h/2\sigma \). The curve shows a discrepancy which is roughly the same than Rosensweig [27] had already found with a more rigorous calculation of the stripe-stripe interaction. These authors have used the same strong assumption than us: the magnetization is supposed to be constant in a MF stripe, \( \mathbf{M}(r) = \mathbf{M}(0) \). We believe that this hypothesis is the reason for the discrepancy between the two calculations and the experiments. An exact calculation of the dipole-dipole interaction is not analytically tractable, and only a numerical approach is possible.
2.2. Bubble Phases. — An experiment performed in the variable thickness cell with \( \Phi_0 = 0.20 \) at low field \( H_{\text{ext}} = 13.2 \, \text{kA m}^{-1} \) gives an assembly of cylindrical droplets of variable radius, placed on a rather hexagonal lattice, as shown in Figure 3b. The calculation of the equilibrium size of the MF droplet is quite similar to the previous one. For a given \( h \), the surface energy, \( E_s \), of a single bubble of diameter \( d \), is:

\[
E_s = \pi \sigma d \left( h + \frac{d}{2} \right). \tag{14}
\]

The magnetic energy, \( E_m \), has the same formula as (6) with \( V = \pi h d^2 / 4 \). We have to reconsider the demagnetizing coefficient. Using the formula (9) in the appropriate geometry, i.e. a cylinder, we get for the dipole-dipole interaction in one MF bubble: \( D_0 = 1 - [1 + x^2]^{-1/2} \), where \( x = d / h \). We can notice that this expression has the correct limit: for \( x \to \infty \), i.e. an infinite 2D sheet, \( D_0 \to 1 \), and for \( x \to 0 \), i.e. an infinite bar, \( D_0 \to 0 \). For the interaction between the bubbles, we use the same approximation and assimilate one bubble to a giant-dipole of total momentum \( M = MV \) parallel to the external field (see Fig. 4b). Thus, the corresponding expression of the demagnetizing field, \( H_{\text{d1}} \), is given by (10), with \( n(r') = \sum \delta (r' - a_i) \), where the \( a_i \) are the vectors of the two-dimensional hexagonal pattern defined in Section 1 \( (a_i = 2\pi / q_i) \). Using the well-known formula given by [13]:

\[
\sum_{\text{hexagonal}} \frac{1}{t^2} \approx 11.034,
\]

we obtain: \( D_1 = (d^2 h / 16)(11.034 / |a_j|^3) \). And finally if we take into account that \( \Phi_0 \) is constant, we get: \( D_1 = \alpha / x \) with \( \alpha \approx 0.840^{3/2} \). Let us notice that the dependence of \( D_1 \) with the geometrical variable, \( x = d / h \), is the same in the both cases (stripes and bubbles). The expression of the total energy of one bubble, \( E_{\text{bubble}} \), interacting with other bubbles in a confined hexagonal lattice, is:

\[
\frac{E_{\text{bubble}}(x)}{\pi \sigma h^2} = x \left( 1 + \frac{x}{2} \right)^{-1} \frac{\chi N_B}{1 + \chi(D_0(x) + D_1(x))} \frac{x^2}{4}. \tag{15}
\]
The equilibrium magnetic Bond number, deduced by a minimization with respect to \( x \), is equal to:

\[
N_B^0(x_0) = \frac{2 \left( 1 + \frac{1}{x_0} \right)}{1 + \chi D(x_0)} \left[ 1 - \frac{\chi x_0/2}{1 + \chi D(x_0)} \left( \frac{dD}{dx} \right)_{x=x_0} \right].
\]  

(16)

The measurement of \( d \) versus \( h \) is reported in Figure 5b, as also the corresponding values computed using formula (16). The calculated quantity \( N_B^0(x_0) \) versus the experimental parameter \( \mu_0 H_{\text{ext}}^2 h/2\sigma \) is reported in Figure 6b. In this case we found, by the least square method, that the value of the bubble-bubble interaction parameter \( \alpha \approx 2 \), while the predicted value is of order of 0.1.

2.3. From the Stripes to the Bubbles. — Figure 7 shows the morphological transition from the bubble shape to the stripe shape in the variable thickness cell, induced by the increasing of the external field. No calculations has been yet performed; an accuracy model is needed to compare the energy of the two phases in order to establish the phase diagram.

3. Ordered Stripe Phase

It is possible to build a particular conformation where the MF stripes are parallel. This structure is a metastable situation of the labyrinthine pattern, meaning that if a rather strong perturbation is applied (for instance a spatial non-homogeneity of the applied magnetic field), the stripe structure is destabilized towards the labyrinthine one. Pragmatically, we obtain the parallel stripes by the following method: the cell is tilted from an angle \( \alpha \) from the horizontal position, consequently the MF falls down. The vertical field, \( H_{\text{ext}} \), is applied, and for a value of the field above the threshold value, \( H_c \), the so-called “peak instability” [28, 29] occurs at the interface between the MF and the oil: a comb made of MF peaks is formed as shown in Figure 8, and the distance, \( \lambda_c \), between two consecutive peaks (i.e. the critical wave length) is related to the capillary length. Another magnetic field, \( H_{\parallel} \), parallel to the cell, is used in order to transform the peaks into stripes by propagation of the MF. When the final pattern is obtained, the cell is delicately turned toward the horizontal position and the horizontal field \( H_{\parallel} \) is switched off, leading to the equilibrium conformation of the system: a pattern of MF parallel stripes alternating with parallel oil stripes. The distance between the stripes, i.e. the period of the pattern and the thickness of the MF stripes are fixed by the magnetic field \( H_{\text{ext}} \) strength for a given value of the control parameter \( \Phi \).

A phenomenological description of stripe phase using effective elastic constants in the framework of the smectic analogy has been already given for the garnet films [14, 15, 18], and for the MF stripes [30, 31]. A compression modulus, \( B \), corresponding to compressional or dilative stress and a bending modulus, \( K \), corresponding to curvature phenomena are measured and determined analytically as a function of the control parameters \( (H_{\text{ext}}, \Phi, h) \) in reference [31]. The minimal expression (i.e. to the lowest order) of the energy for describing the smectic deformation is the same than for a three-dimensional liquid-crystal smectic [32]:

\[
F_{\text{effective}} = \int_{\text{stripes}} dxdy \left\{ \frac{B}{2} (\partial_x u)^2 + \frac{K}{2} (\partial_y u)^2 \right\} + \ldots,
\]  

(17)

where \( u(x, y) \) is the deformation field of the stripes from their equilibrium position, \( x \) is the direction perpendicular to the stripes. We have established that the equilibrium smectic period, \( l_0 \), is a decreasing function of the field strength [31].
Fig. 7. — Photographs of the morphological bubble-stripe transition induced by the increasing of the magnetic field. These experiments are performed in the variable thickness cell ($h$: 1.1 → 4.7 mm) for a surface fraction, $\Phi_0 = 0.20$, and for the magnetic field values: 1) $H_{\text{ext}} = 13.1$ kA m$^{-1}$, 2) $H_{\text{ext}} = 15.7$ kA m$^{-1}$, 3) $H_{\text{ext}} = 16.4$ kA m$^{-1}$, 4) $H_{\text{ext}} = 19.7$ kA m$^{-1}$, 5) $H_{\text{ext}} = 26.2$ kA m$^{-1}$, 6) $H_{\text{ext}} = 32.8$ kA m$^{-1}$. The coexistence between stripe and bubble domains begins in the photograph 3, and finishes in the photograph 5.

The stripe pattern is unstable under the influence of high magnetic field. For $H_{\text{ext}} < H_c$ the stripes are parallel (Fig. 9a), for $H_{\text{ext}} = H_c$ a primary instability occurs: the stripes undulate with a sinusoidal deformation (Fig. 9b). The period of this undulation along the stripes is related to the penetration length in the framework of the smectic analogy: $\lambda = \sqrt{K/B}$ [33]. This characteristic length defined by de Gennes [34] is of order of the smectic period, $\lambda \approx l$. One can notice that the distance between the stripes, $l$, and the thickness of the stripes, $d$, decrease when increasing the field, but the ratio $\Phi = d/l_0$ is constant. This instability can be understood qualitatively as follows: since $\Phi$ is constant and $l_0$ decreases when the field increases, the width, $d$, has also to decrease, and consequently the length of the stripes increases. Since the stripes are built in a confined geometry, the simplest way for increasing the length is to undulate. For higher value of the magnetic field, the undulations contain several modes because
the long range magnetic interaction induces coupling between stripes. This coupling leads to the chevron pattern ("zig-zag") and the transition occurs continuously (Fig. 9c). Since the value of the control parameter $\Phi$ is fixed, it means that no stripe disappears or is created. A secondary instability is visible in Figure 9d: the nucleation of a tip at the edge of the chevrons. For higher value of the magnetic strength the tip grows and becomes a loop, finally we obtain a hairpin pattern (Fig. 9e). A global view of the hairpin structure is given in Figure 10. By increasing the value of the magnetic field, we observe interaction of the hairpin shape like: two hairpins turn together. The symmetry of parity is broken: the hairpin pair can rotate to the left or to the right, depending on the surrounding media (Fig. 9f).

4. Liquid Magnetic Froth Phase

A 2D froth texture whose skeleton is made of MF is obtained if the Hele-Shaw cell is submitted to a homogeneous alternating magnetic field. An extensive study of this new phase is reported elsewhere [35]. The most of the characteristics of the froth is monitored by the external field. For a given amplitude, the thickness of the MF boundaries is fixed, and increases when the field is decreased. Since the total area of the MF is fixed, an evolution of the pattern is obtained as the magnetic field is changed: when decreasing the amplitude of the field, the number of bubbles decreases and the bubbles grow, so the system tends to form a single drop of MF surrounded by the other immiscible liquid in zero field. This evolution is called coarsening, by analogy with the time-evolution of the soap froths.
Fig. 9. — a) Parallel stripes of MF alternating with stripes of the other immiscible liquid, $H_{ext} = 12.4$ kA m$^{-1}$. b) By increasing the magnetic field just above the threshold $H_c$, $H_{ext} = 14.7$ kA m$^{-1}$, the stripes are slightly bent. c) For $H_{ext} = 15.9$ kA m$^{-1}$ the chevron pattern occurs. d) For $H_{ext} = 18.8$ kA m$^{-1}$ another instability occurs, the edge of the chevron forms a double tip (underlined by a white arrow). e) $H_{ext} = 23.1$ kA m$^{-1}$ the tip grows and has a hairpin shape like. f) Two hairpins interact and turn together, $H_{ext} = 34.8$ kA m$^{-1}$. The underlying lines (a millimetric grid) give the macroscopic scale of the system.

This new phase presents analogies with patterns formed by many other physical systems. The example of cellular structures observed in magnetic garnets films [36–38] is the closest of the MF froth. These systems show several patterns when an external magnetic field is applied perpendicularly to the film. In low field, a stripe pattern is formed by domains of uniform magnetization parallel and anti-parallel to the field, and the total magnetization of the film is low. Above a threshold value, a cellular pattern appears, where bubbles of magnetization parallel to the field are surrounded by a skeleton whose magnetization is anti-parallel to the field. Whereas a coarsening of MF froths occurs when the magnetic field is decreased, the pattern in these system evolves towards a single domain of magnetization when the field is increased; the bubbles grow larger at the expense of domains of magnetization anti-parallel to the field because all the dipoles tend to get the field direction.
Fig. 10. — Extensive view of the hairpin pattern \( H_{\text{ext}} = 40.0 \, \text{kA m}^{-1} \), the scale is given by the white bar.

Fig. 11. — a) Wet liquid magnetic froth consisting in circular oil bubbles surrounded by an important quantity of MF. b) Dry froth with faceted oil bubbles. The transition between (a) and (b) is obtained by draining the MF so as to decrease the quantity of MF in the Plateau borders. A little magnet introduced at the edge of the cell creates a magnetic force, which attracts the MF towards the cell edge.

The 2D liquid magnetic froth is also comparable to soap froths confined between two parallel transparent plates. The evolution in time of the soap froth structure has been already well understood [39]. Since the energy of these systems contains only an interfacial contribution, they coarsen in time in order to minimize the interface: the mean size of bubbles increases and the number of bubble decreases so as the pattern turns into a single bubble. In contrast, liquid magnetic froths do not coarsen in time but evolve when decreasing the amplitude of the magnetic field. Indeed, the energy of a liquid magnetic froth contains two additional contributions: the interaction between the magnetic dipoles and
Fig. 12. — Anisotropic liquid magnetic froth. The alternating magnetic field $\vec{H}$ is perpendicular to the cell. A static and homogeneous magnetic field $h$ parallel to the cell is added so as to elongate the oil bubbles in its own direction. We observe groups of cells aligned along the additional field direction.

Fig. 13. — Deformed liquid magnetic froth. Starting from a dry froth as shown in Figure 11b and increasing the amplitude of the magnetic field above a threshold value, the MF skeleton undulates and the oil bubbles get strongly deformed.

the magnetic field, and the magnetic dipole-dipole interactions. In the presence of an external magnetic field, the magnetic particles are aligned in the field direction (in the case of an alternating field, the period of the field is greater than the time during which the magnetic particles turn within their carrier liquid, so the particles follow the motion of the magnetic field). Since the field direction is vertical, the magnetic dipoles are oriented perpendicularly to the plane of the cell. In this configuration, the dipole-dipole interactions are repulsive, and tend to increase the interface between the MF and the oil. There is a competition with the interfacial energy contribution which tends to decrease this interface. Therefore, an equilibrium state can be reached, and the magnetic froth is stable in time. An intermediate case between soap froth and liquid magnetic froth has been experimentally studied by Sudo et al. [40]: using a ferrofluid in which the MF/air surface tension has a very low value, they built a 2D soap-like foam by
Fig. 14. — Transition from the froth to the bubble phase. The surface fraction is equals to $\Phi_0 = 0.23$, the magnetic field is homogeneous and alternating (frequency 50 Hz) with a modulus equals to 18.6 kA m$^{-1}$. For high values of the field strength, or high surface fraction, nucleation of oil holes inside the MF area is followed by the ejection of MF droplets in the oil cells. The photographs 1 to 6 represent the evolution in time of the system towards equilibrium after the magnetic field has been switched on. (1) $t = 0$, (2) $t = 25$ min, (3) $t = 1$ h 20, (4) $t = 1$ h 39, (5) $t = 1$ h 44, (6) $t = 2$ h 16. Finally, we get the dual pattern of the magnetic froth i.e. the MF bubble pattern. We can also notice the existence of MF rings.

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the introduction of air bubbles within the MF. They showed that a magnetic field applied perpendicularly to the plan of the froth stabilizes the structure: from a threshold amplitude of the magnetic field, the number of air bubbles does not evolve in time, whereas the froth coarsens in time in zero field like a soap froth.

We observed a wide variety of MF froths. Figure 11a shows a so-called wet froth: the oil cells are circular and the quantity of surrounding liquid is important around the vertices, i.e. in the Plateau borders. It is also possible to build a dry froth (Fig. 11b), in which the cells are faceted. Figure 12 shows an anisotropic froth built using an additional magnetic field in a parallel direction to the cell. Note that the cells are arranged along some lines parallel to the additional field direction. Finally, in high magnetic field, a deformed magnetic froth is obtained.
as shown in Figure 13. Starting from a dry froth (Fig. 11b) and increasing the amplitude of the magnetic field above a threshold value, the MF skeleton undulates and the bubbles get strongly deformed; this instability is analogous to the bending instability in the stripe phase described in Section 3.

For high surface fractions \( \Phi > 0.5 \) or for high strength of the external field, we observe the transition from the froth pattern to the dual one, \( i.e. \) the bubble phase. The photographs of Figure 14 show this transition, which occurs only with alternating field.

5. Conclusions

To summarise, we present in this paper several conformational instabilities which lead to patterns like stripes, bubbles or the dual situation, froth. A simple analysis of the static behaviour of these modulated phases is given. An experimental evidence of the bubble-stripe transition is shown; numerical calculations is needed for the understanding of this morphological transition. A description of the shape instabilities, like the chevron pattern or the “bending” froth, should also be done. This system presents macroscopic patterns similar to the microscopic ones in the Langmuir monolayers or in the garnet films. Magnetic fluids are one more system to get patterns induced by the competition between attractive short-range interaction and repulsive long-range interaction.

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References

[33] Another way for the obtention of the value of the length penetration, $\lambda$, is to fit the profile of an edge dislocation [31]. The length scale on which the topological defect is extended is proportionnal to $\lambda$.