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The wavenumbers of supercritical surface-tension-driven Bénard convection

By E. L. KOSCHMIEDER AND D. W. SWITZER
College of Engineering and I. Prigogine Center for Statistical Mechanics, The University of Texas at Austin, Austin, TX 78712, USA

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The cell size or the wavenumbers of supercritical hexagonal convection cells in primarily surface-tension-driven convection on a uniformly heated plate has been studied experimentally in thermal equilibrium in thin layers of silicone oil of large aspect ratio. It has been found that the cell size decreases with increased temperature difference in the slightly supercritical range, and that the cell size is unique within the experimental error. It has also been observed that the cell size reaches a minimum and begins to increase at larger temperature differences. This reversal of the rate of change of the wavenumber with temperature difference is attributed to influences of buoyancy on the fluid motion. The consequences of buoyancy have been tested with three fluid layers of different depth.

1. Introduction

As is well known the hexagonal convection cells observed by Bénard (1900) were actually driven by surface tension gradients as was established experimentally by Block (1956) and explained theoretically by Pearson (1958). Pearson's linear theory assumes that the convective motions are driven exclusively by surface tension forces without a contribution of buoyancy, that means that the acceleration of gravity was set to zero. Onset of surface-tension-driven convection is determined by the critical Marangoni number and the cells are characterized by a unique critical wavenumber. The combined surface-tension-driven and buoyancy problem was studied by Nield (1964). Onset of convection in this case is determined in thin fluid layers by the critical Marangoni number and in deep fluid layers by the critical Rayleigh number, and the Marangoni number and Rayleigh number for onset of convection by surface tension gradients in the presence of buoyancy are coupled by a simple equation. The convective motions are again characterized by a unique critical wavenumber. As is well known, linear theories do not permit a determination of the preferred planform of the convective motions which all experiments show to be hexagonal.

Nonlinear theories dealing with supercritical surface-tension-driven convection in fluid layer of infinite horizontal extent have been published by Scanlon & Segel (1969), Kraska & Sani (1979) and Cloot & Lebon (1984). Supercritical surface-tension-driven convection in small circular containers has been studied by Rosenblat, Davis & Homsy (1982). The papers dealing with supercritical surface-tension-driven convection in infinite layers arrive at the conclusion that the hexagonal pattern is the only stable supercritical pattern and hence preferred. Only Cloot & Lebon make a statement about the wavenumber of the supercritical cells. They predict that the wavenumber of the supercritical motions is larger than the critical wavenumber and is non-unique, in the case where the Rayleigh number is zero, i.e. in the absence of buoyancy. On the other hand, the wavenumbers are predicted to be smaller than the critical wavenumber and non-unique for values of the Rayleigh number $400 < R < 500$, that means when buoyancy is present but small.

Little is known from experiments about the variation of the size of the hexagonal convection cells in surface-tension-driven convection when the temperature difference or the Marangoni number is increased above critical. The best information about this can be found in Bénard's (1900) original paper, where it is shown in figure 23 that the size of the cells in very thin and very wide layers varied with the applied temperature difference from larger cells at large ΔT, to a minimum size at moderate ΔT, to a somewhat larger size again at the critical temperature difference. The concept of the critical temperature difference was not known to Bénard but is apparent from the fact that the cells disappeared at this ΔT when ΔT was decreased. The only way Bénard could vary the temperature difference across the fluid was by permitting the metal block at the bottom of the layer to cool down from an initial temperature of 100 °C, thus ΔT decreased in his experiments. To our knowledge, Bénard's observation of the variation of the cell size has not been mentioned in the modern literature although his observations were discussed in a review of the Bénard problem by Koschmieder (1974). Dauzère (1912) published convincing pictures showing that the size of hexagonal convection cells increased when the applied temperature difference was increased above the largest ΔT used by Bénard. This confirms the trend towards larger convection cells with increased ΔT reported by Bénard. Since the cell size is inversely proportional to the wavenumber and since Bénard decreased the applied temperature difference, whereas we increase the temperature difference, we shall here, in order to avoid confusion later on, state Bénard's observation as follows. His observations showed that the wavenumber of the motions increases when after onset of convection the temperature difference is increased, the wavenumber reaches a maximum, and decreases substantially when ΔT is increased further.

The increasing cell size of surface-tension-driven convection observed at large ΔT by Bénard and Dauzère follows the same trend as the increase of the cell size in Rayleigh–Bénard convection. This has been observed in many experiments which have been reviewed by Koschmieder (1974). The increase of the cell size of surface-tension-driven hexagonal convection cells has recently been confirmed by Cerisier *et al.* (1987*b*). The observed increase of the hexagonal cell size seems to contradict Bénard's observation that the cell size had a minimum from which the cell diameters increased with decreased ΔT until they disappeared at the critical condition. We will prove in the following that Bénard's observation of an initially decreasing cell size was correct, and that the decrease of the cell size under slightly supercritical conditions is characteristic for the variation of the cell size of surface-tension-driven convection. We will also show that the supercritical cell size of surface-tension-driven convection is unique within the experimental error.

2. The apparatus

The apparatus is a modified version of the apparatus used by Koschmieder & Biggerstaff (1986, hereinafter referred to as KB) for the investigation of the onset of surface-tension-driven convection. The bottom of the fluid layer was a chromium coated silicon crystal 12.7 mm thick and 17.8 cm in diameter. The surface of the coated crystal was plane to 20 wavelengths and served as the mirror for the shadowgraph visualization. The silicon crystal is an excellent thermal conductor, its

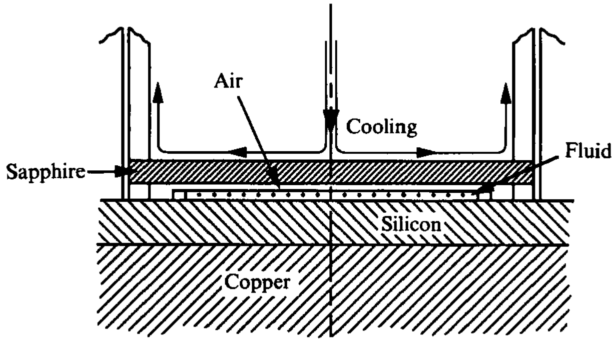

FIGURE 1. Schematic drawing of the essential part of the apparatus.

thermal conductivity is $\lambda = 0.37$ cal/cm s °C, that is $\frac{1}{3}$ of the thermal conductivity of copper ($\lambda = 0.94$ cal/cm s °C). The silicon crystal was heated from below by a copper block, 5 cm thick and 17.8 cm in diameter. The copper block was heated in turn by a regulated electrical current passing through a resistance wire. The arrangement with the silicon crystal and the copper block should provide a practically uniform temperature at the surface of the crystal, by the excellent thermal conductivities of the materials and the thickness of the copper block, in combination with the very poor thermal conductivity of the silicone oil (which is of order of 4×10^{-4} cal/cm s °C) on top of the crystal.

The silicone oil layer on top of the mirror was bounded by lucite rings of 10.48 cm inner diameter and different thicknesses, ranging from 3 to 1.2 mm machined with an accuracy of order of 10^{-2} mm. The nominal thermal conductivity of lucite differs from the thermal conductivity of the silicone oil by only about 10%. On top of the fluid layer was a thin air gap of 0.4 mm depth. The thickness of the air layer was held as low as possible in order to suppress motions of the air on top of the fluid. The air was contained above by a sapphire lid of 5 mm thickness and 13.34 cm diameter whose top and bottom surfaces were polished and optically plane. Koschmieder & Pallas (1974) used such a sapphire for the first time in a convection experiment. The purpose of the sapphire is to provide a transparent lid of good thermal conductivity, which is $\lambda = 0.088$ cal/cm s °C according to the manufacturer (Union Carbide). The thermal conductivity of the sapphire is about 200 times better than the thermal conductivity of the silicone oil. The sapphire was cooled from above by water of constant temperature ($\pm 10^{-2}$ °C) circulated at a rate of about 60 cm^3/s. By the uniform temperature of the lid and by the small depth of the air gap the temperature of the air on top of the fluid was made practically uniform, as is required by theory. The sapphire was held in place by a lucite ring surrounding the ring containing the fluid. The essential features of the apparatus described above are shown in figure 1.

The experimental set-up complies in a very good approximation with the ideal assumptions usually made in theory. These are uniform temperatures on a plane perfectly-conducting bottom as well as uniform temperatures on top of the fluid layer, which is, however, in surface-tension-driven convection necessarily of poor thermal conductivity because air is a very poor thermal conductor. Another assumption usually made in theory is that the fluid layer is of infinite horizontal extent. This assumption cannot be fulfilled in an experiment. This condition has been approximated by the large aspect of the fluid layers, the maximal value of which was 87. The aspect ratio is defined as the ratio of the diameter of the fluid layer divided

ν (cm²/s)	ρ (g/cm³)	α (°C⁻¹)	λ (cal/cm s °C)	κ (cm²/s)	S (dyne/cm)	dS/dT (dyne/cm °C)
1.00	0.968	0.00096	3.7×10^{-4}	0.001095	19.36	−0.050
0.50	0.960	0.00104	3.6×10^{-4}	0.001025	18.35	−0.047

TABLE 1. Properties of the fluids at 25 °C

by the fluid depth. The consequences of the presence of the lateral boundary were always noticeable.

The temperature differences applied to the fluid layers are expressed in non-dimensional form by the Marangoni numbers

$$M = \left(\frac{dS}{dT}\right)\frac{\Delta T d}{\rho\nu\kappa}, \tag{1}$$

and the Rayleigh numbers

$$R = \frac{\alpha g \Delta T d^3}{\nu\kappa}, \tag{2}$$

where dS/dT is the variation of the surface tension coefficient S with temperature, ΔT the temperature difference applied to the fluid layer, d the depth of the fluid, ρ the density of the fluid, ν the kinematic viscosity, κ the thermal diffusivity and α the expansion coefficient of the fluid, with g being the acceleration due to gravity. The properties of the two fluids used are listed in table 1. The onset of convection of purely surface-tension-driven convection under an insulating surface takes place at $M_c = 79.61$ according to Nield (1964) with the critical wavenumber being $a_c = 1.993$; the onset of purely buoyancy-driven convection under an insulating surface takes place at $R_c = 669.0$ with the critical wavenumber being $a_c = 2.086$, according to Nield too. We note that according to Pearson, as well as Nield, the critical wavenumber changes when the upper surface is not insulating, and can increase by 50% if the upper surface is conducting in the surface-tension-driven case. In the experiments to be described the upper surface condition is nearly insulating, that means that the Biot number is near zero.

The actual temperature differences applied to the fluid have to be calculated from the temperature difference between the copper bottom plate and the temperature of the cooling water on top of the sapphire because no temperature sensor can be placed in the narrow air gap or on the fluid surface itself without introducing a disturbance. The temperature difference across the fluid follows from the formula

$$\Delta T_{fl} = \frac{\Delta T}{\left(1 + \frac{\bar{\lambda}_{fl}\Delta Z_a}{\bar{\lambda}_a \Delta Z_{fl}}\right)}, \tag{3}$$

where ΔT is the temperature difference between the top of the sapphire and the copper block, $\bar{\lambda}_{fl}$ the mean thermal conductivity of the fluid averaged over the temperature difference applied to the fluid, $\bar{\lambda}_a$ the mean thermal conductivity of the air and ΔZ_{fl} and ΔZ_a are depth of the fluid layer and the depth of the air. After the onset of convection the vertical temperature distribution in the fluid is no longer linear, that should, however, have no influence on the value of $\bar{\lambda}$. Of great importance for the determination of ΔT_{fl} is the contribution of the air layer, because the thermal conductivity of air ($\lambda = 6.1 \times 10^{-5}$ cal/cm s °C at 20 °C) is so poor. More than half of the temperature decrease between the sapphire and the copper plate occurs in the air layer, although it is so thin. The contribution of the silicon crystal and the sapphire to the temperature decrease are negligible. The temperature difference ΔT_{fl} determined this way is not known with an accuracy better than $\pm 5\%$, mainly because the depth of the fluid layer is not known better than 1%, and the depth of the air layer is not known better than 2%.

3. The experiments

The first series of experiments were made with a fluid layer of 1.90 mm depth. It became apparent immediately that very regular patterns formed, similar to those in figure 2, and that the number of cells increased if the temperature difference was increased in the slightly supercritical range. An increase of the number of cells is equivalent to a decrease of the cell size or an increase of the wavenumber of the motion. Since in Rayleigh–Bénard convection the wavenumber of the convective motions tends to be larger than the critical wavenumber if the heating is time-dependent it was decided to eliminate the possible consequences of time dependence of heating extremely slowly. This is also of importance for the pattern formation because the fluid needs a long time to establish equilibrium in the horizontal plane. This time is characterized by the horizontal relaxation time D^2/κ, which is of order of 28 h. In all experiments described in the following the heating was increased in two steps during a day so that the temperature increase across the fluid was about 0.5 °C per day. That meant that the experiments had to be run continuously for a couple of weeks in order to reach a maximal temperature difference of about 30 °C across the fluid and the air gap, which temperature difference was deemed to be safe for the sapphire and the silicon crystal.

The series of quasi-steady experiments was first made with a lateral ring of 1.90 ± 0.01 mm depth, filled to the rim with silicone oil of 1.00 cm^2/s viscosity. Filling the layer to the rim of the ring eliminates undesirable effects of a meniscus. The depth of the fluid was, however, not known to an accuracy better than ± 0.02 mm, because of the accuracy with which the fluid depth can be measured with a micrometer. Figures 2 and 3 show shadowgraph pictures obtained with such a layer, and figure 4 shows the increase of the number of cells in the layer when the temperature difference was varied. Each point in figure 4 originates from a shadowgraph picture taken 12 h after the last increase in heating. The number of cells is proportional to the wavenumber a of the motions, which for hexagonal cells is given by $a = 4\pi d/3L$, where L is the side length of the cells and d is the depth of the fluid. The wavenumber as given by the number N of the hexagonal cells and the area A of the fluid layer follows from $a = 4\pi d(2.5981N)^{\frac{1}{2}}/3A^{\frac{1}{2}}$. According to this formula, the wavenumber of the fluid motions is proportional to the number of cells in the fluid layer, and the size of the cells A/N is inversely proportional to the wavenumber.

The onset of convection was observed at $\Delta T_c = 7.1 \pm 0.1$ °C. The onset is, however, not an unambiguous spontaneous event, the shadowgraph picture is initially weak with a substantial background of diffuse white light. It is not possible to determine the critical condition with an accuracy better than ± 0.1 °C. The white lines outlining the hexagonal cells in figure 2 indicate cold fluid, because cold fluid focuses the parallel light with which the mirror bottom of the fluid is illuminated from above. The shadowgraph pictures have been discussed in detail by Jenkins (1988), the hexagonal pattern in figure 2 agrees with figure 5(a) calculated by Jenkins for hexagonal cells. The shadowgraph technique is the most suitable visualization for

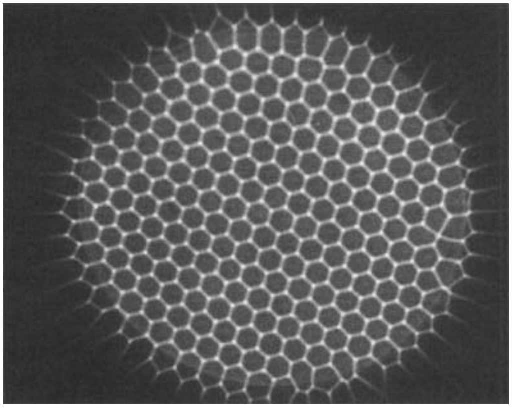

FIGURE 2. Shadowgraph picture of the cellular pattern in the 1.90 mm deep layer of 1.00 cm^2/s viscosity at 1.24 $\Delta T_{c\,exp}$. 200 cells.

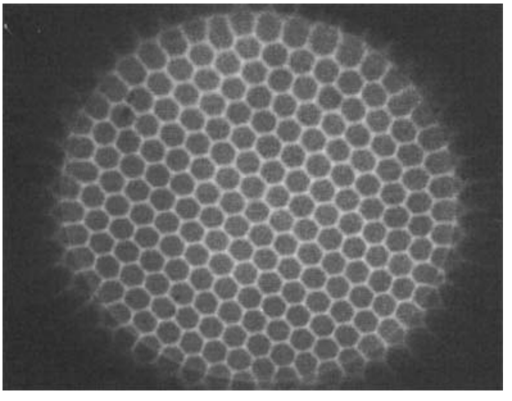

FIGURE 3. Shadowgraph picture of the cellular pattern in the 1.90 mm deep layer of 1.00 cm^2/s viscosity at 1.50 $\Delta T_{c\,exp}$. 217 cells.

our experiments because it permits us to do the experiments with the clear fluid whose surface-tension coefficient is not altered in any way. Note the consequences of the lateral wall on the cells near the rim in figure 2. Of course, these effects cannot be eliminated entirely. The fact that the cells at the rim apparently extend beyond the rim seems to be an optical illusion brought about by refraction of the light in the fluid adjacent to the wall, where the uniformity of the temperature is altered by the different thermal conductivities of the silicone oil and the lucite ring. This effect increases as ΔT is increased.

Increasing ΔT does not only increase the number of cells and the contrast in the shadowgraph, but also changes the shadowgraph picture of the hexagonal cells, see figure 3. The first sign of change was the appearance of bright dots at the vertices of the cell boundaries, as in figure 3. These dots gradually increase in size when ΔT is increased and the lines connecting the vertices gradually increase in width, although the dots at the vertices remain individual dots of greater brightness. We believe that these changes reflect the changes of the temperature field in the cells as the conditions become increasingly nonlinear.

When ΔT approached the maximal ΔT used in our experiments the number of cells seemed to become independent of ΔT, see figure 4. When the maximal ΔT applied

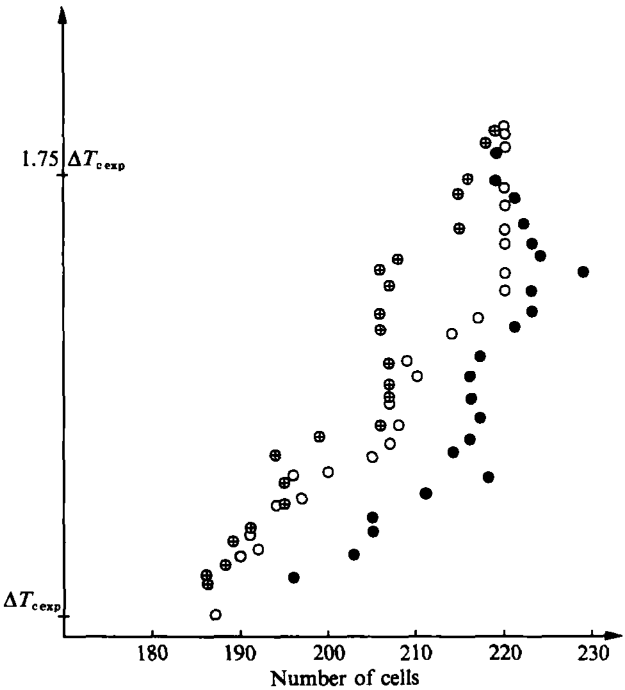

FIGURE 4. The variation of the number of cells in the 1.90 mm deep layer of 1 cm^2/s viscosity. ○, the first increase of ΔT; ●, the following decrease of ΔT; ⊕, the second increase of ΔT.

was reached, the temperature difference was held constant for two days, and afterwards ΔT was decreased just as slowly as it had been increased, in order to see whether there is hysteresis in the wavenumber of the motions. In general the number of cells in the layer then decreased as is shown in figure 4, but the points for decreasing ΔT do not coincide with the points for increasing ΔT. There is usually a small gap between corresponding cell numbers. Directly after the decrease of ΔT was begun the number of cells actually increased with decreased ΔT. These fluctuations in the number of the cells seem to be caused by wall effects. The cells which come into the fluid layer when ΔT is increased do not form as individual additional cells in the interior of the layer, but form in small groups at the rim, where the largest nonconformity of the vertical temperature gradient occurs. Likewise the cells that disappear from the layer when ΔT is decreased do not tend to disappear individually. The interaction of the cells with the rim is different, depending on whether ΔT is increased or decreased. The difference in the number of cells for increasing or decreasing ΔT at a given temperature difference is therefore not a consequence of hysteresis of the wavenumber of the motions, but the result of the boundary conditions at the rim. This will become even more obvious when we discuss the 1.2 mm deep layer.

In order to check on the reproducibility of the cell number as a function of ΔT, the same fluid layer whose wavenumber changes are plotted in figure 4 was heated up again after the pattern had disappeared at ΔT_c and had been held at a temperature difference 3° smaller than ΔT_c for two days. The pattern reappeared at a slightly smaller ΔT_c with a slightly smaller number of cells as shown in figure 4, but these differences result from the difficulties in pinpointing exactly the critical condition through the shadowgraph. Increasing ΔT again in the same slow way as used before produced a much more unsteady increase in the number of cells as compared to the results of the first temperature increase. There was now in particular an extended range in ΔT over which the cell number remained constant. However, in the end, very near the maximal ΔT, the number of cells was practically the same as it was the first time. The differences in the number of cells observed at the same ΔT after the first and second temperature increase only indicate the experimental uncertainty in the determination of the number of the cells. When the temperature difference was increased further, after the uppermost point of the second curve was taken, the fluid made contact with the sapphire lid. Virtually all experiments ended this way. When this happens the fluid apparently creeps across the lateral wall of the layer, up the ring carrying the sapphire and much of the fluid is then pulled by adhesion to the lid.

The qualitative results of this experiment are obviously that the number of hexagonal convection cells in this fluid layer increased as the temperature difference was quasi-steadily increased in the slightly supercritical range, that means that the wavenumber of the convective motions increased. It appears that the increase of the number of cells comes to an end at the upper end of the curves. It also appears that the number of cells or the wavenumber of the motions is independent of the initial condition, within the error of the experiment. These results are in agreement with the outcome of two other experiments increasing and decreasing ΔT quasi-steadily with the same fluid in 1.90 mm deep layers.

The onset of convection occurred in this fluid layer at a temperature difference $\Delta T = 7.1$ °C across the fluid. This corresponds to a Marangoni number $M = 61.8$, using the material properties listed in table 1. The Rayleigh number at the onset of convection was 40. The Marangoni number M_{theor} for onset of convection of the combined surface tension–buoyancy problem follows from the relation

$$\frac{M}{M_c} + \frac{R}{R_c} \approx 1, \tag{4}$$

according to Nield (1964). It follows that in this layer $M_{\text{theor}} = 74.8$, and therefore it is $M_{c\,\text{exp}} = 0.83 M_{\text{theor}}$. The implication of this will be discussed later. The wavenumber of the motions in the hexagonal cells follows from the area of the fluid layer, the depth of the fluid and the number of the cells, using the relation $a = 4\pi d/3L$, as stated before. With 185 cells at onset of convection we find $a_c = 1.85$, while the theoretical critical wavenumber under an insulating surface is $a_c = 1.993$, according to Nield (1964). The wavenumber found will be compared with the wavenumbers found in the other layers and with the theoretical critical wavenumber later on.

The qualitative results with the 1.9 mm deep layer raised the question of whether they do not contradict the observations of Dauzère (1912) and Cerisier *et al.* (1987 *b*). On the other hand they confirm the first part of Bénard's (1900) curve for the number of cells versus temperature difference. Since the increase of the number of cells in our experiment seemed to have come to a halt at the highest ΔT we worked with, it was decided to extend these experiments to higher Marangoni numbers. This can be accomplished by using a fluid of smaller viscosity. The results of two quasi-steady experiments made with silicone oil of 0.50 cm^2/s viscosity and 1.90 mm depth are shown in figure 5. It is apparent from this figure that the initial increase of the number of cells changes to a gradual decrease of the number of cells if the Marangoni number is made sufficiently large. This then is in agreement with the observations of Dauzère and also of Cerisier *et al.* in the case of large temperature differences or Marangoni numbers. Furthermore, figure 5 is in complete agreement with Bénard's observations, it shows an initial decrease of the cell size, a minimum of the cell size, and a subsequent increase of the cell size.

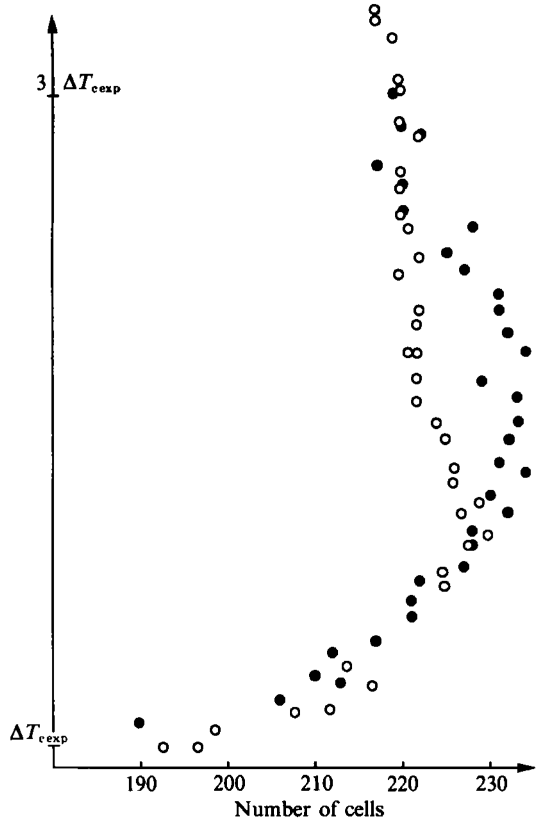

FIGURE 5. The variation of the number of cells in the 1.90 mm deep layer of 0.50 cm^2/s viscosity. ○, the first increase of ΔT; ●, a second experiment with increasing ΔT.

These observations raise the question of how it can come about that the variation of the wavenumber with temperature difference changes sign at a moderately supercritical temperature difference. There is, at present, no theory that would predict or explain this. We note however that the wavenumber of buoyancy-driven Rayleigh–Bénard convection decreases with increased Rayleigh number. We therefore assumed that the decrease of the wavenumber of the hexagonal cells in the upper part of figure 5 is induced by buoyancy, which is also present in our experiments because the Rayleigh number is not zero, and the value of the Rayleigh number increases with increased ΔT. A way to test this hypothesis experimentally is to vary the relative importance of the Rayleigh number, which can be easily done by varying the depth of the fluid layer. Since the Rayleigh number is proportional to d^3 while the Marangoni number is proportional to d, the importance of the Rayleigh number increases in deeper fluid layers. If the increase of the wavenumber of the hexagonal cells is caused, as assumed, by the contribution of buoyancy to our experiments, then the decrease of the wavenumber should appear sooner in deeper fluid layers and later, that means at higher Marangoni number, in thinner fluid layers.

The importance of the surface-tension effects should increase if the fluid depth is decreased. We have therefore determined the variation of the number of hexagonal cells in a 1.20 mm deep layer of 10.48 cm diameter. Since the critical temperature difference necessarily increases as the fluid depth is decreased we had to use for this

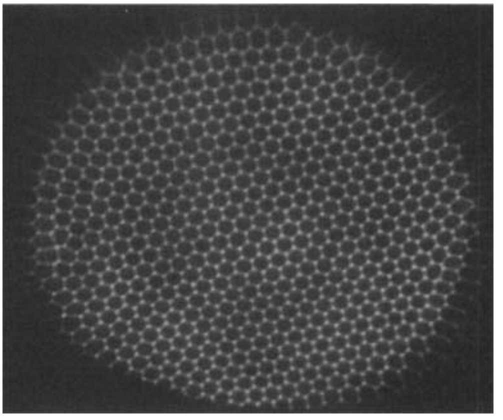

FIGURE 6. The cellular pattern in the 1.20 mm deep layer at 1.32 $\Delta T_{c\,exp}$. Note the lines of cells along the curved lateral wall. 511 cells.

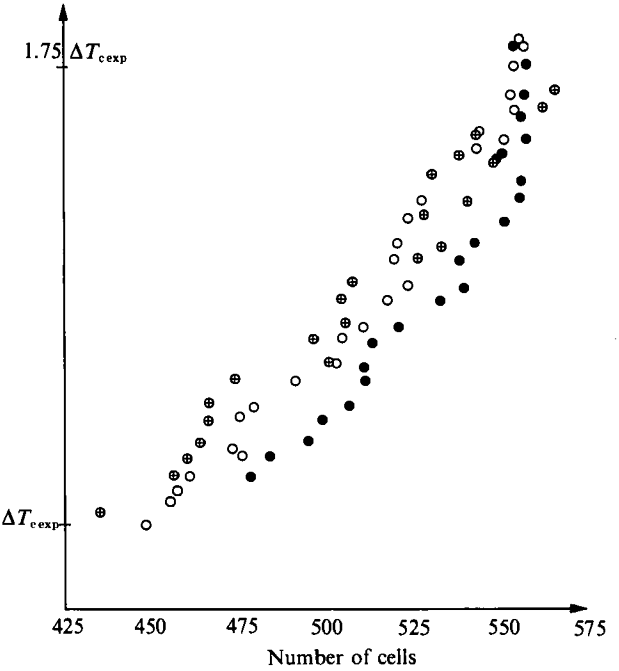

FIGURE 7. The variation of the number of cells in the 1.20 mm deep layer. ○, first temperature increase; ●, the following decrease of the temperature difference; ⊕, the second temperature increase.

experiment silicone oil of 0.50 cm^2/s viscosity. The aspect ratio for this layer was 87. Excellent patterns with 450 and more cells appeared, as shown in figure 6. When ΔT was increased, the number of cells increased, as in the 1.9 mm deep layer. The variation in the number of the cells with increased ΔT is shown in figure 7. In this experiment the temperature difference was quasi-steadily increased, later on quasi-steadily decreased to subcritical values, and then quasi-steadily increased again. In this layer with its increased aspect ratio the consequences of the lateral wall should be smaller than with the 1.9 mm deep layer. The influence of the aspect ratio on the critical wavenumber of hexagonal cells has been investigated experimentally by

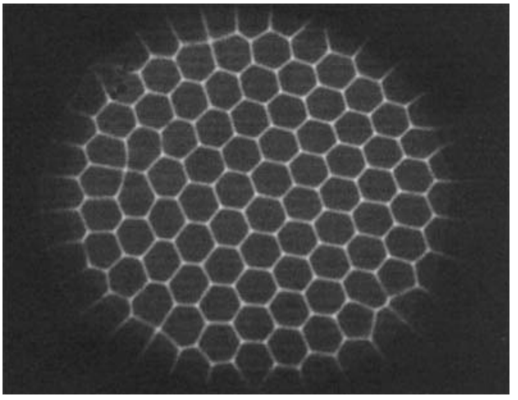

FIGURE 8. The cellular pattern in the 3.00 mm deep layer at 1.52 $\Delta T_{c\,exp}$.

Cerisier & Zouine (1989) who found that within the experimental error the critical wavenumber is independent of the aspect ratio. They also found the critical wavenumber at around 2.1, whereas the theoretical value is 1.993. The pattern in the 1.2 mm deep layer was noticeably affected by the lateral ring, forming long lines of cells along the ring, instead of individual cells adjusting their form to the shape of the wall. When additional cells came in as ΔT was increased they did not come in individually in the interior of the layer, but in groups or even lines at the lateral wall. The addition of entire lines of cells to the pattern was particularly noticeable when the layer was heated for the second time. This was reminiscent of the cell changes with the second heating in figure 4. Overall the scatter of the data in figure 7 is about $\pm 3\,\%$ and is smaller than the scatter of the data in figure 4.

As can be seen in figure 7 the number of the cells increased steadily over the range of ΔT we could work with. The data points end at the same maximal temperature difference $\Delta T = 30$ °C across the fluid layer and the air gap at which the experiments in figure 4 ended. Within the error of the experiment the number of cells appeared to be independent of the initial condition. This experiment shows that the increase of the number of cells (or the increase of the wavenumber) is more pronounced when the contribution of buoyancy to the convective motions is reduced in a less deep layer, or when the Rayleigh number is much smaller. The results of this experiment confirm the qualitative conclusion reached earlier that the cell size of surface-tension-driven hexagonal convection cells decreases in the slightly supercritical range, as long as there is no contribution of buoyancy to the motions.

Onset of convection in the 1.20 mm deep layer occurred at a temperature difference of 5.48 °C at a Marangoni number $M = 61.4$ and a Rayleigh number $R = 18.4$. The experimentally determined Marangoni number is therefore $M_{c\,exp} = 0.79\, M_{theor}$. The critical wavenumber of the pattern with 450 cells is $a_{ce} = 1.85$, the theoretical critical wavenumber for an insulating upper surface is $a_c = 1.993$. The slope of the variation of the wavenumber with the Marangoni number is $\Delta a/\Delta M = 0.00425$, assuming that the variation is linear over the entire range observed. We note that the variation of the wavenumber observed in our experiments does not depend on the Marangoni number only, but also on the Rayleigh number, because as the Marangoni number increases the Rayleigh number increases as well, because both numbers depend on the ΔT. Since the wavenumber $a = a(M, R)$ it follows that the $\mathrm{d}a/\mathrm{d}M$ we observe is given by $\mathrm{d}a/\mathrm{d}M = \partial a/\partial M + (\mathrm{d}R/\mathrm{d}M)\,(\partial a/\partial R)$.

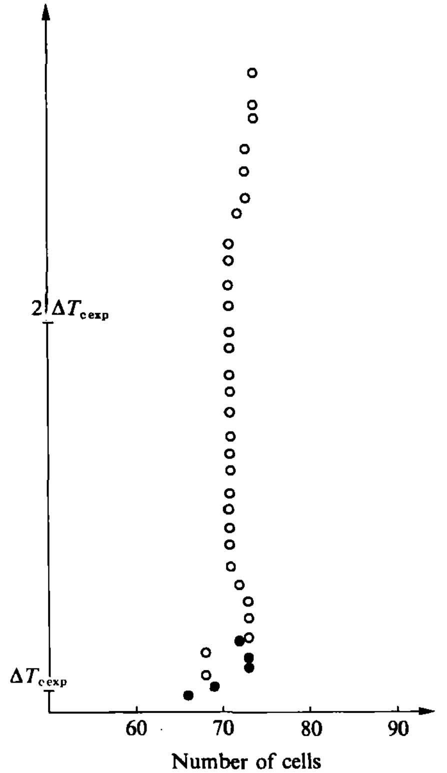

FIGURE 9. The variation of the number of cells in the 3.00 mm deep layer. ○, from an experiment with increasing ΔT; ●, from another experiment increasing ΔT, which ended when the fluid made contact with the lid.

The variation of the wavenumber of hexagonal convection cells was also determined in a 3.00 mm deep layer of silicone oil of 1.00 cm^2 viscosity of 10.16 cm diameter. Increasing the fluid depth decreases the aspect ratio, in this case to nearly 30. That, of course, reduced the number of cells significantly. However, the pattern remained clearly hexagonal as shown in figure 8. The variation of the number of cells with temperature difference is shown in figure 9. Except for a very small decrease of the number of cells near the onset of convection the number of cells remained virtually constant as ΔT was increased. The initial decrease of the number of cells was confirmed by another experiment, which ended when the fluid made contact with the lid. In an earlier experiment, Koschmieder (1967) found that the number of hexagonal cells in a fluid layer of 4.3 mm depth and 20 cm diameter increased from 123 to 126 when ΔT was increased to about 2.4 times the critical temperature difference. In this experiment the Rayleigh number at onset of convection was about $R = 230$, that means that buoyancy probably contributed to instability. The experiment with the 3 mm deep layer shows that at this depth the initial increase of the number of the cells had nearly disappeared, the experiment with the 4.3 mm deep layer shows that the fluid does not pick up the decrease of the wavenumber of buoyancy-driven convection very quickly when the fluid depth is increased.

Onset of convection in the 3 mm deep layer occurred at a temperature difference of 5.10 °C which corresponds to a Marangoni number $M = 71$. The Rayleigh number at onset of convection was 116, the relation between the experimental Marangoni number and the theoretically predicted Marangoni number M_{theor} is then

$M_{c\,exp} = 1.08 M_{theor}$. The wavenumber of the pattern consisting of 68 cells is $a_c = 1.86$. The critical wavenumber according to theory is $a_c = 1.99$ if the upper surface is insulting.

We summarize the results of experiments with the different fluid layers in table 2. As we see there, the observed critical wavenumbers of the experiments are in the range from 1.85 to 1.90. The experimental uncertainty of the wavenumbers is 2–3 %. It seems to be fortuitous that the value of $a_{c\,exp}$ is about 1.85 in three of the four layers. We attribute the 7 % difference between the experimental value 1.85 and the theoretical value 1.99 to the effects of the lateral wall. A small wavenumber means larger cells, and the cells at the wall are obviously larger than cells in the interior of the layer. We also note that a very large fraction of the number of cells is affected by the wall; in the 1.90 mm deep layer more than 20 % of the cells are in contact with the wall. We therefore believe that the observed wavenumbers at the onset of convection are in reasonable agreement with the theoretical prediction.

The results concerning the onset of convection in the different fluid layers are presented in table 2 in the column with the ratio M_{ce}/M_{ct}. There seems to be a decrease of this ratio as the depth of the fluid is decreased. But we note that we cannot expect the experimental Marangoni numbers to be more accurate than ± 10 % on an absolute scale. This is so because we do not know the value of $\mathrm{d}S/\mathrm{d}T$ with an accuracy better than ± 5 %, and we also do not know the value of ΔT with an accuracy better than a few per cent. This is so because ΔT has to be calculated with equation (3) in which ΔZ_{air} is only known to a few per cent, because the fluid depth is only known to about a thousandth of an inch (= 0.025 mm), which is a large fraction of the nominal depth of the air gap which is 0.4 mm. The experimental Rayleigh numbers are for similar reasons of equal uncertainty. On a relative basis, however, the ratios M_{ce}/M_{ct} should be much more accurate, because $\mathrm{d}S/\mathrm{d}T$ drops out, so the observed decrease of M_{ce}/M_{ct} seems to be fairly certain.

However, theory states that there should be no motion in the fluid at Marangoni numbers smaller than M_{ct}. But we have observed onset of surface-tension-driven convection at values below M_{ct} before in KB. Our current observations confirm to a surprising degree the experimental Marangoni numbers M_2 at which onset of convection in the form of hexagonal cells was found in KB. The onset of convection in a 2.57 mm deep layer of 1.00 $\mathrm{cm^2/s}$ silicone oil occurred then at $M_2 = 76.8$, in a 1.81 mm deep layer of 1 $\mathrm{cm^2/s}$ silicone M_2 was 61.0, and in a 1.34 mm deep layer of 0.50 $\mathrm{cm^2/s}$ silicone oil it was $M_2 = 68.0$. Although the depths of the layers are not the same as those here, the Marangoni numbers M_2 are similar for similar depth in table 2. There is certainly the same tendency for the ratios M_{ce}/M_{ct} to decrease with decreased depth. There is also the same apparent failure to continue the decrease of M_{ce} between 3 mm depth and 1.90 mm depth to the 1.20 mm layer that we see in table 2. However, the viscosity of the fluid in the 1.20 mm layer was only 0.50 $\mathrm{cm^2/s}$, and so was the viscosity on the 1.34 mm deep layer in KB. We do not need to expect complete agreement with the results of KB, because the heating in our present experiments was much slower than in KB and the visualization was different, and there are some subtle differences in the apparatus.

Finally, we note that in our present experiments onset of convection occurred at $M_{ce} = 1.08 M_{ct}$ in the 3 mm deep layer. According to theory, onset of convection must occur at M_{ct}. The difference between experiment and theory may simply be due to the experimental uncertainty in the determination of the absolute value of M. On the other hand, the difference in the critical value of M may be genuine, and may be caused by the upper boundary condition, which is not necessarily completely

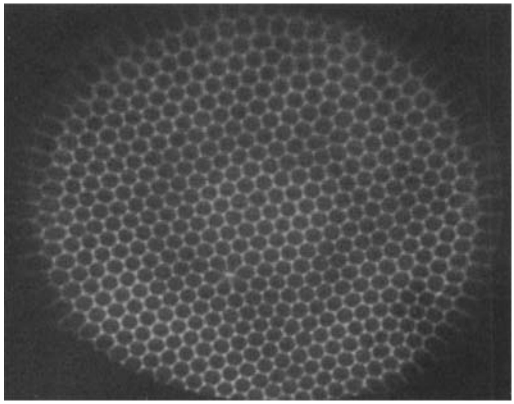

FIGURE 10. A defect on the cellular pattern in the 1.20 mm deep layer in the form of a star in the lower centre part of the picture at $\Delta T = 1.11\,\Delta T_{\mathrm{cexp}}$. 476 cells.

Depth (mm)	Viscosity (cm^2/s)	a_{ce}	M_{ce}	R	M_{ce}/M_{ct}	Aspect ratio
3.00	1.00	1.855	71.0	115	1.08	34
1.90	1.00	1.854	61.8	40	0.83	55
1.90	0.50	1.904	72.1	54	0.99	55
1.20	0.50	1.851	61.4	18	0.79	87

TABLE 2. Results of the experiments

insulating, as is the case when we set $M_{ct} = 79.6$. The thin air layer on top of the fluid which is covered by the well-conducting sapphire is not a good representation of an insulating upper surface. We have determined the value of the non-dimensional parameter L which describes conduction on top of the fluid layer in KB, which applies also for our present experiments because the set-up is essentially the same. We found that L is of order of 0.2 under these conditions. With $L = 0.2$ the critical Marangoni number is $M_c = 87$ according to Nield. Our experiments with the 3 mm deep layer are compatible with theory if we use $M_c = 87$ for the onset of convection.

Finally we want to discuss a characteristic defect of the cellular patterns which appeared in our experiments. Over the course of the experiments the patterns were usually very regular, with truly hexagonal cells dominating, except of course right at the wall. Naturally, imperfections of the patterns occurred. Such imperfections are referred to as defects. Defects have to be characteristic in order to be significant. A photograph with a defect characteristic for hexagonal cell patterns is shown in figure 10, where one can see what we call a 'star' in the lower part of the layer. Such a star can also be found on figure 3(*c*) of Cerisier *et al.* (1987*a*), where many other defects are discussed. A star is the combination of six deformed cells which have a common centre point, each of the six cells having only five vertices. There seems to be a point-like disturbance at the centre of the star, producing a sink. Usually the stars appeared spontaneously in the layer after a temperature increase, and usually the stars stayed at a fixed position for quite a while, a couple of days or a couple of temperature increases. In one particular experiment there were two stars simultaneously in the layer. We have noticed the existence of such stars much earlier, see figure 4 in Koschmieder (1974). In the present series of experiments we cannot blame a scratch on the bottom for the formation of the star, as we did then. There may have been an impurity at the centre of the star, such as a grain of dust or a piece of lint, but we have never been able to locate such a disturbance after the experiment. It may be simply that the formation of the star is a natural way for the pattern to accomplish a change of the wavenumber.

4. Conclusions

The principal result obtained from these experiments is the observation that the wavenumber of surface-tension-driven hexagonal convection cells increases when the Marangoni number is increased in the slightly supercritical range. This observation confirms the same observation made by Bénard (1900) in his original experiments. The decrease of the cell size of the hexagonal cells has implications for nonlinear stability in general. The increase of the wavenumber of surface-tension-driven convection is in startling contrast to the experimentally well-established decrease of the wavenumber of buoyancy-driven Rayleigh–Bénard convection under supercritical conditions which was discovered by Koschmieder (1966). The decrease of the wavenumber of supercritical Rayleigh–Bénard convection is discussed in detail in Koschmieder (1974). The variation of the wavenumber of the convective motions is a consequence of nonlinear effects and is of prime interest for the nonlinear theory of these instabilities.

The second result obtained from our observation is the apparent uniqueness of supercritical surface-tension-driven convection. This is important because according to linear theory the flow can be non-unique at a given supercritical Marangoni number, the theoretical non-uniqueness permitting wavenumbers smaller as well as larger than the critical wavenumber. The nonlinear theory of Cloot & Lebon (1984) also has non-unique solutions. We cannot compare our finding of apparent uniqueness of the wavenumber of the hexagonal cells with corresponding results of buoyancy driven Rayleigh–Bénard convection, because it has not been established whether the wavenumber of supercritical Rayleigh–Bénard convection is unique or non-unique. However, there seems to be a consensus that time-independent supercritical convection motions always have wavenumbers smaller than the critical wavenumbers, and that not all wavenumbers smaller than a_c permitted by the Eckhaus (1965) diagram are actually stable. Whether or not the remaining interval of supercritical convective wavenumbers smaller than a_c is only a line plus–minus the experimental uncertainty has so far eluded experimental work.

The third result of our experiments was the observation that the wavenumber of the hexagonal convection cells decreases when the Marangoni number exceeds a certain moderately supercritical value. This is in agreement with Bénard findings, as well as those of Dauzère (1912) and Cerisier *et al.* (1987*b*). The wavenumber of the cells cannot continue to increase at its original pace because it would intersect the marginal curve of linear theory for surface-tension-driven convection. But that does not necessitate that the wavenumber begins to decrease with increased ΔT. We assume that the decrease of the wavenumber at larger Marangoni numbers is caused by contributions of buoyancy to the flow. We find that assumption confirmed by our experiments with fluid layers of different depth. Whether buoyancy does indeed cause the ultimate decrease of the wavenumber of the hexagonal cells remains to be clarified by theoretical studies or by a microgravity convection experiment.

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