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Structural disorder in Benard-Marangoni convection

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Résumé. — Une étude systématique de l'influence de l'écart au seuil, de la forme et de la taille du récipient sur la structure de l'instabilité de Bénard-Marangoni (B-M) a été entreprise. Au moyen de certains outils théoriques, souvent utilisés pour décrire la fusion dans les milieux à deux dimensions, ces influences sont étudiées quantitativement. Les résultats expérimentaux montrent que le désordre est minimum lorsque le récipient est de forme hexagonale et que dans ce cas il passe par un maximum lorsque le rapport d'aspect est d'environ 53. Le désordre est une fonction croissante de l'écart au seuil.

Abstract. — The influence of the vessel shape, the vessel size and heating on convective patterns in the Bénard-Marangoni (B-M) instability has been analysed. These influences can be quantitatively studied by means of some theoretical tools, often used to describe melting in two dimensions. Experimental results show that a hexagonal geometry minimizes the disorder in the hexagonal pattern of B-M convection. This disorder is maximum when the aspect ratio is about 53. The disorder increases when the distance to the threshold increases.

1. Introduction

In recent years there has been an increasing interest in the study of patterns that appear in systems far from equilibrium [1]. Some systems, when driven out of equilibrium by means of an external parameter, develop very regular patterns of motion. One of the challenges in macroscopic physics is to understand and to interpret the mechanisms responsible for these regularities. At a next step one will also be interested in the complexities that arise when the control parameter continues to increase, finally leading to turbulent or chaotic motions [2].

A prototype of these nonequilibrium ordered structures is given by the Rayleigh-Bénard (R-B) problem [3]. In this instability, a pattern of rolls develops in normal liquids as seen in experiments and in some weakly nonlinear analyses. Irregularities are always present in these patterns, including dislocation-like defects and orientational disorder [4]. These complicated patterns may be characterized by a two-dimensional horizontal wavevector field that, from the theoretical point of view, can be obtained from amplitude equations [5].

The problem of disorder is linked to the very basic one of the wavenumber selection mechanisms in dissipative structures and it seems to play a decisive role in the transition to turbulence in these confined systems [6, 7].

In this article we present results on disorder in a slightly different convective instability, the Bénard-Marangoni (B-M) instability [8]. Historically, this was discovered before the R-B one, but since the latter is simpler than the B-M instability, it has been more studied from both the theoretical and experimental points of view. In the B-M convection the upper surface is free and then the surface tension variations provide a competing instability mechanism [9].

In the B-M case, a pattern of hexagons is the most stable structure in usual situations. Photographs of these patterns always show some defects that are mainly pentagon-heptagon pairs and aggregates of irregular polygons [10]. It is interesting to note that this kind of defects are also observed in different natural hexagonal structures, for example, in honeycombs, in crystals, in liquid crystals or in interfacial patterns during solidification [11]. It is widely recognized that simple, regular, and symmetric patterns are exceptional in convection experiments.

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In the scarce theoretical analyses undertaken in the B-M convection, the dynamics of these irregular patterns has not been analysed. Until now, only one experimental study exists [10] in which the formation, mutual transformation and evolution of defects in hexagonal patterns are described. As a complete theory of defect dynamics is still lacking, it can be useful to use the analogy between these patterns and layered materials in two dimensions (2D) [12]. Occelli et al. [13] have proposed a quantitative description of spatial disorder using some theoretical tools used to describe melting in 2D hexagonal lattices. Although similarities between 2D layered structures and B-M patterns exist, it must be noted that the differences are very stringent. The main ones are that 2D structures are static and are made of material particles, while convective patterns are purely dynamic and are formed by convective cells whose number is not conserved, i.e., they can be created or annihilated (the total number of cells in the pattern suffers small fluctuations).

The aim of the present work is to determine the influence of the vessel size, the vessel shape and the distance to the threshold on the disorder in B-M convection. With the help of theoretical methods that are described in § 2. The experimental apparatus and procedures are described in § 3. In § 4 the main experimental results are quoted. Finally, a summary of the main conclusions and a discussion of the results are gathered in § 5.

2. Measurements of the disorder.

As shown in a previous paper [10] the presence of pentagon-heptagon pairs is the main manifestation of disorder in a hexagonal convective pattern. (These pairs introduce a non-zero Burger’s vector.) Therefore, a direct and easy technique to measure disorder is to count the number of these defects or to quote their percentage $d_D$ in the pattern.

This measure only gives general information on the disorder of the structure, but not on the local or global distortions introduced by these pairs into the adjacent cells. These modifications may be quantitatively characterized by radial and orientational correlation functions often used in 2D systems [14]. If $u$ represents the displacement at a point $r$, and $K$ is the set of vectors in the reciprocal lattice, one can define a complex radial order parameter

$$ \rho_K(r) = \exp[ i K \cdot u(r) ] $$

Similarly, if $\theta_j(r)$ stands for the orientation with respect to a reference axis of a bond between two nearest neighbour cells, an order parameter for bond orientation can be defined

$$ \psi_6(r) = \frac{1}{6} \sum_{j=1}^{8} \exp 6 i \theta_j(r) $$

The correlation functions over the set of convective cells are defined by the expressions

$$ G(r) = \langle \rho_K(r) \rho_K(0) \rangle $$

for translation

$$ G_6(r) = \langle \psi_6(r) \psi_6(0) \rangle / \| \psi_6(0) \|^2 $$

for orientation

where brackets mean an average over the pattern. They are introduced to measure a loss of order, to characterize phase transitions (mainly melting) in 2D solids and, in particular in the Kosterlitz-Thouless-Halperin-Nelson (KTHN) [15] theory of dislocation-disclination mediated melting of 2D solids.

In 2D solids at large distances, $(G(r) - 1)$ decreases to zero as $r^{-n}$, showing a quasi long range translational order, while $G_6(r)$ is constant, that is equivalent to a long orientational order. On the contrary, in a liquid, both radial and orientational correlation functions decrease exponentially and then only a short range order exists.

Nelson and Halperin [15b] suggested that a new phase, called hexatic, may exist in 2D systems. Then a solid can melt into a liquid via two continuous phase transitions. The hexatic phase is different from crystals because it does not have a long range orientational order. The correlation functions decrease in this phase as $r^{-n}$ for orientation and as $\exp(-ar)$ for translation (here $a$ is a constant). This corresponds to a quasi long range orientational order and a short range translational order. During the last years many experimental and numerical works have been undertaken to show this new phase. The aim here is not to discuss this basic problem on 2D systems, but to use the analogy between disorder in this static 2D phase and the B-M nonequilibrium patterns.

The disorder density $d_D$ and the two correlation functions are only useful when patterns have a number of cells sufficient to make statistics. (In particular we have calculated correlation functions when the pattern contained more than 380 cells.) In small vessels, with only a few cells, the structure does not have any defect ($d_D = 0$) but almost regular hexagons (Fig. 1), whereas some distortion is still present in the pattern. Therefore, even in this situation, it is important to have a quantitative measure of this distortion to determine whether the

Fig. 1. — Structure without dislocations for a small aspect ratio ($\Gamma = 27.7$, $\epsilon = 0.05$).
pattern is stationary. In order to achieve this goal, we propose the following function

\[ F_D = \sum_{i=1}^{N} \frac{1}{n_i} \sum_{j=1}^{n_i} |l_{ij}/d| \]

where \( N \) is the number of complete cells in the pattern, \( n_i \) is the coordination number of the \( i \)-th cell, \( l_{ij} \) is the distance between the centre of the \( i \)-th cell and the centre of its \( j \)-th neighbours, and \( d \) stands for the length between the centres of two neighbour cells averaged over the whole pattern. (To make this average, only hexagonal cells are taken into account.) In the following we refer to this function as the disorder function \( F_D \). This is an average of the deviations of the distance between the centres of cells on the whole pattern with respect to a simple, completely regular pattern. In analogy with the entropy in regular honeycomb lattices [14] a logarithmic function is chosen. The absolute value of this function is taken in order to have contributions that, do not cancel each other, when the length between two centres is smaller or bigger than average. These contributions are averaged over the nearest neighbours and, finally, all these contributions are added over the whole pattern. Note that with this function one can also account for distortions in patterns with very few convective cells.

3. Experimental device.

The experimental device consists of a thin horizontal layer of silicon oil in a vessel with a copper block bottom at uniform temperature \( T_s \), and lateral walls made of plexiglass. The upper surface of the liquid is free, at temperature \( T_u \), and in contact with the ambient air. These temperatures are measured by means of a series of thermocouples and the liquid depth \( d \) by means of micrometer comparators. A more detailed description of this device and of the measurement methods can be found in reference [16].

The external parameter which controls the instability is the temperature difference \( \Delta T = T_s - T_u \) across the layer. Usually, it is more useful to take a normalized parameter

\[ \epsilon = (R - R_{c})/R_{c} = (M - M_{c})/M_{c}, \]

where \( R \) and \( M \) are the Rayleigh and Marangoni numbers [3, 9], and the subscript \( c \) stands for the corresponding threshold value. Parameter \( \epsilon \), known as the distance to the threshold, is an important parameter in convection. When increasing \( \epsilon \), new instabilities can appear, leading ultimately to turbulence.

Lateral walls impose important constraints on the liquid motions, through the lateral boundary conditions. The vessel size is taken into account by means of a nondimensional parameter, the aspect ratio \( \Gamma \), the ratio between a characteristic horizontal length and the liquid depth. In our experiments, instead of using the usual definition of \( \Gamma \), the following form is taken \( \Gamma = \sqrt{S/d} \) (\( S \) is the surface area). This form permits us to compare vessels with a similar number of cells but with different shapes. Besides \( \epsilon \) and \( \Gamma \), the vessel shape is important to determine the external effects on the disorder of the pattern.

Therefore, experimental measurements have been made in vessels with the same shape but with different aspect ratios \( \Gamma \), and in vessels with different shapes (square, hexagonal and circular) with comparable aspect ratios \( \Gamma \), both at different distances to the threshold \( \epsilon \).

Liquid motions are visualized by means of a shadowgraphic technique [16] or by direct observation after addition of aluminium powder in the fluid. At fixed values of \( \epsilon \), photographs of the convective structure are taken at regular time intervals (12 min) for a long period (at least 8 h). Then photographs are digitized [13]. This information and a suitable software allow us to obtain the values of the relevant functions \( (d_D, F_D, G(r) \text{ and } G_6(r)) \).

4. Experimental results.

From a general point of view we must stress that the pattern is not stationary but fluctuates. These fluctuations are a consequence of the dynamics of defects: they grow, evolve and sometimes annihilate. These changes are responsible for strong fluctuations in the direct two measures of disorder, namely \( d_D \) and \( F_D \), observed in this experiment (Fig. 2). It must be noted that these fluctuations do not behave monotonically. (This fact suggests that a potential model, like that proposed by Bestehorn and Haken [17], is not completely valid for B-M convection). The histogram of the corresponding values shows, however, that they follow a normal law for each value of \( \epsilon \). The uncertainty \( \Delta d_D/d_D \) reaches about 15\% and

![Fig. 2. — Fluctuation of the density of defects \( d \) as a function of time \((\Gamma = 60.3, \epsilon = 0.05)\).](image)
it is independent of $\varepsilon$. On the contrary, the amplitude of the fluctuation increases with $\varepsilon$. In small aspect ratio case $d_D = 0$, but $F_D$ is different from zero and still displays small fluctuations.

We now quote the main results of different three series of measurements to determine the role of the external constraints on the convective patterns.

4.1 INFLUENCE OF THE CONTAINER SHAPE. — With the same liquid we have filled three different vessels with similar aspect ratios ($\Gamma \approx 85$) but with different shapes: hexagonal ($V_h$), cylindrical ($V_c$) and square ($V_s$) vessels. They are heated in the same way (the same distance to the threshold $\varepsilon = 0.05$). An example of pattern arising in these vessels can be seen in figure 3. It is noteworthy that marginal cells, i.e., those in contact with walls, always have sides perpendicular to the walls (a similar feature can be observed in the R-B case [4]). This strong constraint seems to be one of the causes of disorder, mainly in boxes with a small aspect ratio.

Experimental results for $d_D$ and $F_D$ are listed in table I. The density of defects $d_D$ is minimum for $V_h$ and maximum for $V_s$. This can be understood simply from topological arguments. The two conditions, a) a perfect hexagonal arrangement and b) having all the marginal cells perpendicular to the walls can be fulfilled in a hexagonal container, and not in cylindrical or square containers. Then, at least theoretically, one can have a completely regular pattern of hexagons in that container. (However, in convective patterns some defects are always present in big hexagonal vessels).

In contrast, in cylindrical or square containers some topological defects must always be present. (From Euler's theorem, a hexagonal arrangement in a cylindrical container must have at least five pentagons (18)). Among the shapes considered here the square is the most incompatible geometry with the conditions mentioned above, thus it is not surprising to find the maximum of defects in the vessel $V_s$.

The density $d_D$ is greater in $V_s$ than in $V_h$. But strains induced by a cylindrical symmetry seem to be more uniformly distributed: the distortion in the lattice is small. These two effects taken together (more defects but less strains from walls) could explain that $F_D$, which is a global measure of the distortion in the pattern, is about the same for $V_c$ and for $V_h$ (Table I). Likewise, it can be noticed that the amplitude of $d_D$ fluctuations is the lowest for $V_c$, probably in connection with the uniform distribution of strains. For $V_s$, $F_D$ is significantly higher, in agreement with previous comments.

A study of the correlation functions may lead to similar conclusions. The orientational order (Fig. 4) is fairly kept in $V_h$ and $V_c$. On the contrary, for $V_s$ it decreases fastly when $r$ increases. A similar behaviour is obtained for the radial correlation function, as can be seen in figure 5: a certain order is kept up to 9 for $V_h$, 7 for $V_c$ and 5 for $V_s$.

The function $G_6(r)$ and the envelope of $(G(r) - 1)$ decrease to zero either as $r^{-n_1}$, or as $\exp(-n_2 r)$. The twelve coefficients $n_1$ and $n_2$ are shown in table II. In $V_h$ and $V_c$ the correlation functions are better fitted by a power function $r^{-n}$, corresponding to quasi long range translational and orientational order. The results in the square vessel $V_s$ correspond to an exponential behaviour, typical of short-range order. As discussed in a

Table I. — Disorder function $F_D$, density of defects $d_D$ and standard deviation $\beta$ for various vessels ($\varepsilon = 0.05$, $\Gamma = 85$).

<table>
<thead>
<tr>
<th></th>
<th>$F_D$</th>
<th>$d_D$ (%)</th>
<th>$\beta$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>hexagonal</td>
<td>0.028</td>
<td>3.7</td>
<td>1.5</td>
</tr>
<tr>
<td>cylindrical</td>
<td>0.027</td>
<td>6.1</td>
<td>0.9</td>
</tr>
<tr>
<td>square</td>
<td>0.035</td>
<td>8.2</td>
<td>1.6</td>
</tr>
</tbody>
</table>

Fig. 3. — Convective structures in a finite container ($\Gamma \approx 85$, $\varepsilon = 0.05$) a: hexagonal ; b: circular ; c: square.
previous paper this fact does not allow us to conclude that the pattern in $V_h$ or in $V_c$ corresponds to a hexatic phase because the difference between power law and exponential function is not significant. But these results show that the disorder in a pattern of cells is strongly influenced by the shape of the container. The pattern is more disordered in $V_s$ than in $V_c$, the latter induces more disorder than $V_h$.

4.2 Influence of the aspect ratio. — To determine the influence of the aspect ratio $\Gamma$ on the convective motions we have observed the pattern for the same liquid depths at the same distance to the threshold ($\epsilon = 0.05$) in hexagonal boxes with seven different side lengths (Table III). The main results are shown in figure 6 where, for each $\Gamma$, the average value of $d_D$ and the associated standard deviations of the fluctuations are drawn. For small $\Gamma$, $d_D$ is zero and it remains small up to $\Gamma \approx 53$ (corresponding roughly to $N = 200$ cells), while, as obtained in a previous study [13], for $\Gamma \approx 200$, $d_D$ is constant, about 2.5%. The disorder function exhibits a similar behaviour, but the variations are less pronounced.

Two states of disorder can be distinguished. For small $\Gamma$ the structure is very sensible to the form of the walls. In the case of a hexagonal container, there are no defects ($d_D = 0$) but some distortion ($F_D \neq 0$), mainly orientational, remains. For a very large aspect ratio ($\Gamma \approx 70$) the layer can be considered as being infinitely extended so that the strains induced by the walls do not reach the central part of the pattern. In these wide vessels, complicated patterns may be understood from the existence of an orientational degeneracy and of a band of
stable wavenumbers. For $\Gamma \approx 50$, a competition between those strains and the domains of hexagons with different orientations induces a maximum of disorder. This feature seems to be analogous to structures in 3D metallic heaps. As observed by Farges [19] when there are few atoms, they tend to form a icosahedral structure while, when there are many atoms (for $N \approx 60$) a hexagonal symmetry prevails. A competition between space filling and the best possible piling up is proposed as an explanation of this interesting fact.

4.3 INFLUENCE OF THE DISTANCE TO THE THRESHOLD. — To determine how disorder depends on $\epsilon$, we have used a hexagonal vessel in which, as seen above, the disorder is minimized. This vessel contains a layer of fluid which is submitted to different heatings. The measurements are repeated for three liquid depths with the same vessel, corresponding to $\Gamma = 82.3$, $71.4$ and $28.5$ respectively. The main results can be seen in figure 7: when $\epsilon$ increases, more defects appear in the pattern. Within the precision of our measurements, the curves obtained for the variation of $d_D$ (or $F_D$) with $\epsilon$ have the same form for the three aspect ratios considered. This means that there is no coupling between $\epsilon$ and $\Gamma$ at a fixed shape. For small $\epsilon$, $d_D$ increases very fast, while for higher $\epsilon$, the number of defects grows more slowly. As already noticed by Occelli et al. [13] a dramatic increase of the disorder is observed when $\epsilon$ increases: for $\epsilon = 5$, $d_D$ is greater than 50%. So that, all the polygons are irregular, so the pattern cannot be considered as being hexagonal.

These experimental results can be fitted with an exponential law $\exp(-B/\epsilon)$ (with $B = 0.56$), analogous to the Arrhenius law for the density of point defects in real crystals. This fact supports the analogy between these convective structures and real crystals, already underlined in a preceding article [10]. In this sense, $B$ could be understood as an activation energy for defects in these convective patterns.

Finally, the orientational and radial correlation functions are shown in figures 9 and 10, respectively for two values of $\epsilon$ ($\epsilon = 0.05$ and 1.02), only for the
largest aspect ratio $\Gamma = 82.3$. Both functions display a fast decrease of order with increasing $\varepsilon$. For $\varepsilon = 1.02$, $G_6(r)$ decreases as $\exp(-1.13r)$ and the envelope of $(G(r) - 1)$ as $\exp(-0.91r)$. These values correspond to a short range order as in 2D liquids. When $\varepsilon$ increases the structure varies from a quasi-long-range orientational and translational order ($\varepsilon = 0.05$, few defects) to a short-range order ($\varepsilon = 1.02$, many defects) but without displaying the characteristics of a transition from a 2D « hexatic » to a 2D liquid phase.

5. Conclusions.

In this paper we show how the use of theoretical tools useful to describe melting in 2D systems can be applied to study quantitatively the influence of three external parameters (the vessel shape, the aspect ratio and the heating) on the disorder of hexagonal patterns in B-M convection. Experimental results confirm that vessels with a shape that favours hexagonal symmetry (hexagonal, cylindrical) minimize the disorder. As in R-B instability, the number of defects is shown to increase with heating.

The results concerning the influence of the aspect ratio are more surprising. In particular we point out the existence of a value of $\Gamma$ for which a disorder maximum appears, probably due to a competition between lateral constraints and the orientational degeneracy in the pattern.

Another interesting point concerns the existence of stationary states in these convective patterns. It was generally thought that disorder must vanish when convection is very near the threshold [20]. In the R-B convection, however, numerical simulations and carefully controlled experiments allow us to conclude that some disorder remains near the threshold in high or intermediate aspect ratio conditions [21-22]. An extreme case was found in liquid helium where turbulent motions appear as soon as convection starts [22a].

In B-M convection our measurements suggest that there is an intrinsic disorder, not due to external constraints, in hexagonal patterns. In view of the temporal fluctuations of the disorder functions ($d_D$ and $F_D$) observed in time it seems that even in patterns with few convective cells a stationary state does not exist. However, it may be asked whether a simple symmetrical pattern can be unstable with respect to the more complicated patterns observed. A partial answer can be given after forcing initially a regular pattern, following its time evolution. (A thermal technique developed recently allows us to do this in hexagonal patterns [23]).

These results can be compared with numerical simulations of hexagonal patterns [17]. The present measurements indicate, however, that the behaviour of the defects and their fluctuations cannot be described by means of variational principles, and therefore by potential models, like that proposed by Bestehorn and Haken [17], in which the pattern becomes stationary after a sufficient time.

The present experiments have been made with a liquid open to the atmosphere. Thus it could be argued that air motions could distort the pattern. This would be responsible for part of the fluctuations in thin layers but not in thicker layers. Moreover the experiments of Koschmieder [20], made with a careful regulation of the temperature difference across the liquid layer plus a small air gap, always show some defects that evolve with time.

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