Experimental observation of dynamic mode switching in interfacial-tension-driven convection near a codimension-two point

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The container geometry's effect on flow patterns that form near the onset of interfacial-driven convection were studied. Numerical studies indicate that the flow patterns at the onset of convection should be steady, while experiments showed *a dynamic switching between two different flow patterns*, or modes. This phenomenon is a function of the container's radius over height (aspect ratio), and it is believed to result from the coexistence of energy states. Unsteady convection has significant implications in liquid encapsulated crystal growth where the aspect ratios are ever changing due to the solidification process. [S1063-651X(96)50510-6]

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Liquid encapsulated crystal growth is a process where the aspect ratios are ever changing due to solidification. Fluid convection in the melt that is associated with this growth is caused by density and interfacial tension gradients. It is therefore useful to understand the effect of lateral boundaries on the flow pattern that arises when interfacial and gravity driven convection are present. In this study an unusual and interesting phenomenon has been uncovered, manifested by a dynamic switching between two different flow patterns or modes.

A liquid layer with a free surface, subject to a thermal gradient can convect by two principal mechanisms. The first process is buoyancy or Rayleigh [1] convection, while the second type of thermally induced convection is interfacial tension driven or Marangoni convection [2]. Imagine a closed container of fluid initially at a quiescent state. When a vertical temperature difference is applied across the fluid layer, the fluid remains in a static state until the temperature difference reaches a critical or bifurcation point. At the bifurcation point, the fluid begins to convect and forms a particular pattern. The pattern that is formed is a function of the container's radius over height (aspect ratio). A review of recent works on the Rayleigh-Marangoni-Bénard convection is given by Davis [3] and Lebon [4].

Studies in pattern formation are often performed far past the onset condition into deeply nonlinear regimes. Nonetheless, curious behavior may be obtained in a careful and systematic study using linear analytical tools near the onset of convection.

Table I gives the critical Marangoni number for azimuthal modes 0, 1, 2, and 3 for two different aspect ratios, 1.5 and 2.5. The Marangoni numbers were calculated from the linearized Boussinesq equations in a cylindrical geometry. The liquid was bounded on the vertical sides by a rigid, no slip side wall. The lower surface was rigid and assumed to be at a constant temperature. The upper surface was in contact with a passive gas and surface deflections were assumed negligible. Note that the linearized Boussinesq equations are independent of the Prandtl number as it is scaled into the temperature. For every calculation performed with this model, the largest eigenvalue was found to be real; that is, the fluid did not oscillate at the onset of convection.

Each mode in Table I represents a different flow pattern. For example, $m=0$ represents the ''toroidal'' flow pattern (Fig. 1). The mode which has the smallest Marangoni number will be the mode which is present at the onset of convection. The calculation in Table I reveals that the $m=0$ or toroidal flow should be expected at an aspect ratio of 1.5. It is important to note the difference between the smallest (i.e., critical) Marangoni number and the next smallest. For the 1.5 aspect ratio, the difference is quite large about 12%. For the 2.5 aspect ratio, the difference between the first mode, $m=0$, and the next mode, $m=2$, is quite small, about 1.5%. This difference becomes important experimentally when one tries to resolve which flow pattern is present at the onset of convection. In special cases, two modes coexist at the smallest Marangoni number. These points are known as codimension-two points. The 2.5 aspect ratio is near a codimension-two point. The linearized instability calculations near the codimension-two points for Marangoni [5] and Rayleigh [6] convection indicate that only one steady pattern should be seen, but not both.

The prime motivation for this study was to investigate the pattern behavior close to a codimension-two point where a change in pattern was expected as the system crossed the critical location. To further investigate the codimension-two point experimentally, a single liquid layer of 100 cS (where 1 $S=1\ cm^2/s$) silicone oil with an upper air gap was placed between two conductive plates. The upper plate consisted of zinc selenide, which is transparent to infrared radiation in the 8 to 12 μm range. Precautions were taken to ensure that the temperature was uniform across both upper and lower plates. A temperature difference was imposed between the two plates. This difference was increased in a quasisteady fashion until the fluid began to convect and exhibited a pattern. The

TABLE I. Critical Marangoni numbers associated with each mode for aspect ratios [(radius)/(height)] of 1.5 and 2.5 [5].

Mode	1.5 Aspect ratio	2.5 Aspect ratio
0	90.45	69.37
1	101.3	70.84
2	112.02	70.41
3	129.81	72.98

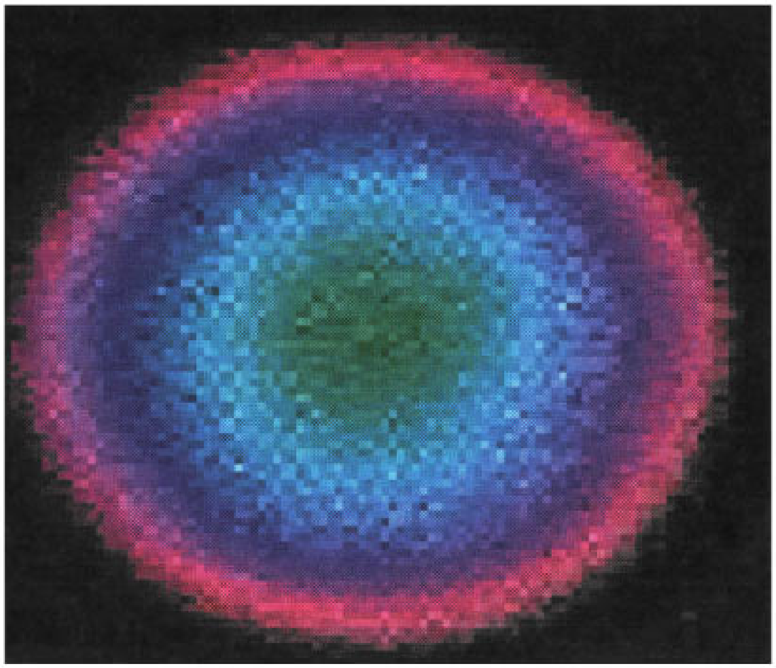

FIG. 1. An infrared image of the toroidal flow pattern in a cylindrical container. The radius over height aspect ratio is 1.5. The cooler fluid is colored purple and the hotter fluid green. The hotter fluid is 1.5 °C hotter than the cooler fluid.

standard deviation, due to temperature control, was under 2% of the imposed temperature difference. The depth of the silicone oil was 5.0 mm and the air gap was 11.2 mm for all the experiments reported here. This system gives a ratio of the Rayleigh (Ra) to Marangoni (Ma) number around 4.6. The flow visualization technique used an infrared (IR) camera to measure the temperature field of the upper liquid surface. The IR camera was placed above the apparatus and viewed down upon the surface. As IR radiation is readily absorbed by silicone oil, the radiation detected by the camera was only from the very upper layer of the silicone oil.

The first experiments used an aspect ratio of 1.5. The flow pattern predicted to be seen from numerical calculations in Table I, is a toroidal flow. The toroidal flow shows fluid rising up from the center, moving radially across the top of the surface, and then falling down the sides of the container. Indeed, as seen in Fig. 1, the IR camera captured this flow pattern at the onset of convection. Further moderate increases in temperature difference did not change the flow pattern.

The second set of experimental runs used an aspect ratio of 2.5, which is near a codimension-two point. Again the temperature difference was increased until fluid convection was seen near the predicted critical value. The first pattern seen was the toroidal flow. The temperature difference was increased and then held constant, within experimental error, for several hours. After a few minutes, the flow patterns started to transform into a dynamic state and maintained this state for as long as the temperature difference was held fixed. The period of the oscillations was constant and was seen to be about one fourth of the horizontal thermal diffusive time constant (92 min), and about seven times the vertical thermal diffusive time constant. This experiment was reproduced and verified several times.

Figure 2 depicts snapshots of the flow pattern at an aspect ratio to the left of a codimension-two point. Additional experiments were also performed for a slightly larger aspect ratio of 2.6, where the pattern observed at the onset of convection was different from the 2.5 aspect ratio. The pattern

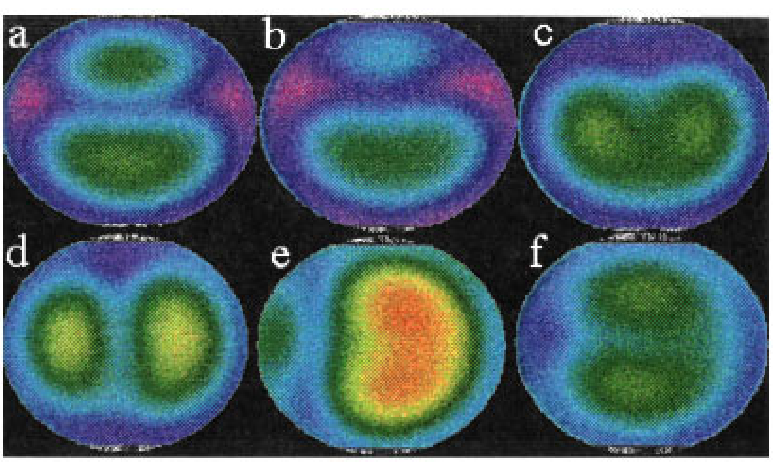

FIG. 2. Time sequenced infrared images showing the switching between flow patterns. The aspect ratio is 2.5 and to the left of a codimension-two point. The yellow-red color signifies the hotter fluid and the purplish-blue is the cooler fluid which is 2 °C cooler. The flow patterns start as an $m=2$ flow with two cells (a). One of the cells increases in size, trying to form the toroidal flow (b). In doing so, it eliminates the other cell. Before the toroidal pattern forms completely, it splits, like an amoebae, into two cells (c). These two cells shift into the two cell formation seen in part a offset by 90° (d). Before this cell achieves a complete toroidal flow (e), it too splits into two cells. The two cells shift and arrive at the exact flow pattern seen in (a) (f). As long as the temperature difference is held constant, the dynamic state of convection cells growing and splitting continues and repeats itself with a regular time interval.

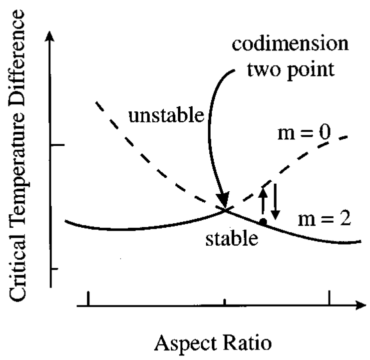

FIG. 3. Schematic of the codimension-two point. The experiment is usually performed at some point close to but not exactly at the codimension-two point. Self-excited perturbations in the supercritical region can cause the fluid to switch from one mode to another.

appeared as a steady superposition of $m=2$ and a double concentric toroidal pattern. Pattern formations near codimension-two points have been observed [6] in pure buoyancy flows in containers. It was observed there that the mode switching did not occur at a codimension-two point. This study reveals that codimension-two points lead to dynamic pattern switching in a cylindrical container near the onset where interfacial or Marangoni forces are present. Similar work [7] has been done where pattern switching has been observed in square containers and is attributed to a Takens-Bogdanov bifurcation that is associated with codimension-two points.

A closer look at Fig. 3, which is a schematic of a codimension-two point, may reveal why the flow pattern switches between different modes. Self-excited perturbations in temperature will cause the fluid to remain in a dynamic state of mode switching. For codimension-two points that may occur at smaller aspect ratios, the angle between the lines of constant flow pattern is considerably larger. It is much more difficult to place the system such that small perturbations in the temperature difference will cause the flow pattern to switch from one mode to another.

Theoretical work has been performed to describe weakly nonlinear behavior around codimension-two points. For example, Erneux and Reiss [8] looked at codimension-two points for supercritical bifurcations. They noted that when the supercritical bifurcation was symmetric, and no imperfection was introduced, the steady solutions would branch off into a secondary solution as the bifurcation parameter was increased, but the secondary solution would always be steady. However, they showed that when an imperfection to the base state was introduced, Hopf bifurcations to a secondary solution were possible. Rosenblat, Davis, and Homsy [9] performed a weakly nonlinear analysis for the pure Marangoni problem; that is, they neglected buoyancy effects. In their analysis they showed that for a $m=0$ and $m=1$ codimension-two point, it was possible for Hopf bifurcations to occur. However for the $m=0$, $m=2$ codimension-two points, they did not find a Hopf bifurcation. It is important to note the many differences between their paper and the physical experiment, the most important being the lack of gravitational effects and the assumption of an unphysical, vorticity free sidewall boundary condition. This condition will cause the modes to unfold in a different order than what is observed in the experiment: the vorticity free sidewall generates $m=1$, $m=2$, $m=0$ modes as the aspect ratio is increased, whereas the no slip sidewall [5] has $m=1$, $m=0$, $m=2$ modes as the aspect ratio is increased. Echebarría, Krmpotic, and Pérez-García [10] also found oscillatory mode interactions when they took into account the Rayleigh or gravitational effects. However, their work only concentrated on the $m=1$, $m=2$ mode interactions.

In this study we were definitely able to show the existence of a codimension-two point by both linearized calculations and by performing experiments. The experiments showed the onset of a different pattern on each side of the codimension-two point. Weakly nonlinear analysis has shown that codimension-two points can lead to Hopf bifurcations or oscillatory states. The computed critical Marangoni number and the predicted flow pattern were found to agree well with the experimental results. This study has implications in crystal growth, where multiple mode interactions can be another mechanism for unsteady convection, which in turn can lead to dopant stratification.

Future progress on this system may answer such questions as, can this behavior be seen at different codimension-two points, does oscillatory behavior only occur at codimension-two points, and can the flow switch between more than two patterns? Answers to these questions certainly warrant continued study in this fascinating phenomenon.

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