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Convective patterns in a binary mixture with a positive separation ratio

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Résumé. — Nous présentons une étude expérimentale des propriétés spatiales et dynamiques des structures convectives obtenues dans les mélanges binaires présentant un rapport de séparation positif. Dans ce dernier cas, le champ de concentration et celui de température sont déstabilisants mais ils conduisent respectivement à des structures de type différent. En utilisant un dispositif quantitatif de mesure optique, nous avons mesuré les vecteurs d'ondes et l'amplitude de la convection. Pour des petites différences de température, seul le champ de concentration est déstabilisant et participe à l'établissement de structures de type carrés. Lorsque la différence de température appliquée à la couche de fluide est importante, le champ de température devient aussi déstabilisant. Nous nous sommes intéressés à la compétition entre les structures de type carré, liées au mode de concentration, et celle de type rouleaux, liée au mode de température. En étudiant la dynamique de ces structures, nous avons mis en évidence des ondes propagatives qui ont été récemment prédites par Linz *et al.* [1].

Abstract. — We present an experimental study of the convective patterns and their dynamics occurring in a binary mixture presenting a positive separation ratio. In this situation, both the concentration and the temperature fields are destabilizing, and they lead to two different kinds of convective patterns. Using quantitative optical methods, we have measured the convective motion wavevectors and their non-linear amplitude. For small temperature differences, only the concentration field is destabilizing and the convective motions are organized in a square pattern. We study the evolution of the wavevector and the amplitude of the square pattern. For larger temperature differences both fields are destabilizing and we investigate the competition between the square pattern, reminiscent of the concentration field, and the roll pattern associated with the temperature field. By studying the dynamics of this competition we have evidenced the existence of traveling waves which have been recently predicted by Linz *et al.* [1].

Introduction.

The original motivation of this study is the investigation of the square pattern and its competition with a roll pattern. In a previous experiment. Le Gal *et al.* [2] observed, in convection using silicone oil, a square pattern and a transition to a roll pattern involving an

oscillation between the two sets of rolls which constitute the square pattern. At that time the authors attributed the effect to the horizontal boundary conditions which were of moderate thermal conductivity. Studying water-ethanol mixtures, Moses and Steinberg [3] observed a very similar behavior when the so-called separation ratio of the mixture was positive. Furthermore, these authors gave a qualitative explanation of the phenomenon and provided further quantitative tests to be made. More recently, Müller and Lücke [4] have investigated this problem using an extended Lorenz model. Using a numerical simulation of their model, they confirmed the conjecture presented by Moses and Steinberg and found the oscillations between square and roll patterns. In a very similar approach, Linz *et al.* [1] have shown that, in the case of a mixture with a positive separation ratio, even the simple roll pattern becomes time dependent and leads to traveling waves. All these results were unexpected for binary mixtures with a positive separation ratio and challenge further experiments. We have investigated a binary mixture having a strong separation ratio in order to obtain unambiguous effects.

To confirm the Moses and Steinberg interpretation, the first test consists in demonstrating that the silicone oil used in [2] effectively behaves as a mixture with a positive separation ratio. Silicone oil is actually a mixture of polymers with a distribution of mass and was shown [5] to exhibit a small segregation when a temperature gradient was applied to an oil layer. This effect may be modelled with a positive separation ratio of the order of 3×10^{-3} .

Convection in binary mixtures has encountered a very important experimental [6] and theoretical [7] interest during these last years, the main reason being that the convective motions may have two origins. In a simple fluid, only the density variations related to the thermal expansion may lead to an unstable situation. In a binary mixture this mechanism still exists but the density may also vary with the concentration of the mixture and this constitutes the second mechanism leading to convection. Experimentally, it is easy to apply a temperature gradient on a fluid layer but, on the other hand, it is more difficult to control a concentration gradient externally, except by using the Soret effect. This effect corresponds to the cross coupling between the heat flow and the molecular flow of the components of the mixture. In other words, applying a temperature gradient to a layer of binary mixture produces a heat flow and also the preferential migration of one component of the mixture in the direction of the temperature gradient. Owing to the impermeable nature of the usual boundary condition, a gradient of concentration builds up in the layer. In that way the externally applied temperature gradient also creates a concentration gradient. Both may lead to convective motions. Although the temperature difference is at the origin of the convective motions, their behaviour is determined by the interplay between the temperature field and the concentration field. The coupling between these two fields is given by the separation ratio ψ which is the ratio of the density gradient produced by the concentration one, to the density gradient produced by the thermal expansion of the mixture : $\psi = \beta \nabla C / \alpha \nabla T$ where α is the thermal expansion coefficient $\alpha = \frac{1}{\rho} \cdot \left(\frac{\partial \rho}{\partial T} \right)$ and β is the concentration expansion coefficient $\beta = \frac{1}{\rho} \cdot \left(\frac{\partial \rho}{\partial C} \right)$.

The separation ratio depends on the mixture considered and on its mean temperature, the concentration gradient ∇C is related to the temperature gradient through the Soret coefficient : $\nabla C = (D_T/D) C_0 \cdot (1 - C_0) \cdot \nabla T$ where C_0 is the mean concentration of the mixture and D_T/D is the Soret coefficient, with D being the diffusivity of the mixture components. The main motivation of most actual studies of binary mixtures concerns the case where both fields are opposed to each other, that is the thermal field is destabilizing whereas the concentration one leads to a stabilizing density gradient. This case corresponds to the negative separation ratios, the density stratification caused by the concentration field leads to

waves. The destabilizing temperature gradient results in an amplification of these waves under appropriate conditions. The result is an oscillatory behavior of the convective rolls at the threshold of the convection, which has been predicted [7] and observed [6].

The situation that we investigate here corresponds to the opposite situation, where both density gradients are destabilizing. In this case, the linear theory predicts a dramatic decrease of the convective threshold but no wave solution, that is a stationary pattern. In fact a rough approximation is to consider that both fields have their own Rayleigh number, their expressions being : $R_a = \alpha g d^3 \Delta T / \kappa \nu$ for the temperature and $R_s = \beta g d^3 \Delta C / D \nu$ for the concentration (this definition is not the usual one in binary mixture, but corresponds to the actual Rayleigh number if the concentration field is involved alone). Owing to the Soret coupling we have $R_s = \psi R_a / L$ where $L = D / \kappa$ is the Lewis number. In a first approximation when both fields are present, the Rayleigh number of the mixture is the sum of the temperature and concentration one, this will define the threshold of the convective motions. In most liquids, the Lewis parameter, which compares the diffusivity of both fields, is rather small and of the order of 10^{-2} ; the separation ratio depends on the mean concentration C_0 of the mixture and is typically of the order of 0.1 when $C_0 \approx 0.01$. It follows that, in most cases, the Rayleigh number of the mixture is dominated by the concentration contribution. The instability which first appears may be considered as nearly a pure concentration instability, the temperature gradient being just a mean to establish the concentration gradient.

Provided that we take the different time scales into account, there is an analogy between the temperature field and the concentration field, and the convective motions which they involve are similar. However, the concentration instability differs from the usual thermal instability in its horizontal boundary conditions : $\partial C / \partial z = 0$ at $z = 0.1$ to compare with $T = T_0 + \Delta T$ at $z = 0$ and $T = T_0$ at $z = 1$. The boundary conditions experienced by the concentration field are equivalent to that of the temperature in the case where the thermal conductivity of the top and bottom plates is much smaller than that of the fluid. This situation has not received an extensive experimental study since it is somewhat difficult then, to control the temperature difference applied to the fluid layer but in the case of pure water with plexiglass as horizontal boundaries, Le Gal and Croquette have shown that convection appears with square cells [8]. This result was predicted theoretically by Busse, Riahi, Proctor and Jenkins [9] who have determined the stability of square cells *versus* rolls at the threshold of convection as a function of the relative conductivity of the boundaries with the fluid. Referring to this result, Steinberg and Moses infer that square cells should occur in the same way for concentration convection. The analysis performed by Müller and Lücke [4] constitutes the first theoretical approach concerning the non-linear evolution of the pattern and its stability *versus* Rayleigh number. Within the framework of their simplified model, these authors show that squares are not stable in the concentration regime but may be stabilized by a small sidewall forcing. On the other hand, they also show that thermal-like convection occurs when the Rayleigh number R_a exceeds its critical value and definitely leads to a roll pattern. They also address the transition from squares to rolls and find that the oscillation between the two sets of rolls constituting a square pattern is a pure non-linear effect.

Mixtures with a positive separation ration have at least two distinct fields of interest : the pure concentration convection and the competition between patterns when the thermal mode is also unstable. We have addressed these two fields by choosing a mixture with a large enough separation ratio so that they correspond to well separated Rayleigh number domain. Owing to the different nature of interest of these two fields, we have chosen to gather our experimental observations on each domain, with the referring discussion.

Experimental apparatus.

We have studied the convective patterns which appear in a mixture of methanol and a small concentration of carbon tetrachloride. We have chosen this mixture since the index difference between the two compounds is substantial, the Soret coefficient is known and does not vary very much with the concentration of carbon tetrachloride. Finally, owing to the large density difference between its two compounds, this mixture leads to a large separation ratio. The concentration of tetrachlorocarbon is 9.15 % in weight, according to Story and Turner [10] the Soret coefficient is $D_T/D = -3.3/T$ where T is the room temperature, which leads to a separation ratio $\psi = 0.32$. Although mixtures with positive separation ratio are less subject to systematic error in the determination of their Soret coefficient [11], the uncertainty on its value, and thus on ψ , may be as 50 %. The molecular diffusion coefficient D of tetrachlorocarbon in methanol equals $2.1 \times 10^{-5} \text{ cm}^2/\text{S}$, so that the Lewis parameter of the mixture is $L = D/\kappa = 0.02$ where κ is the heat diffusion coefficient.

Comparing to the ethanol water mixture, the methanol tetrachlorocarbon exhibits index variation with temperature which is several times greater. Moreover we are able to use a very thin cell which reenforces index gradients and decreases the characteristic time scales. The major drawback of this mixture is its chemical incompatibility with a lot of materials.

The fluid mixture is confined between two glass plates and a cylindrical cell made of kel-f, the sealing is achieved by a teflon ring surrounding the two glass plates. Two metallic washers are screwed so that they compress the teflon ring on the edge of the glass plates. The depth of the cell is defined by three spacers of 1.57 mm. The kel-f inner diameter is 80 mm leading to a very large aspect ratio $\Gamma = R/d = 25$. The temperature of the glass plates is determined by water circulations. We measure the temperature difference between the two plates using thermocouples, this temperature difference is not the one applied to the fluid layer since the glass plates have a temperature conductivity only 5 times greater than the fluid mixture. To determine the temperature difference really applied to the fluid layer requires a small computation : In the conductive regime and in the Soret regime, the convective velocities are null or extremely small and the relation between the total temperature difference and that applied to the fluid layer is. Each glass plate being 8 mm thick, we have $\Delta T^* = \Delta T/3$ (where ΔT^* is the temperature difference across the fluid layer). When thermal convection occurs, the heat conductivity of the fluid layer is enhanced so that we have used the prediction made by Schlüter [12] *et al.*, for the Nusselt number of square patterns to estimate this correction. We have used :

$$(\Delta T^* - \Delta T_{cl}^*)/\Delta T_{cl}^* = 0.68 \times (\Delta T - \Delta T_{cl})/\Delta T_{cl} \quad (1)$$

this relation is valid when $\Delta T > \Delta T_{cl}$ with $\Delta T_{cl}^* = \Delta T_{cl}/3$.

The water circulation channels are closed by glass windows which provide vertical optical access to the cell. We have used the shadowgraphic method to visualize the whole pattern of convection [13]. The pictures presented in this article have been obtained by this method with variable defocusing distances. Furthermore we have used a more quantitative measurement technique which enable us to record the components of the horizontal index gradient on either a line or a two-dimensional array of points covering 30 % of the cell surface [14]. This is achieved by scanning a laser beam using a pair of galvanometric mirrors, either on a line or on a grid of 64×32 points and recording at each point the light beam deviations by a position sensitive photodiode. The numeric images may be obtained in 1 second and may be considered as snapshot of the pattern. Using this measuring technique we have computed the temperature field in the cell and, performing the 2D Fourier transform, we have determined the wavevector of the convection pattern and the magnitude of the convective field.

Moreover the 2D Fourier transforms enable us to characterize regular patterns and to distinguish squares from rolls. The experimental procedure that we have followed consists in increasing or decreasing the temperature difference by small steps and after typically two hours, taking pictures and recording index gradient maps. The characteristic time of the molecular diffusion $d^2/\pi^2 D$ equals only 120 seconds so that the concentration gradient was always steady when we proceeded to the measurements. We have not encountered any noticeable hysteresis in our measurements of the amplitude of convection but the pattern overall organization has extremely long characteristic times.

Experimental results.

Depending on the temperature difference, we have found two types of convective regime differing by the intensity of the index gradient that they produce. The first regime which occurs at low temperature differences leads to weak index gradients. The second one leads to strong index gradients and appears above a well defined temperature ΔT_{c1} . Those two regimes may be easily seen in figure 1 where we have plotted the square of the index modulation *versus* the temperature difference. The two regimes are characterized by the very different slopes. The rapid change of slope occurs at the temperature difference ΔT_{c1} as may be seen in the graph of figure 1.

THE Soret REGIME. — Let us first consider the weak regime. Starting from the same temperature on top and on bottom plates, and increasing the temperature of the bottom plate leads first to the usual conductive regime where no convection is detected. But very soon, a weak cellular convective pattern appears as the temperature difference exceeds 0.5 K. We

Fig. 1. — Evolution of $S_i(k_0)$ corresponding to the amplitude of the convection pattern *versus* the temperature difference applied to the glass plates. $S_i(k_0)$ is determined using the 2D Fourier Transform and corresponds to the spectral density integrated on an annulus in reciprocal space, with a radius of k_0 and a width Δk_0 . The temperature difference ΔT used in the horizontal axis is not the one applied to the fluid layer, the relation is given in (1). Figure 1a shows the evolution of the amplitude of convection in the full range of ΔT . Due to the strong difference in the strength of the convection in both regimes, the concentration convection is barely visible, so that we have blown up this region of the graph in figure 1b. The different symbols correspond to various runs increasing or decreasing ΔT . $\Delta T_{c1} = 5.7$ K delimits the two convective regimes.

Fig. 2. — Shadowgraphic picture of the cellular pattern of convection in the regime where the concentration field is dominant, $\Delta T = 5.59$ K. Although the pattern contains defects, the squares are clearly visible. The index modulations induced by concentration gradients being smaller than those produced by thermal gradients, the contrast of this picture is rather poor.

present a picture of such a pattern in figure 2, the contrast is rather poor since the index gradients are particularly small but the modulations reveal a cellular pattern consisting mainly of square cells forming a texture. We must emphasize that the temperature difference in this weak convective regime is much too small to allow usual thermal convection to appear, we shall see that the threshold of pure thermal convection coincide with ΔT_{c1} . On the other hand, convection induced by concentration gradients may appear in a mixture and since the chemical diffusivity is fifty times smaller than the thermal diffusivity, this instability appears more easily. According to [15] this instability should appear when $\psi \cdot R_a / (L + 1) = 720$, where R_a is the usual Rayleigh number $R_a = \alpha g d^3 \Delta T / \kappa \nu$. If we assume that $R_a = 1\,708$ when $\Delta T = \Delta T_{c1}$ we find that the temperature difference at which the concentration convection should appear is ΔT_{c2} which equals $\Delta T_{c1}/17$ within our experimental conditions and coincide well with the exact computation of Zielinska and Brand [16]. ΔT_{c2} is thus rather small: 0.36 K leading to extremely small concentration modulations. To detect the occurrence of convection, we have preferred to use the quantitative index gradient measurements. We have recorded images of 32×64 points and averaged 16 of these images to increase the signal-to-noise ratio, we then compute the Fourier transform and obtain the image of the pattern in reciprocal space. In figure 3 we represent two typical examples of such images. In figure 3a the cellular pattern was obtained by increasing the temperature and the ring shape of the so-called Bragg peaks indicates that the structure is more powder-like. The pattern is indeed extremely disordered even on the scale of a few rolls. On the other hand figure 3b represents the reciprocal space of a cellular pattern obtained by decreasing the temperature and after annealing most of the structural defects at high temperature differences. The square structure is there obvious since four peaks appear with equal amplitudes and at ninety degrees apart. In both cases the Fourier transform presents an unambiguous proof of convection, we have computed the spectral power density $S(k, \theta)$ as a function of k and θ (where k is the wavevector and θ is the polar angle in the reciprocal space). When we could detect the convective motion, we have found that $S(k, \theta)$ always

Fig. 3. — Perspective and contour lines of the reciprocal spaces of the concentration convective motion patterns calculated by 2D FFT on a grid of 32×32 points. Both cases were obtained at the same temperature difference $\Delta T = 5.02$ K, but they correspond to a natural pattern extremely disorganized (Fig. 3a) and an annealed pattern (Fig. 3b). In this case, four peaks equal in amplitude and at ninety degrees, as expected for a perfect square pattern, are visible. The scale of figure 3b is reduced by a factor 5/9.

presents a well defined peak at a finite wavevector k_0 with a rather small width Δk_0 . In order to find the convection threshold experimentally we have measured $S_i(k_0)$, k_0 and Δk_0 as a function of the temperature difference. With :

$$S_i(k_0) = \int_0^{2\pi} \int_{k_0 - \Delta k_0}^{k_0 + \Delta k_0} S(k, \theta) k dk d\theta .$$

Figure 1b represents the evolution of $S_i(k_0)$ in this weak convective regime, as the evolution of k_0 and Δk_0 are reported in figure 4. In fact the accuracy of our measurements is not sufficient to determine the threshold of the concentration convection unambiguously. At 0.5 K the peak $S_i(k_0)$ disappears in the noise, and the only way we have to determine the actual threshold is to extrapolate the linear behavior of $S_i(k_0)$ to zero. The wavevector

Fig. 4. — Evolution of the wavenumber k_0 of the convective field, and of its width Δk_0 as a function of the temperature difference applied to the glass plates ΔT (the same remark as in Fig. 1 holds for this variable). At $\Delta T = 5.7$ K the wavevector is relatively close to the usual value of 3.117. Below 5.7 K, the wavenumber decreases gently to reach the value 1.5 at the lowest temperature difference where we are able to detect the occurrence of the convective motions.

k_0 decreases when we approach the convective threshold ΔT_{c2} , roughly it follows a linear behaviour with ΔT reaching a value very near k_c when $\Delta T = \Delta T_{c1}$. When $S_i(k_0)$ disappears in the noise, k_0 is approximately equal to $k_c/2$. The wavenumber distribution Δk_0 is usually rather small but tends to increase when ΔT approaches ΔT_{c2} . The wavenumber measurements depend slightly on the fact that they were made while increasing or decreasing the temperature difference, more precisely $S_i(k_0)$ is fairly insensitive but k_0 is smaller when ΔT is increased than when ΔT is decreased, the difference never exceeds 30 %.

The study of this concentration convection is motivated by the fact that this kind of convection leads to square cells instead of the usual roll pattern in the pure thermal convection case with good thermal conductor on top and bottom. Actually we shall demonstrate that the squares induced by the concentration convection did persist over some range of temperature difference above ΔT_{c1} before the usual roll pattern appears. The existence of square pattern for concentration convection was expected since the boundary condition for the concentration modulations is precisely the same on that the temperature experiences when top and bottom plates are nearly insulating. In such a case the existence of a square pattern has been demonstrated theoretically [9] and experimentally [8]. However the pattern wavevector is predicted to be zero or of the order of $1/\Gamma$ where Γ is the aspect ratio of the container, in the condition where the boundary conditions are really $\partial T/\partial z = 0$ [9]. The first experiment where squares were observed [2], the following investigations by Steinberg and Moses [3] and this study all confirm the existence of the square pattern but with a wavevector of order 3.0 in the first two cases and between 1.5 and 3 in this study. These values are very similar to the usual value of 3.117 observed for thermoconvective rolls with good thermal boundary conditions. If we consider the results that we present in figure 4, it is true that we cannot determine the wavevector just at the threshold of the concentration convection. At this threshold, expected around 0.4° , we have measured that $k_0 \approx k_c/2$ for a temperature difference only two times larger. This implies that the k_0 should drop from this

value to nearly 0 on this small temperature difference range and indeed such a sudden drop contrasts very much with the gentle evolution of k_0 with ΔT that we have observed all the way up to ΔT_{c1} (see Fig. 4).

A possible explanation was proposed by Moses and Steinberg who present a diagram, figure 4a of reference [3], where they claim to see a single convective cell in their large container when the temperature difference just exceeds the threshold of concentration convection. They propose that this form of convection is replaced by convective cells having a wavevector nearly 3 when the temperature difference approaches ΔT_{c1} . In this description a transition between the two convective regimes should occur for a temperature difference smaller than ΔT_{c1} but larger than ΔT_{c2} ; such a transition has not been reported so far. More recently Lhost and Platten [17] have performed an experiment in a long and narrow cell with a mixture of water and isopropanol having a positive $\psi = 0.38$. They use a Laser Doppler Velocimeter to measure the fluid velocity locally. They observe that just above ΔT_{c2} the horizontal velocity component of the fluid becomes significant and by scanning their measuring point they show the existence of a large roll extending all along the cell. Unfortunately these authors have not studied the convective pattern appearing at ΔT_{c1} and especially the square-to-roll transition so that the issue of a competition of patterns having two very different wavelengths or a wavenumber selection is not resolved. We wish to emphasize that such an issue is a challenge for experimental techniques since none of those currently used in convection is well adapted to characterize patterns having small and large wavelengths at the same time : optical techniques such as shadowgraph or index gradient measurements are sensitive to a spatial derivative of the temperature modulation and thus are rather unsensitive to large cell detection. The Laser Doppler Velocimetry has not this limitation but is a local measurement difficult to use in characterizing the pattern especially if it is time dependent. We conclude that another kind of measurement is necessary in order to detect unambiguously the nature of the pattern at the threshold of concentration convection.

A partial answer to this question was given by Giglio who has developed a very elegant way to detect accurately the threshold of the concentration convection by measuring the vertical index gradient [18] ; unfortunately this method does not reveal the pattern of convection. Nevertheless, by measuring the critical slowing down of the convective mode, Giglio infers that the wavevector must be of order $1/\Gamma$.

These various incomplete observations should challenge further investigations of the threshold of the concentration convection, one stimulation being the understanding of wavenumber selection. In the picture given by Moses and Steinberg, based upon linear theory, concentration convection should appear above ΔT_{c2} with $k_0 \cong 0$ and the thermal mode should appear above ΔT_{c1} with $k_0 \cong k_c$. When $\Delta T_{c2} < \Delta T < \Delta T_{c1}$, the strong concentration nature of the convection should lead to very small wavevector until $\Delta T = \Delta T_{c1}$ where a new thermal-like convective pattern should superimpose with $k_0 \cong k_c$. Our experiment supports a different picture, at least in the vicinity of ΔT_{c1} : as ΔT approaches ΔT_{c1} , the wavevector k_0 gently reaches k_c indicating that the wavenumber selection, which we have measured for the concentration like convection, is strongly determined by the occurrence of the thermal convection. This suggests that this wavenumber selection should be very sensitive to the ψ value of the mixture.

THE THERMAL CONVECTION REGIME. — Above ΔT_{c1} thermal convection occurs, leading to stronger index modulations, as illustrated in figure 1, $S_i(k_0)$ evolves linearly until it reaches the highest temperature difference that we have investigated, that is 15 K. At this point the pattern appears like the usual Rayleigh Bénard rolls as may be seen in figure 5. This structure contains a few defects, dislocations and grain boundaries but the square tendency has nearly completely disappeared and this pattern corresponds to the usual thermal convection. The

Fig. 5. — Shadowgraph picture obtained at $\Delta T = 2.79 \times \Delta T_{c1}$, ($\Delta T = 15.93$ K), the pattern is now clearly roll like and resembles very much patterns obtained in pure fluid convection. Nevertheless a faint square tendency is still visible.

main motivation of this work was to investigate the square-to-roll transition and its associated dynamics. In similar situations, oscillations between squares and rolls were observed [2, 3] as a way to switch from squares to rolls. As we shall show, we have observed here a different behavior : first, probably related to our larger value of ψ , the square tendency remains until large temperature differences and really disappears when we reach $2.5 \times \Delta T_{c1}$. In all the temperature range between ΔT_{c1} and $2.5 \times \Delta T_{c1}$ we have found that the pattern was time dependent and topologically unstable. Although we cannot completely exclude the fact that this time dependence was related to oscillations between sets of rolls, as already observed [2, 3], we have evidenced another mechanism related to the existence of traveling waves in patches of rolls. These traveling waves resemble very much those observed in binary mixtures with negative separation ratio [6], except that they are much slower in our case. Nevertheless this kind of time dependence leads to texture-like patterns and indeed all our attempts to produce defect-free patterns have failed since grain boundaries seem to nucleate spontaneously connecting patches of rolls with an inside square tendency. One typical example of such patterns is given in figures 6 and 7. In the first figure we show the entire pattern whereas in the second we have tried to present the pattern time evolution using four consecutive numerical images, this time scale is of the order of a few hours. The instability of these patterns renders their study difficult. During one experiment we have obtained a relatively large patch of rolls nearly filling up the area scanned by our laser beam : all the data that we present concerning the relative amplitudes of the two perpendicular sets of rolls correspond to this peculiar pattern. On the other hand the determination of $S_i(k_0)$ and k_0 does not require a perfect pattern and was done on either highly or weakly disordered patterns.

Let us first present our results concerning $S_i(k_0)$ and k_0 . The occurrence of thermal convection appears smoothly, as may be seen in figure 1, $S_i(k_0)$ increases regularly and only a change of slope indicates this occurrence. The pattern evolves gently growing from the square cells of the concentration mode, no hysteresis appears in either $S_i(k_0)$ or k_0 , provided that we wait for the pattern relaxation. The behavior of k_0 with ΔT is noteworthy : as we have already mentioned in the Soret regime, k_0 increases so that when ΔT_{c1} $k_0 \cong k_c$, the critical value usually observed in pure fluid convection, afterwards k_0 remains nearly constant.

Fig. 6. — Shadowgraph picture obtained at $\Delta T = 1.58 \times \Delta T_{cl}$, ($\Delta T = 9.04$ K) illustrating the square to roll competition.

Fig. 7. — Reconstruction of the temperature field obtained from the temperature gradients observed at $\Delta T = 10.63$ K. Figures 7a, b, c, d, are consecutive pictures separated from each other by 750 seconds. They illustrate the time evolution of a disordered roll pattern with a strong square tendency. The reorganization through the motion of grain-boundary is easy to see, the traveling motion of patches of rolls is present but less obvious to realize.

The characterization of a disordered and time dependent pattern is not simple, and we have focused on two points : the square tendency and the traveling rolls' behavior. We have expressed the square tendency by the ratio $\nu = (A^2 - B^2)/(A^2 + B^2)$, where A^2 is the square amplitude of one set of rolls and B^2 the square of the amplitude of the orthogonal set of rolls. The choice of the numerator is arbitrary in the sense that it could also be the opposite, thus ν and $-\nu$ have the same meaning except that the dominant roll set is not in the same direction. In the Soret regime where $\Delta T \leq \Delta T_{c1}$, ν is equal to 0. At high temperature difference, in the Rayleigh regime, the ratio ν is either close to 1 or close to -1 . We observe that this limit is reached when $\Delta T \approx 2.5 \Delta T_{c1}$. The ratio ν has a continuous behavior as a function of the temperature difference has, but presents a characteristic bifurcation at $\Delta T_{c3} = 8.5^\circ$, as may be seen in figure 8. At this temperature difference $A^2 - B^2$ increases linearly with ΔT and very soon approaches $A^2 + B^2$. In this experiment we have not observed any periodic exchange between the two sets of rolls as reported in [2, 3]. No pure and continuous oscillations could be observed since the pattern became rapidly disordered with many defects involved.

Fig. 8. — Evolution from square to roll, the diamond represents $A^2 - B^2$ and the crosses $A^2 + B^2$. Until $\Delta T = 8.5$ K the pattern is made of pure squares while a bifurcation clearly occurs and leads to a progressive transition to rolls.

Fig. 9. — Space and time evolution of the temperature gradient recorded along a line in the pattern, this picture clearly demonstrates the existence of a traveling roll pattern, $\Delta T = 7.97$ K. Rolls parallel to the measuring line give rise to the apparently erratic motions on each side of the recording.

For temperature difference above ΔT_{c1} we have observed the existence of traveling rolls. In order to characterize this phenomenon, we have measured periodically in time the temperature gradient along a single line during a long period of time. The scanning line was chosen to be perpendicular to a large patch of rolls at the beginning of the recording. As may be seen in figure 9, the rolls slowly and homogeneously move perpendicular to their axis. The typical frequency is $\omega_0 = 2 \times 10^{-3}$ radians per second. We have tried to follow these oscillations while decreasing the temperature difference, however we have not measured significant variation of ω_0 , because the nucleation of defects, the occurrence of squares and the pattern rotation make any quantitative measurements impossible, especially near

$\Delta T \approx \Delta T_{c1}$. In figure 7, we present this time dependent state by four consecutive snap shots, the evolution of grain boundaries is easy to see, the traveling motion of the rolls is less obvious but exists. However all our recordings indicate that patterns are always time-dependent and that traveling rolls are always involved. The occurrence of squares prevent us from determining in which way the traveling patterns emerge from stationary patterns and how their frequency changes with the temperature difference.

Discussion.

The stability of rolls and squares has been recently considered by Müller and Lücke using a few modes Galerkin model in binary mixtures with positive separation ratios [4]. Their conclusions correspond well to the experimental observations, except for the Soret regime for which they found that rolls are the stable pattern. Although the stability of the rolls is weak and the instability of the square pattern is also weak, these authors argue that a small sidewall forcing could reverse the stability in experiments and lead to square pattern. We are not convinced by this argument, since sidewall forcing in a cylindrical cell will only favour rolls perpendicular to the walls. Our pictures demonstrate that the patterns feel this influence on a rather short distance, thus the square patterns are uncorrelated with the cell boundary. Moreover, the result of Müller and Lücke is in contradiction with that of Busse, Riahi and Proctor [9]. We believe that squares are stable in the Soret regime and that it might well be that a more sophisticated model will also lead to this conclusion. The moderate thermal conductivity of the top and bottom boundary used in this experiment is not low enough to stabilize square patterns for pure thermal convection. Our observations of square patterns can only been explained by concentration convection. The oscillating regime between square and roll is difficult to compare with the experiments described in this paper since we have not been able to observe regular oscillations. However, the oscillations found by Müller and Lücke are certainly those observed in references [2] and [3]. On the other hand, Müller and Lücke found that, at high Rayleigh number, only rolls are stable. This is in reasonable agreement with our experiment but we observe that the rolls are not pure, and a clear orthogonal roll set persists on a wide Rayleigh number range. This observation does not correspond to the prediction.

In another study, Linz, Lücke, Müller and Niederländer [1] study a very similar model but intended to describe traveling wave solutions. Although their main results concern the case of negative separation ratios, they also consider with some interest the positive ψ case and find that traveling rolls should also be observed. Traveling rolls should emerge from the stationary roll pattern slightly above the Rayleigh regime. Our observations coincide with this prediction in a qualitative way, a quantitative comparison being impossible due to the occurrence of squares and the strong tendency to get disordered pattern. The existence of traveling waves for positive separation ratio was a surprising and interesting prediction of Linz *et al.* Our experimental observations confirm the existence of these traveling waves even though a detailed comparison is not possible now. This point is important since the Lorenz model used in [1] and [4] may not be valid above ΔT_{c1} , where the high velocity of the convective motions mixes up the fragile concentration gradient, shrinking them only to small boundary layers [19]. The origin of these traveling rolls is an open question. They could be studied experimentally using an annulus cell small enough in the radius direction so that only rolls could develop. Theoretically, the possible connection with phase instability mechanism is a challenging question. The pattern selection has received a special interest recently confirming the stability of squares in our experimental conditions [20].

Conclusion.

We have studied convective patterns in a binary mixture with a positive separation ratio. In

the Soret regime, we have confirmed the existence of square cells. Our measurements indicate that the wavenumbers involved in the square patterns are smaller than the one usually encountered for thermal convection but are not very small as predicted using linear theory. Furthermore, we observe a continuous evolution of this wavenumber until the threshold of the thermal convection is reached. In the thermal regime, we observe that the rolls are the stable pattern. However the transition from square to rolls appears to be very progressive and the square tendency remains on a large range of temperature differences. Finally, we demonstrate that, in the same temperature difference range, the rolls are unstable to traveling rolls as predicted recently.

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