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Marangoni convection in multiple bounded fluid layers and its application to materials processing

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A brief review of multilayer convective phenomena that is associated with materials processing is presented. Several instability phenomena that can occur in a bilayer of two fluids heated from either above or below and the effect of laterally and vertically confined geometries are explained. In particular it is shown that such confinement can lead to the occurrence of codimension-two points and pure thermal coupling that is initiated by convection in an upper gas phase during liquid–gas bilayer convection. Experimental evidence that shows the effect of geometrical restrictions is given.

Keywords: Rayleigh; Marangoni; interfacial tension driven convection; buoyancy driven convection; multiple fluid level convection

1. Introduction and physics

Much of the work reported in this paper has been motivated by the need to understand a technique for growing certain crystalline materials, known as the liquid-encapsulated vertical Bridgman (LEVB) crystal growth method. Liquid-encapsulated crystal growth is a process for producing III-V semiconductor crystals from bulk liquid melts. The demand for crystals of increasingly higher purity and lower defects requires us to understand this process in much greater detail. Some examples of crystals grown using this technique are gallium arsenide (GaAs) and indium phosphide (InP). Taking GaAs as an example, when GaAs is melted, it has a tendency to decompose, releasing arsenic gas and destroying the desired stoichiometric ratio. To prevent this decomposition, a liquid encapsulant of boric oxide (B_2O_3) is placed on top of the gallium arsenide. In addition, an inert gas may be placed on top of the B_2O_3. These three layers are placed in a crucible, which is lowered through a temperature gradient created by a furnace. The lower end of the crucible is cooled, thereby solidifying the gallium arsenide. This configuration is shown schematically in figure 1. The heating configuration generates vertical as well as radial temperature gradients and, consequently, interfacial-tension-gradient-driven convection, also known as Marangoni convection, and buoyancy-driven convection, also called Rayleigh convection, can occur at the liquid–gas and liquid–liquid interfaces as well as in the bulk fluid regions.

While the LEVB technique is the motivation for this study, only by considering simple systems can we have a clearer understanding of the physics of the convective process. Radial gradients of temperature, creeping of the encapsulant along the vertical sidewalls and solutal gradients all have a complicated effect on the convection.

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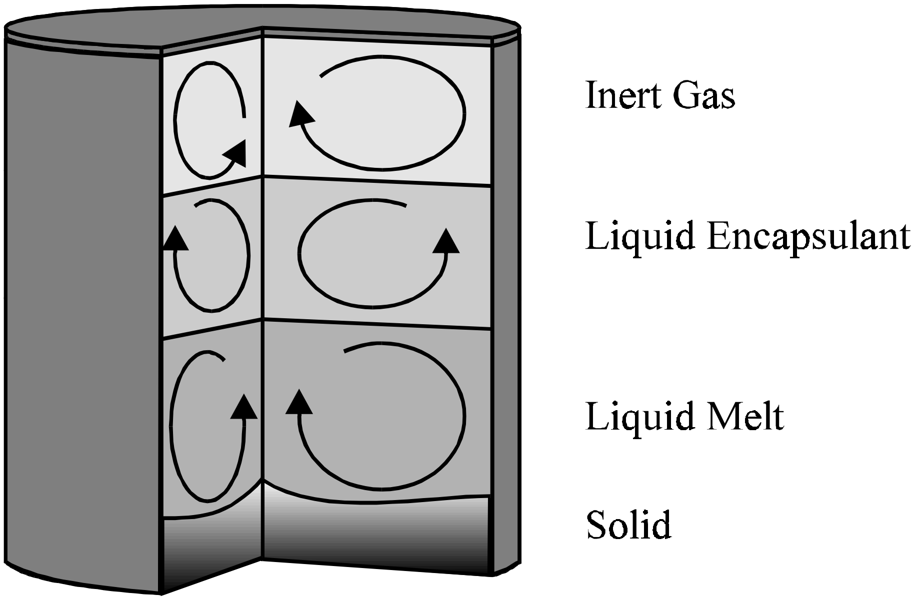

Figure 1. Schematic of a liquid encapsulated crystal grower: a system of three convecting fluid layers. Convection in the GaAs liquid influences the quality of the GaAs solid.

Indeed the onset of convection in an actual LEVB system occurs simultaneously with the application of any temperature gradient. However, a clear understanding of convection in LEVB and many other materials processing methods requires us to consider problems where classical fluid mechanical procedures may be employed, thereby simplifying the mathematics while simultaneously revealing the essential physical features.

One such problem is the Rayleigh–Marangoni problem. Here a gas or another liquid superimposes a liquid layer and a vertical temperature gradient is applied. Suppose that the density and interfacial tension of the liquid decreases with increasing temperature. As the liquid is heated from below, it is top heavy. A small disturbance can upset this arrangement if the overall temperature difference is large enough and flow can ensue in the form of buoyancy or Rayleigh convection. However, flow can occur even in the absence of gravity. For example, in the quiescent state the liquid–gas interface is flat and a small disturbance to it causes a transverse temperature gradient at the interface causing fluid to flow from warm regions of low interfacial tension to cold regions of high interfacial tension. Hot fluid from below rises to the interface and cold fluid from the interface moves down to maintain continuity of fluid flow and the convection continues as Marangoni convection. For small values of the vertical temperature gradient, the fluids remain quiescent and transport heat by pure conduction. However, when the temperature gradient reaches a critical value, even the smallest disturbances imposed on the system amplify with time and the system reaches a steady or time-periodic steady state. In other words a critical temperature gradient is required for convection to occur. More details on the nature of this type of convection are available in the reviews of Koschmieder (1993) and Davis (1987). We will explain the physics of single and multilayer convection in laterally bounded geometries where the layers are heated from above making them gravitationally stable and where the layers are heated from below making them gravitationally unstable. The explanation of physics in multilayers will be followed by a report on two sets of

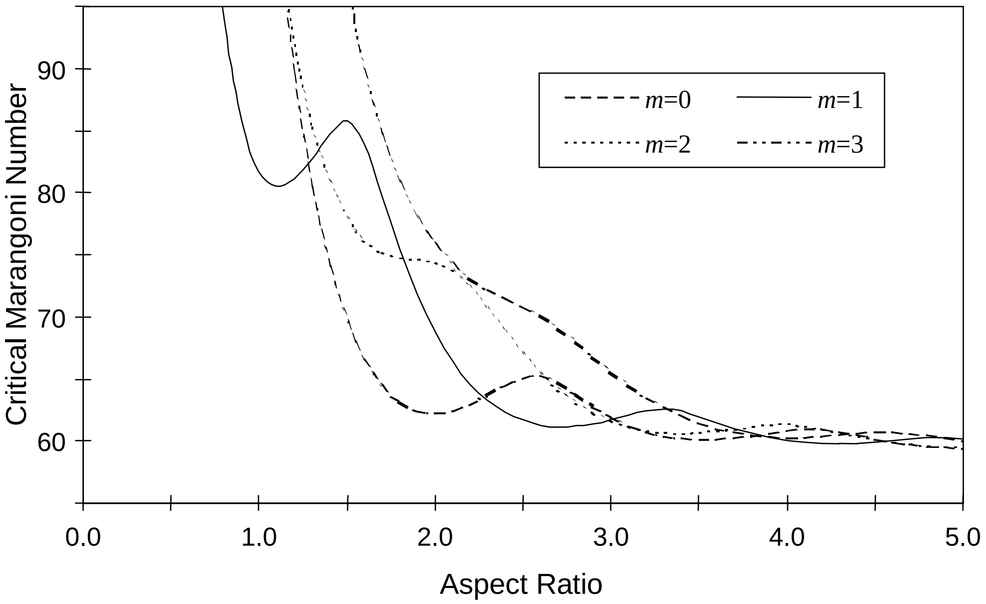

Figure 2. Plot of the critical Marangoni number versus the aspect ratio of a cylinder. The mode, m, with the smallest Marangoni number at a given aspect ratio is the mode or flow pattern at the onset of convection.

experiments. Noticeably absent from this paper will be the effects of solutal convection, some aspects of which have been covered by several other authors (McFadden *et al.* 1984; Turner 1985). It should be noted that this paper is a brief report of the work done by us and a few other researchers. Greater detail is available in the thesis by Johnson (1997) as well as papers by Johnson & Narayanan (1996, 1997, 1998). Fuller explanations of the effects of convection on crystal growth are given by Hurle (1994), Müller (1988) and Schwabe (1981).

The extent of convection is often characterized by a dimensionless temperature difference represented by the Marangoni or Rayleigh numbers. The Marangoni number is proportional to the depth of the liquid, the temperature difference and the variation of the surface tension with respect to the temperature and inversely proportional to the dynamic viscosity and thermal diffusivity. The Rayleigh number is proportional to the cube of the liquid depth, gravity, the temperature difference and the thermal expansion coefficient and inversely proportional to the kinematic viscosity and the thermal diffusivity. In a physical system, fixing the temperature difference necessarily fixes both the Rayleigh and Marangoni numbers.

We begin by confining our discussion to a single layer of fluid, heated from below, with a free surface. In this configuration both buoyancy and interfacial-tension forces become important. For larger depths, buoyancy is more important than interfacial-tension effects, and when the fluid depth is small, interfacial-tension forces dominate convection.

(*a*) *Physical effects of a bounded geometry*

Consider a single layer of fluid bounded below by a rigid conducting plate and whose upper surface is bounded by a passive gas. By a passive gas we mean a gas which has no viscosity and only conducts heat away. The lower plate here is at a higher temperature than the passive gas. In a fluid of infinite horizontal extent,

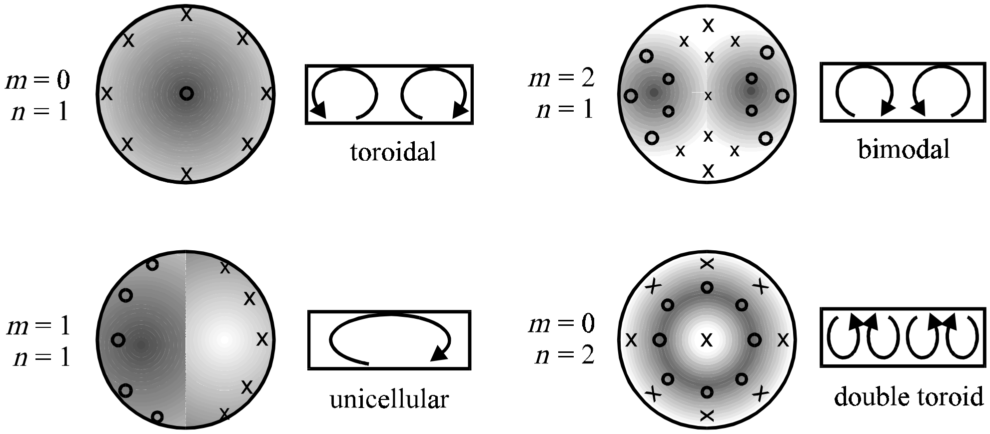

Figure 3. Schematic of four different flow patterns: ○, fluid flowing up; ×, fluid flowing down.

there is no limit on the number of convection cells. However, in a bounded finite-sized container only a finite number of convection cells may exist. Physically this means that at the onset of convection in a bounded cylinder, only one flow pattern will usually exist. As the aspect ratio (radius/height) of the container increases, more convection cells will appear. Figure 2 is a representative calculation of the critical Marangoni number for various aspect ratios and for different azimuthal modes m. The Biot number, which is a dimensionless surface heat-transfer coefficient is equal to 0.3.

In a bounded cylinder, each flow pattern is associated with an azimuthal mode, m, and radial mode, n. For example, at an aspect ratio of 1.0 in figure 2, there is an $m = 1$, $n = 1$ flow pattern (see figure 3). For an aspect ratio of 1.5, there exists an $m = 0$, $n = 1$ flow pattern. The azimuthal mode is the number of times the azimuthal component of velocity goes to zero, and the radial mode is the number of times the radial component of velocity goes to zero starting from the centre for a given vertical cross-section.

At particular aspect ratios, where the fluid switches from one flow pattern to the next, there coexist two different flow patterns. These aspect ratios are known as codimension-two points. For certain codimension-two points, the flow patterns will interact nonlinearly to yield oscillatory behaviour (Rosenblat *et al.* 1982; Johnson & Narayanan 1996). This phenomenon will be shown later in § 2.

(*b*) *Physical effects of multiple fluid layers*

Imagine a less dense immiscible layer of fluid above the lower layer of fluid. Here the lower layer is bounded below by a rigid conducting plate and another rigid conducting plate bounds the upper layer. Once again let the temperature of the lower plate be greater than the upper plate. The interface between the two fluids may deform and is capable of transporting heat and momentum from one layer to the other. We will now consider the various types of convection that can occur in a bilayer of two fluids.

In order to distinguish the various convection mechanisms, we introduce phrases such as 'convection initiating in one layer or another'. Strictly speaking, convection occurs in both fluids simultaneously, although one layer may be more unstable than the other, driving flow in the other layer.

Turning now to various convective mechanisms, consider figure 4. Suppose that convection initiates in the lower layer. The upper layer responds by being dragged,

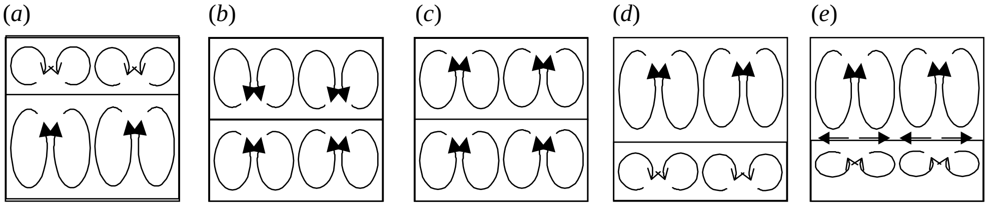

Figure 4. Schematic of the different types of convection-coupling: (*a*) lower dragging mode; (*b*) viscous coupling; (*c*) thermal coupling; (*d*) upper dragging mode; (*e*) pure thermal coupling. Moving from (*a*) to (*e*), the buoyancy force in the upper layer increases. Gas–liquid thermal coupling, with surface-driven flow, is caused by the upper fluid buoyantly convecting and simultaneously inducing interfacial-tension- or buoyancy-driven convection in the lower layer near the interface.

generating counter rolls at the interface. Hot fluid flows up in the lower layer and down in the upper layer. The upper layer is not buoyant enough and moves by a combination of viscous drag and the Marangoni effect. This is seen in figure 4*a*. The sign of the velocity switches and the maximum absolute value of the lower-layer velocity is much greater than the maximum absolute value of the velocity of the upper layer.

When the buoyancy in the upper layer increases and the upper layer begins to convect, one of two things can happen. The first possibility is that the two fluids are *viscously coupled*. Physically this can be shown in figure 4*b* as counter-rotating rolls in the two fluids. This can also be denoted by the vertical component of velocity switching sign at or near the interface, while the temperature perturbations indeed switch sign at the interface itself. If the temperature perturbation switches sign near the interface in either layer near the interface we would say that the bilayer is nearly viscously coupled. In particular if the switch takes place in the upper fluid near the interface, then the lower layer is slightly more buoyant. If the temperature perturbation switches sign in the lower layer, then the upper layer is more buoyant. The Marangoni phenomenon, for fluids, whose interfacial tension decreases with an increase in temperature, plays an ambiguous role here. The hot fluid flowing up in the lower layer causes the fluid at the interface to move in the same direction. However, the colder fluid moving down in the upper layer contradicts this. The exact effect the Marangoni phenomenon has on the two fluids depends on where the thermal perturbations change sign. For the situation where the thermal perturbations switch sign at the interface there is no Marangoni effect.

The second possibility is *thermal coupling* where the rolls are corotating (figure 4*c*). Here hot rising fluid from the lower layer causes hot fluid in the upper layer to flow up. The maximums of the vertical component of velocity and the temperature perturbations have the same sign in each fluid layer. Strictly speaking, the transverse components of velocity should be zero at the interface. However, thermal coupling is sometimes referred to the case when a small roll develops in one of the layers so as to satisfy the no-slip condition at the interface. In this situation, when the interfacial tension decreases with an increase in temperature, the Marangoni effect encourages flow in the lower fluid layer, and discourages the flow in the upper fluid.

Another interesting phenomenon is present at certain fluid depths where both thermal and viscous coupling can occur. At these depths, a competition arises between the two types of convection. As both convection configurations cannot occur simultaneously, the fluids begin to oscillate between these two states. This phenomenon was

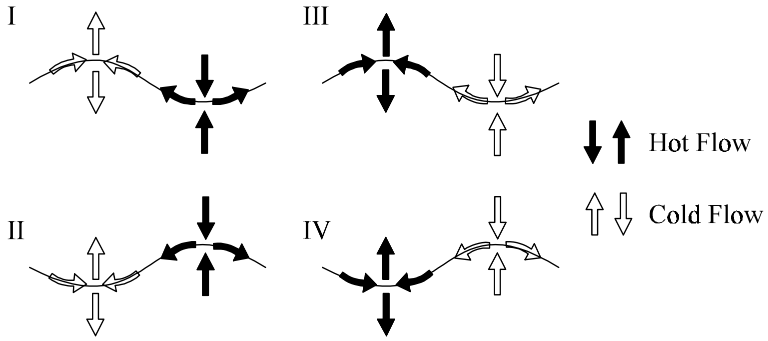

Figure 5. The four possible interfacial structures at a fluid–fluid interface. Each structure can give information about the driving force of the convection.

first reported by Gershuni & Zhukhovitskii (1982), and has recently been confirmed by Andereck *et al.* (1996).

As the buoyancy continues to increase in the upper layer, convection initiates in only the upper layer and the lower layer is viscously dragged (figure 4*d*). This situation only occurs when the upper fluid is a liquid, as gases are very tenuous and will not exert much shear. The vertical component of velocity in this case switches sign and the magnitude of convection in the upper fluid is much greater than the magnitude of convection in the lower fluid.

The last figure (figure 4*e*) is an example of what may be called *pure thermal coupling*. This typically occurs in a liquid–gas system where buoyancy convection is predominant in the gas layer. The convecting gas then simultaneously creates a non-uniform temperature profile across the liquid–gas interface and generates either Marangoni or buoyancy-driven convection in the lower layer (Johnson *et al.* 1998). Notice that the convection in the lower layer is now generated purely by horizontal temperature gradients at the interface and not by viscous dragging. To maintain the no-slip condition at the interface a small counter-roll may develop in the gas-phase. This roll is not shown in figure 4*e*.

(*c*) *Physics of interfacial structures*

Another indicator of what is occurring in bilayer convection can be inferred from the fluid–fluid interface instead of the bulk convection. In a paper by Zhao *et al.* (1995), four different interfacial structures were identified for any given convecting bilayer with a deflecting interface. Each of these structures depends upon whether fluid was flowing into or away from the trough or the crest, and whether the fluid was hotter or cooler at the trough or the crest of the interface. Hot fluid flowing into a trough defines the first interfacial structure. The second interfacial structure has hot fluid flowing into a crest. The third structure has hot fluid flowing away from a crest and the fourth structure has hot fluid flowing away from a trough. Each of these four scenarios is given in figure 5.

One of the important factors to consider in interfacial structures is the direction of the flow along the interface. As interfacial tension is usually inversely proportional to temperature, at cooler regions of the interface, the interfacial tension will be higher and will pull on the interface. Where the interface is hotter, the interfacial tension will be lower causing the fluid to move away from warmer regions. Another important factor is the direction of the flow into or away from a crest or a trough. One reason the interface deflects is due to bulk convection, caused by buoyancy effects, pushing against the interface. Consider two fluids whose dynamic viscosities are equal. If buoyancy-driven convection is occurring mostly in the lower layer, then the fluid will flow up from the lower layer into a crest. If the fluid flows down from the top layer into a trough, then one would argue that buoyancy-driven convection occurs mostly in the upper fluid.

In each of the four cases, the interfacial structure can be used to indicate the driving force of the convection. In the first interfacial structure, the dominating driving force is interfacial-tension-gradient-driven convection. This is seen as the cold fluid, with the higher interfacial tension pulling the fluid up into the crest. The first interfacial structure can also occur by buoyancy-driven convection in the upper layer, when the density of the upper layer increases with an increase in temperature. In the second interfacial structure, buoyancy drives convection in the lower phase. The hot rising fluid pushes the interface upwards. As the fluid moves along the interface, it cools and eventually sinks back down. The third interfacial structure is dominated by buoyancy-driven convection in the upper phase or by interfacial-tension-driven convection where the interfacial tension increases with respect to temperature. The fourth interfacial structure only occurs when the lower fluid has a positive thermal-expansion coefficient. In other words, the density increases with an increase in the temperature, causing the cooler lower fluid to flow up into a crest.

Knowledge of interfacial structures will be beneficial in the understanding of certain materials processing problems such as drying of films, coatings and deposition.

(*d*) *Physics of heating from above*

In the previous subsections we talked about some of the phenomena that occur in single- and multiple-fluid layers heated from below. However, in an attempt to avoid convection in crystal growth, the crucible is often cooled from below and heated from above. This heating configuration changes the physics of the problem, which is the topic of this subsection.

When a layer of fluid is being heated from above, it creates a stable density stratification. Therefore not only does the buoyancy force not cause convection, it acts to inhibit other instabilities. Marangoni convection, though, may still occur in fluids being heated from above.

First we will consider a single layer of fluid superposed by a passive gas. If the upper gas is truly passive, then pure Marangoni convection will not occur. For example, suppose some random perturbation causes some part of the surface to become warmer than the rest of the surface. The interfacial tension will decrease in this region and the tension will pull fluid away from this hot spot. By continuity, fluid lying below the hot spot will rise up to replace the displaced fluid. As the lower fluid is cooler than the surface this region now cools off and the interfacial tension increases, thereby restabilizing the region. However, in real systems, the upper fluid is never truly passive. Given the same scenario, fluid movement along the interface will also drag warmer fluid from above. This warmer upper fluid will further increase the temperature in this region, and, depending upon the ratio of thermal–physical properties, reinforces the instability.

By this argument, it appears that an active upper fluid is necessary to have Marangoni convection when the system is being heated from above. However, this is not the case if the buoyancy effects are included. In Rednikov *et al.* (1998), it was demonstrated, theoretically, that oscillatory onset of convection may occur for a single layer of fluid with a purely passive upper gas. The explanation is as follows.

If a small volume of fluid is displaced within the bulk of the fluid, the density stratification acts as a restoring force, causing a dampened oscillation within the fluid. These are often referred to as internal waves. The Marangoni force acts similarly, as discussed above, also giving dampened oscillations. Apparently when these two forces act together they can overshoot one another leading to sustained oscillatory convection. Indeed, as was demonstrated in their paper, this only occurs in certain fluids, at certain depths, where the buoyancy and interfacial-tension forces are approximately equal.

A completely different type of instability is also possible in two layers of fluids being heated from above. Gershuni & Zhukhovitskii (1981) first demonstrated this phenomenon by analysing two immiscible fluids where the interface between the fluids was assumed flat and the Marangoni phenomenon was neglected. They found the onset of steady convection when the thermal conductivity and thermal expansivity of the lower fluid was much greater than that of the upper fluid.

The mechanism of this instability is as follows. Suppose an element of fluid in the upper layer, near the interface, is displaced towards the lower layer. Because the thermal expansion of the upper fluid is so small, this element remains in a relatively neutrally buoyant state. Also, as the thermal conductivity of the upper fluid is small, it cools very slowly. When the two fluids are heated from above, the displaced fluid will be warmer than its surroundings. This element of fluid then heats part of the lower fluid near the interface. The lower fluid, with its relatively large thermal conductivity and thermal expansivity, quickly heats up and expands horizontally. This expansion then causes convection in the lower fluid layer and propagates by viscously coupling with the upper fluid layer. The Marangoni phenomenon, if it were considered, would act to enhance this instability.

Another case of interest is convection induced by the Rayleigh–Taylor instability. This phenomenon can occur in two immiscible fluid layers being heated either from above or below, when the densities of the two fluids are approximately the same and the thermal expansivity of the lower fluid is much greater than the thermal expansivity of the upper fluid. Upon heating, the density of the lower fluid will decrease and become less than the density of the upper fluid. Consequently, the heavier upper fluid will begin to sink causing large deformations in the liquid–liquid interface, generating the Rayleigh–Taylor instability (Chandrasekhar 1961). This problem has been investigated extensively in Renardy & Renardy (1985) and Renardy (1996), but is of application to materials processing only if the densities of both layers are similar.

2. Some experimental observations

The experiments were used to investigate both the oscillatory behaviour near codimension-two points and the pure thermal coupling of air with silicone oil (see figure 4*e*). Details on the experimental procedure are available in Johnson (1997) and Johnson & Narayanan (1996, 1998).

(*a*) *Experimental apparatus and procedure*

The experiments consisted of two compartments: one for the lower fluid and one for the air. Lucite inserts were used to give a variety of different fluid depths and aspect ratios. A copper plate was placed below the liquid insert and the air insert was bounded above by a high-thermal-conductivity infrared transparent zinc selenide (ZnSe) window. Heating of the copper plate was done by an enclosed stirred water bath that was in turn heated by a hot plate. The top of the ZnSe window was kept at a constant temperature by accurately controlling the temperature of the overlying air. The overall temperature control was kept within ±0.05 °C.

The flow patterns that developed at the silicone oil–air interface layer were visualized with an infrared camera. Although other flow visualization techniques could have been used, such as shadowgraphy or particle tracing, the IR camera was chosen to prove the viability of its use with opaque materials, such as gallium and gallium arsenide. The IR imaging technique is also useful in observing weak thermocapillary flow near the surface, whereas shadowgraphy requires some strength in the domain flow.

To guarantee that the flow pattern seen was indeed the flow pattern at the onset of convection, the temperature difference applied across the bilayer system was carefully increased. At first a temperature difference was applied that was less than the critical temperature difference necessary for the onset of convection. This, and all temperature differences, were kept constant for several characteristic time constants; *ca.* 3–4 h. If no flow pattern was seen, the temperature difference was then increased by as little as 0.05 °C. This was repeated until the temperature profile at the interface changed to some distinct pattern, indicating that the fluid had begun to flow. At this point, the flow pattern was recorded and the temperature difference noted.

(*b*) *Experimental observation of codimension-two points*

As was noted in § 1, there exist certain liquid aspect ratios where two different flow patterns coexist. For example, in figure 2 at an aspect ratio of 2.3, there exists a codimension-two point between the azimuthal modes $m = 0$ and $m = 1$. The questions we want to answer are: What happens at these aspect ratios? Does one flow dominate over the other? Do the different flow patterns coexist as a superposition of both states, or do they oscillate and interact between these two states?

Rosenblat *et al.* (1982) have performed a weakly nonlinear analysis to investigate these questions. They found that all three of these possibilities may occur, depending on the Prandtl number, the particular codimension-two point being investigated, and on which side of the codimension-two point the aspect ratio lies. To simplify their calculations, a vertical and tangential vorticity-free side-wall was assumed. Later a more realistic no-slip condition was applied (Zaman & Narayanan 1996; Dauby *et al.* 1997), where it was noted that the order of azimuthal modes, as the aspect ratio was increased, was different than the vorticity-free condition. These latter calculations were done assuming a linearized instability analysis. Therefore, a direct comparison of the nonlinear analysis with the experiment is not currently possible. Nonetheless, some of the qualitative features should still hold true.

A series of experiments were performed to first find the codimension-two points and then determine the flow patterns at or near the codimension-two point (Johnson & Narayanan 1996). A 5.0 mm-high 2.5 aspect-ratio liquid insert was used in conjunction with a 11.2 mm air height. Table 1 shows the calculated critical Marangoni numbers for four different azimuthal modes for a 2.5 aspect ratio. The table predicts that an $m = 0$ flow pattern should be seen at the onset of convection. However, the critical Marangoni numbers for $m = 1$ and $m = 2$ are also very close to the onset point. Physically, this means that for temperature differences slightly above the critical temperature difference, these modes may affect the flow pattern.

At the onset of convection, a very faint $m = 0$ double-toroidal-flow pattern could be seen. When the temperature difference was increased by just 0.05 °C, the flow

Table 1. *Critical Marangoni number for the first four azimuthal modes for a 2.5 aspect ratio.*

modes	Marangoni number
0	69.37
1	70.84
2	70.41
3	72.98

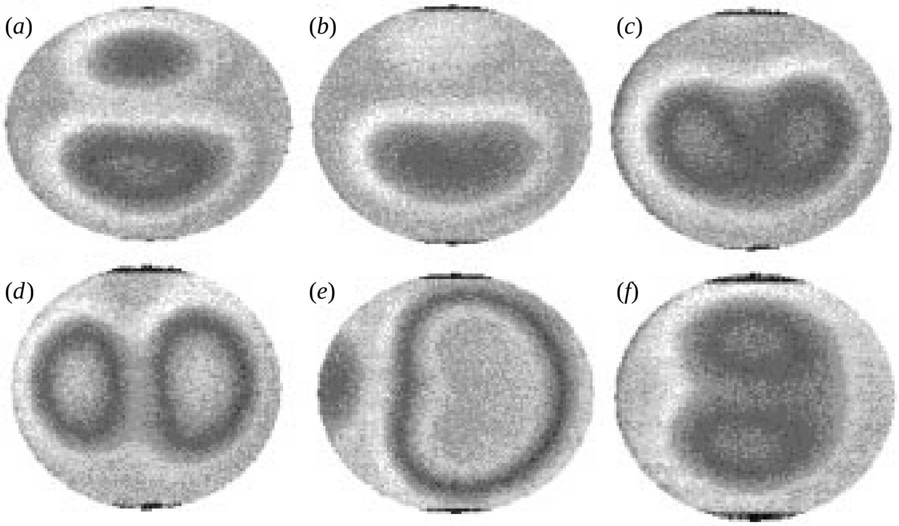

Figure 6. Infrared images showing the mode-switching behaviour in the paper by Johnson & Narayanan (1996). The experiment used 91 cS silicone oil and a 5.0 mm, 2.5 aspect-ratio insert.

pattern changed from the double toroid to a dynamic switching between two and one flow cells (see figure 6).

At first, two symmetric cells appeared (figure 6*a*). Then, one of the cells would grow and push the other cell out of the picture, forming a superposition of the $m = 0$ and $m = 1$ flow pattern (figure 6*b*). Next one cell would grow (figure 6*c*), then split into two cells, rotated by 90° (figure 6*d*). This process would then repeat itself (figure 6*e*) arriving back to the original $m = 2$ flow pattern (figure 6*f*). As long as the temperature difference was held constant, this dynamic process would continue repeating itself approximately every 20 min.

It is noteworthy that oscillatory convection is of particular importance in crystal growth. It has been shown (Hurle 1994) that fluctuating temperatures in the liquid melt have a deleterious effect on the crystal quality, leading to a higher dislocation density.

(*c*) *Experimental observations of thermal coupling*

The thermal coupling of air with the lower fluid was originally discovered by a series of experiments using the same experimental apparatus (Johnson & Narayanan 1998). As was explained in § 1, air can thermally couple with the lower fluid caus-

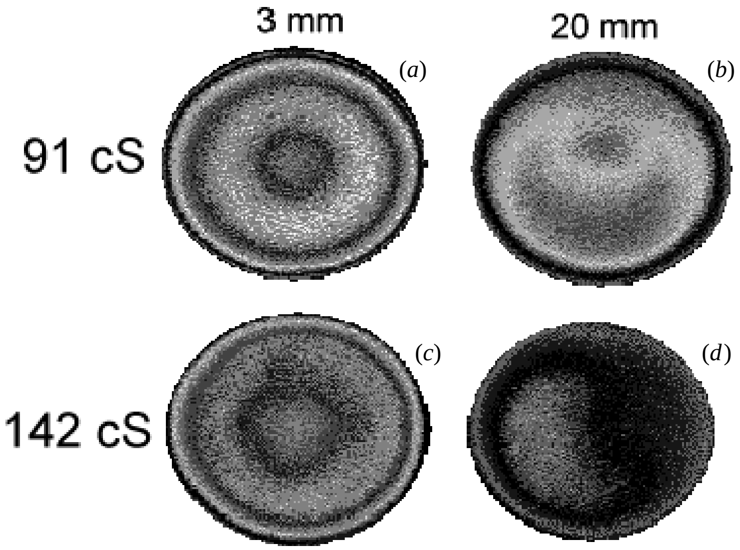

Figure 7. Infrared images of the flow pattern for different air heights and different viscosities of silicone oil. (*a*) and (*b*) used a 91 cS silicone oil. (*c*) and (*d*) used a 142 cS silicone oil. (*a*) and (*c*) had a 3 mm air height, (*b*) and (*d*) had a 20 mm air height.

ing interfacial-tension-driven flow in the lower fluid. To explore this, a set of four experiments was performed.

In all of the experiments, a liquid aspect ratio of 2.0 was studied. Two different air heights (3 and 20 mm) and two different viscosities were used. When the air height of 3 mm was used, the flow patterns did not change with viscosity but the temperature difference across the liquid did increase proportionally, indicating that convection was controlled by the liquid phase. The liquid convection pattern also agreed with calculations and the experimental result is seen pictorially in figures 7*a*, *c*.

When a deeper air height was used, the temperature difference across the liquid at the onset of convection did not change substantially between the experiments that employed different fluid viscosities. This indicated that convection was controlled by the dynamics in the air layer. It may be argued that air convection, when dominant, acts like buoyancy-driven convection between two rigid conducting plates as the lower liquid is much more viscous and more conductive than the air above it. A comparison was therefore made between the measured temperature drops across the air for the deeper air heights and the numerical calculations of Hardin *et al.* (1990). The experimental and theoretical results compared remarkably well and the flow pattern predicted theoretically also compared favourably with the experimental results. This confirmed our hypothesis that deep air heights interact with the lower liquid and drive thermally coupled flow through the interface.

This phenomenon of thermal coupling may not be as important in LEVB because the crucible is often heated from above. However, this may be much more applicable to other important processes, such as evaporation and drying of films.

3. Future work

The research of convection in multiple fluid layers has revealed and continues to reveal many interesting phenomena. However, further work is needed to elucidate some of the details more fully in a realistic system. One of the more interesting areas involves analysing some of the basic instability phenomena in bounded containers. To do this, two-fluid-layer numerical models that take into account realistic no-slip conditions will be necessary. With a proper model some of the effects, such as the Rayleigh–Taylor instability, the Gershuni–Zhukhovitskii instability and oscillations between thermal and viscous coupling, can be studied for containers with small aspect ratios.

To date, few experiments have been performed in small aspect-ratio containers. As was demonstrated in the codimension-two point experiments, new and interesting dynamics are present in small aspect-ratio containers, which are not present in large aspect-ratio containers. It would be interesting to show the interaction of codimension-two points with such instabilities as the oscillations between thermal and viscous coupling. Additionally, several of the phenomena discovered with theoretical models have yet to be shown in experiments. Two examples are the Gershuni–Zhukhovitskii instability and the oscillations shown by Rednikov *et al.* (1998).

By investigating some of the basic physics of multilayer convection, we obtain a better understanding and appreciation for the liquid-encapsulated crystal-growth process and other fluid materials processing problems where temperature gradients are employed. Further research into this field should lead to improvements in such an important industrial process.

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Discussion

J. R. Helliwell (*Department of Chemistry, University of Manchester, UK*). Reference has been made to oscillatory convection flow patterns in fluids and that this is known to cause defects (dislocations and fault lines) in crystal growth. My own particular research interests include the growth of protein crystals for X-ray crystal-structure analysis and how the quality of protein crystals can be improved, and thereby exploited, for higher resolution X-ray crystallographic data collection. I have been using CCD and interferometry diagnostic monitoring of protein crystal growth, and have seen benefits of microgravity if the crystals do not move, and if the mother liquor is not subject to convection (including Marangoni convection). The benefits of these conditions manifest as reduced crystal mosaicity and likewise a lack of, or only a few, mosaic blocks in X-ray topographs of crystals in such cases. In his experiment, how can Dr Johnson be sure that it is specifically oscillatory flow patterns that especially caused defects in his type of crystal?

D. Johnson. We cannot be sure that oscillatory convection is always responsible for defects in crystals. However, research cited by Hurle (1994) has indicated that oscillatory behaviour generated through double diffusion is the cause of striations along the growth axis in directional solidification. The point of this paper, however, is to show that oscillatory behaviour need not arise merely from opposing forces that are seen in thermo-solutal, otherwise known as double-diffusive, convection. Such oscillatory behaviour can arise by geometrical effects. Indeed as the crystal grows, the aspect ratio of the liquid phase changes and there are certain aspect ratios where the energy states may coexist leading to codimension-two points that can cause oscillatory convection.

S. K. Wilson (*Department of Mathematics, University of Strathclyde, Glasgow, UK*). I complement the authors on a penetrating investigation of a complicated physical situation. As I understand it, they have found examples of slow oscillations between two different steady flow patterns in the vicinity of codimension-two points calculated theoretically using linear theory for the onset of steady convection in a finite-sized container. May I ask if truly oscillatory (rather than quasi-steady) convection is ever observed, and if it would be possible to undertake the same kind of investigation for the onset of oscillatory convection?

D. Johnson. Yes, under certain circumstances, we believe that oscillatory convection can be observed in liquid–gas bilayer experiments. Theoretical calculations that were done indicate the absence of such convection at the onset. In that case what is the origin of oscillations in our experiments? The answer lies in the fact that a Hopf bifurcation lies in the vicinity of the onset but only in the post-onset region. The Hopf bifurcation mode or oscillatory mode was excited by the presence of codimension-two points. These points were generated by the fact that at certain aspect ratios two competing flow states coexist and in a manner of speaking the system wants to choose between the flow states leading to continual oscillations.