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Convective temperature oscillations in molten gallium

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An experimental study of the occurrence of temperature oscillations in molten gallium contained in a rectangular boat and heated from the side is reported. The dependence of the critical temperature difference across the boat for which oscillations set in upon the boat dimensions and upon the strength of a transverse magnetic field is described. The dependence of the frequency of oscillation on these parameters is also reported together with measurements of the variation of the phase of the oscillations over the top surface of the melt. The results are discussed in relation to the theory in the companion paper by Gill (1974).

1. Introduction

Very recent studies of finite amplitude convection in Bénard cells have shown that regular oscillatory motions can occur superimposed on top of the usual steady cellular motions. Oscillatory temperature variations have been obtained in gases (Mitchell & Quinn 1966; Willis & Deardorff 1970), liquid metals (Harp & Hurle 1968; Verhoeven 1969) and inorganic liquids (Krishnamurti 1970*a*, *b*; Willis & Deardorff 1970; Carruthers & Pavilonis 1968). Very little theoretical work has been performed in this field. However Fromm (1965) computed temperature and flow distributions for two-dimensional roll cells of finite amplitude in a laterally confined Bénard cell and found that for Rayleigh numbers greater than 10^6 the distributions oscillated in time. Very recently Busse (1972) has proposed a model to account for the oscillatory motion of roll cells in low Prandtl number fluids which are heated from below.

This paper is concerned with the occurrence of an oscillatory motion superimposed upon a steady convective flow but for the case in which vertical surfaces are maintained at different temperatures. In this case there is no threshold driving force; any finite difference in temperature between the two surfaces leads to steady convective motion with fluid rising at the hot surface and descending at the cold one. Hurle and co-workers (Hurle 1966, 1967; Hurle, Gillman & Harp 1966; Jakeman & Hurle 1972) have reported some preliminary experiments using a horizontal cell containing molten gallium on which is impressed an axial heat flow. These authors observed the onset of regular sinusoidal oscillations of temperature in the melt at a critical axial temperature gradient. In this paper we present the results of a detailed study of this phenomenon, which include measurements of the dependence of the frequency and the critical temperature gradient on cell dimensions and on the strength of a transverse magnetic field.

Somewhat similar measurements using liquid mercury have recently been made by Skafel (1972). Finally we compare our results with the theory of Gill (1974). This theory is the only one which appears to relate at all to the experimental results but it does not account for certain significant features such as the dependence of frequency on spatial dimensions.

Skafel (1972) performed most of his experiments with an annulus having radial heat flow. He measured mean temperature profiles and the power spectra of the fluctuations observed above threshold. Spatial correlation measurements indicated the existence of a progressive wave in the circumferential direction. Quantitative comparison with the present results is not possible but observations on temperature profiles and the dependence of the critical Rayleigh number on spatial dimensions are in qualitative agreement.

The present work was undertaken in an attempt to understand the origins of unsteady convection in fluids which have a low Prandtl number and to devise means of eliminating it because it has recently been shown (Hurle 1967; Müller & Wilhelm 1964; Chedzey & Hurle 1966; Utech & Flemings 1966; Cockayne & Gates 1967; Kim, Witt & Gatos 1972) that this form of convection is the cause of an unwanted inhomogeneity in the distribution of solute in single crystals of metals, semiconductors and oxides grown from the melt. Basically the fluctuating temperature produces fluctuations in the rate of growth of the crystal which in turn produce a banded distribution of solute in the crystal.

2. Apparatus

2.1. *Convection cell*

The experiments reported in this paper were performed using an improved version of the apparatus described in a previous paper (Hurle 1966). A photograph of the improved apparatus appears as figure 1 (plate 1). Molten gallium was contained in a U-sectioned pyrophyllite channel of length 10 cm having internal depth 1·5 cm, internal width 1·3 cm and a wall thickness of 0·5 cm. Additional experiments were performed with channels of internal width 1·0 cm and 0·65 cm. The ends of the channel were defined by a pair of copper thermodes which were maintained at different temperatures (shown schematically in figure 2).

Heat was supplied to one thermode by a resistance furnace fed from a constant-voltage transformer via an autotransformer while the other thermode was water cooled from a constant-head supply (flow rate 0·55 l/min). The end faces of each thermode were protected from corrosion by $\frac{1}{16}$ in. thick molybdenum tips. Although the thermodes fitted closely in the boat it was necessary to seal the hot thermode in place with 'araldite' in order to prevent loss of gallium. Losses from around the cold thermode were prevented by a little silicone vacuum grease smeared on the sides. The cold thermode could therefore be moved, enabling the length of the melt to be varied. To prevent oxidation of the gallium and vertical heat loss a thin film of DC705 silicone oil was applied to the melt surface. Lateral heat loss from the thermodes was minimized by enclosing exposed parts in Sindanyo packing pieces. The boat, thermodes and muffle furnace were mounted on a rigid supporting framework which also carried a $1\frac{1}{2}$ in.

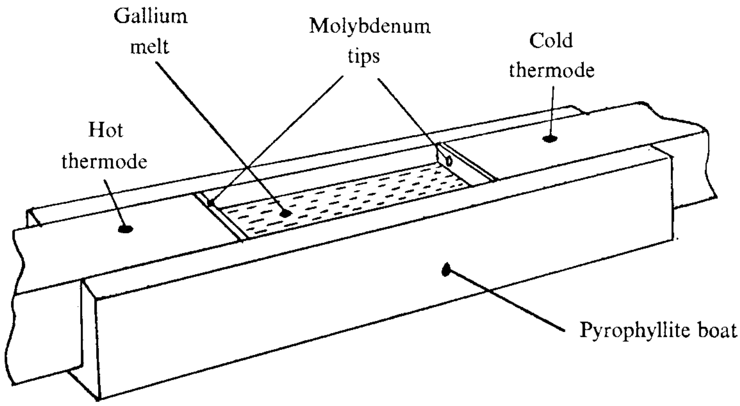


FIGURE 2. Schematic diagram of convection cell.

electromagnet, water cooled from a separate high-pressure supply. The boat was positioned on a Sindanyo supporting block midway between the pole-pieces so that a horizontal transverse magnetic field could be applied to the whole melt. A micromanipulator, allowing three degrees of freedom, was rigidly fixed alongside the cold thermode so that a thermocouple temperature probe could be positioned anywhere within the melt. The whole apparatus was provided with levelling screws and rested on a slate bed table to minimize vibration.

2.2. *Measuring thermocouples*

Earlier experiments had indicated the need for a very fine thermocouple probe if temperatures deep in the melt were to be measured without upsetting the local flow patterns. After some unsuccessful attempts to sheathe 0·002 in. chromel–alumel (type K) wires in separate glass capillaries and then to draw these out inside a third capillary, it was decided to attempt to draw down 3·15 mm O.D. twin-bore silica tube and then to pass the wires through the narrow bores (attempts to draw the tube with the wires already inside were unsuccessful: the wires broke under the strain). By this method twin-bore silica capillaries were made as small as 0·010 in. O.D., through which 0·002 in. thermocouple wires could be easily passed. The thermocouple junction was protected by being painted with a suspension of boron nitride in water. After evaporation a hard cap remained which was impervious to attack by the gallium and was also a good thermal conductor.

The principal improvement in the present apparatus over that used in the initial experiments was the use of open-ended boats which permitted a more nearly axial heat flow. (The thermal conductivity of heat-treated pyrophyllite is $3{\cdot}5$–$5{\cdot}5 \times 10^{-3}$ cal cm^{-1} s^{-1} °C^{-1} depending on the orientation with respect to the bedding plane (Carte 1955), while the value for molten gallium is $8{\cdot}1 \times 10^{-2}$ cal cm^{-1} s^{-1} °C^{-1}, from which it can be seen that the radial heat flow through the pyrophyllite will be small.)

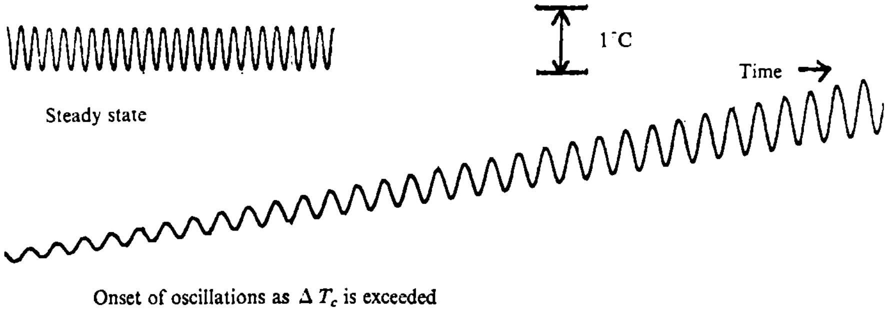


FIGURE 4. Typical recorder traces just above threshold conditions.

2.3. *Instrumentation*

The thermocouple output was fed via a backing-off circuit employing a Mallory cell and precision decade resistors to either of two Honeywell–Brown pen-recorders having a full-scale deflexion of 1 mV.

Phase measurements were made using a second thermocouple, identical to the first, placed in one corner of the melt and plotting Lissajous figures using the two thermocouple outputs. The thermocouple outputs were fed through a pair of Keithley type 149 milli-microvoltmeter/amplifiers which were adjusted to remove the d.c. level and which amplified the small a.c. components (of order a few μV) up to $\sim$ 1 V. The response of the amplifiers was flat up to 1 Hz. To remove harmonic components from the signals, the outputs from the amplifiers were fed to a pair of tunable band-pass active filters and then, through variable attenuators, to an X–Y plotter. By this means relative phases could be measured with a precision of better than $\pm 10°$.

Screened low-noise switching circuits were used to switch the thermocouple outputs either through the amplifier and filter or onto the pair of potentiometric recorders for display. A photograph of the whole apparatus (but excluding the X–Y plotter) is shown in figure 3 (plate 2). The band-pass active filters were tunable over the range 0·02–2 Hz and could be set to have a 'Q' factor of 20 or 40.

3. Results

Small amplitude sinusoidal oscillations of frequency ω set in when the axial temperature difference exceeds a critical value ΔT_c. Both ω and ΔT_c depend on the dimensions of the boat and ΔT_c depends additionally on the strength H of the transverse magnetic field . Typical signals, just above threshold conditions, are shown in figure 4.

The critical temperature difference exhibits some hysteresis: the value of ΔT_c at which oscillations first set in on slowly increasing ΔT is slightly greater than the value at which oscillations become undetectable on cooling slowly. The quantity measured was the value obtained on slowly increasing ΔT.

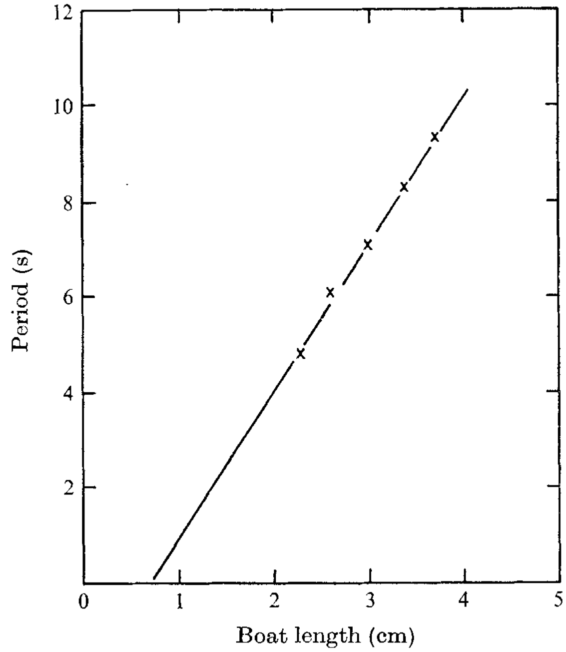


FIGURE 5. Variation of the period of oscillation with boat length for a boat of width 1·3 cm and a depth of gallium of 1·2 cm.

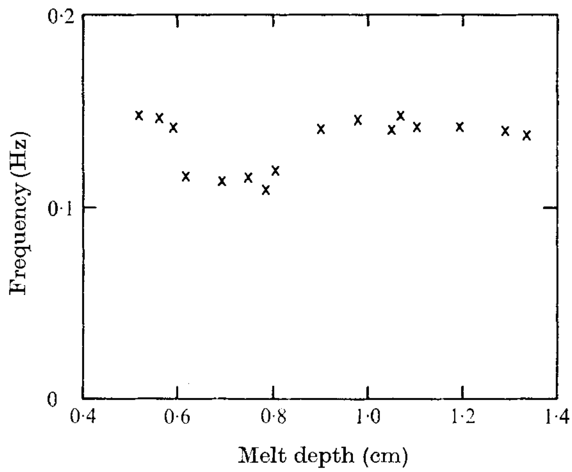


FIGURE 6. Variation of the frequency of oscillation with depth of gallium for a boat of width 1·3 cm and length 3·0 cm.

3.1. *Dependence of frequency on boat dimensions*

3.1.1. *Dependence on length.* The period $2\pi/\omega$ of the first oscillation to appear as ΔT was increased was measured for a boat of width 1·3 cm with a depth of gallium of 1·2 cm for various boat lengths (with no magnetic field). The results are shown in figure 5. The period varies linearly with length with an intercept on the length axis of $\sim$ 0·7 cm. This reproduces the results obtained by Hurle

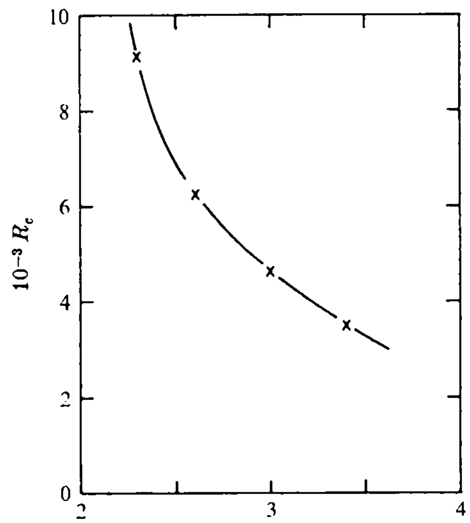


FIGURE 7. Dependence of the critical Rayleigh number on boat length for a boat of width 1·3 cm and a depth of gallium of 1·2 cm.

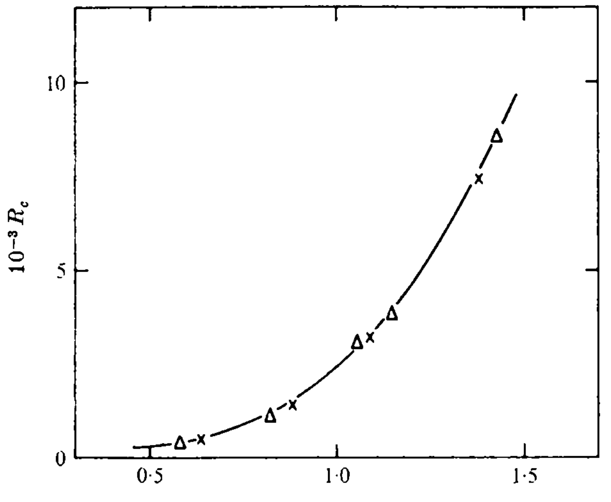


FIGURE 8. Dependence of the critical Rayleigh number on depth of gallium for a boat length of 3 cm and widths of (*a*) 1·3 cm (crosses) and (*b*) 1·0 cm (triangles).

(1967) on the original apparatus. Second and third harmonic components were sometimes observed as in the previous study (Hurle 1967). For $\Delta T \gg \Delta T_c$ large amplitude 'noisy' signals were obtained (Hurle *et al.* 1966).

3.1.2. *Dependence on depth.* Measurements as in §3.1.1 were made with a boat of width 1·3 cm and length 3·0 cm but with varying depth. The results are shown in figure 6. In the ranges 0·5–0·6 cm and 0·9–1·35 cm the period is essentially constant at 7 s. In the intermediate range of depths the period increases

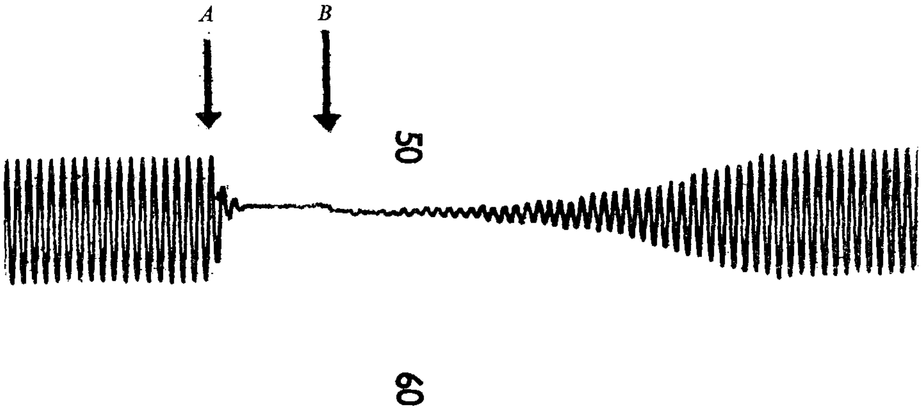


FIGURE 9. Effect of transverse magnetic field on oscillations. Field applied at A and removed at B. Oscillation period $= 6{\cdot}1$ s; maximum amplitude approximately $1{\cdot}3$ °C peak to peak.

to $\sim$ 9 s. There appears to be a change of mode over this range of depths but this has not been investigated further.

3.1.3. *Dependence on width.* This has not been systematically investigated but one experiment was performed with a boat of width 0·65 cm and length 3 cm and a melt depth of 1·2 cm. A frequency of 0·4 Hz was obtained. In another experiment with a boat of width 1·0 cm and the same length and depth a frequency of 0·12 Hz was obtained. This compares with the value, from figure 5, of 0·14 Hz for a width of 1·3 cm. These results are inconclusive; it is possible that the frequency observed with the narrowest boat was a harmonic of the basic mode.

3.2. *Dependence of ΔT_c on boat dimensions*

3.2.1. *Dependence on length.* The temperature difference at which oscillations first appeared was measured for a 1·3 cm wide boat with a 1·2 cm depth of gallium for various boat lengths. The temperature difference was obtained by moving the thermocouple along the centre-line of the boat until it touched each thermode in turn. The results are shown in figure 7, where ΔT_c has been expressed as a critical Rayleigh number $R_c = \alpha g \Delta T_c d^4/\nu\kappa l$, where α is the expansion coefficient, g the acceleration due to gravity, ν the kinematic viscosity, κ the thermal diffusivity, d the melt depth and l its length. Inserting values for gallium, $R_c = 320\,\Delta T_c d^4/l$.

3.2.2. *Dependence on depth.* Measurements, as in §3.2.1, were made with a boat length of 3·0 cm, boat widths of 1·3 cm and 1·0 cm and with varying depths of gallium. The dependence of R_c on depth is shown for the two boats in figure 8. R_c approaches a constant value of $\sim$ 300 for depths less than $\sim$ 0·5 cm and rises as d^n for greater depths, where $n = 3{\cdot}4 \pm 0{\cdot}5$. Any difference between the two boat widths is within the experimental error.

3.3. *Dependence of ΔT_c on magnetic field H*

The effect of a transverse magnetic field on the oscillations is shown in figure 9. A field just sufficient to quench the oscillation was applied at A and removed

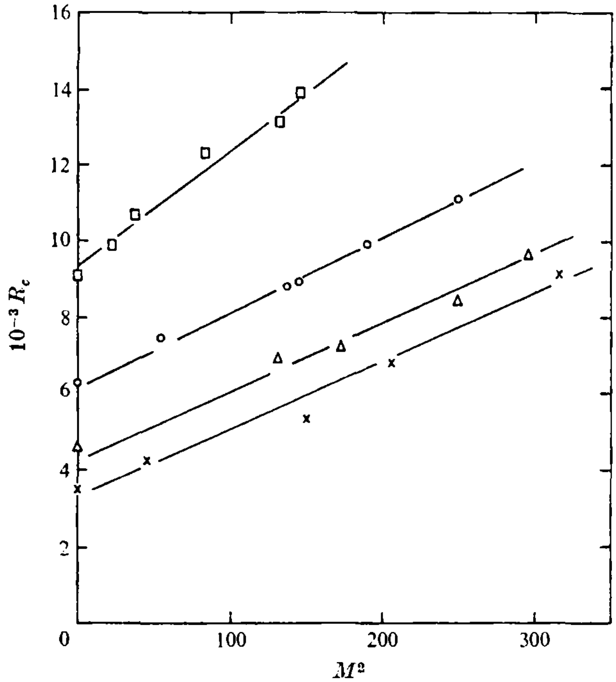


FIGURE 10. Dependence of the critical Rayleigh number on the square of the Hartmann number for a boat of width 1·3 cm and a depth of gallium of 1·2 cm. Boat length: □, 2·3 cm; ○, 2·6 cm; △, 3·0 cm; ×, 3·4 cm.

at B. Note the rapid rate at which the oscillations are quenched compared with their rate of regrowth.

The temperature difference $\Delta T_c(H)$ for each of a number of furnace current settings was measured as described above when the transverse magnetic field had been adjusted so as just to damp out the oscillations. The width and depth of melt were respectively 1·3 and 1·2 cm. Results, shown in figure 10, were obtained for boat lengths of 2·3, 2·6, 3·0 and 3·4 cm.

The magnetic field is expressed as the non-dimensional Hartmann number $M = (\sigma/\rho\nu)^{\frac{1}{2}}\mu dH$, where σ is the electrical conductivity, ρ the density and μ the permeability. For gallium $M = 4{\cdot}78 \times 10^{-2} dH$, where d is in cm and H in gauss.

The critical Rayleigh number is seen to increase linearly with the square of the Hartmann number. (There is some scatter in the data and it may be that some other functional dependence would give a better fit.) The coefficient of proportionality appears to increase with decreasing length of the melt over the range studied.

The Rayleigh number is observed to be proportional to the square of the Hartmann number at onset of stationary convection in a Bénard cell (Chandrasekhar 1961, p. 171).

Experiment	Conditions	Field (gauss)	Figure
1	$\Delta T < \Delta T_c(0)$	0	11 (*a*)
2	$\Delta T < \Delta T_c(900)$	900	
3	$\Delta T > \Delta T_c(0)$	0	11 (*c*)
4	$\Delta T_c(0) < \Delta T < \Delta T_c(70)$	70	
5	$\Delta T < \Delta T_c(3800)$	3800	11 (*b*)
6	$\Delta T > \Delta T_c$, off-axis flow	0	
7	$\Delta T_c(0) < \Delta T < \Delta T_c(3800)$, off-axis flow	3800	
8	$\Delta T < \Delta T_c$, off-axis flow	0	

TABLE 1

3.4. *Temperature distribution in the melt*

Time-averaged three-dimensional temperature distributions were constructed for several values of ΔT and H in order to obtain information about the nature of the convective flow. The boat dimensions used were: length 3 cm, width 1·3 cm with a depth of gallium of 1·1 cm. Temperature measurements were made at 30 s intervals at 128 points on a three-dimensional grid and a check for temperature drift during the measurements was made. This was never greater than 2 %. Temperature plots were made with and without the melt temperature oscillating and in the presence of a transverse magnetic field. In addition plots were made with an off-axis heat flow produced by covering half of each thermode with a thin strip of mica. Table 1 gives a complete specification of all the runs performed. Perspex models depicting the more instructive plots were made and are shown in figure 11 (plate 3). On these models the narrow black lines represent intervals of $100\,\mu$V ($\sim$ 2·5 °C), whilst the broad lines represent $500\,\mu$V ($\sim$ 12·5 °C) intervals.

3.5. *Phase measurements*

An initial measurement on a particular mode indicated a phase difference of 2π between the ends of the boat and, naively, it was at first thought that the oscillation consisted of a plane wave travelling along the boat (with wave velocity equal to the fluid velocity $\sim$ mm/s). However a more careful study showed that the phase measured along the centre-line of the boat did not change linearly with distance and it was decided to map the lines of constant phase over the entire top surface of the boat on a 1 mm grid. This proved to be very tedious but was very revealing.

The result of a typical plot, for a boat of length 3 cm and width 1·3 cm with a depth of gallium of 1·1 cm, is shown in figure 12. There exist two points, one near each end of the boat, into or out of which 2π's worth of lines of constant phase disappear (or appear). The question arises as to what determines the sense of rotation of these 'phase vortices'. The answer to this, illustrated in figure 13, appears to be the direction of the heat flow relative to the side walls of the boat. Heat flow which was slightly off-axis was obtained by covering one half of each thermode with mica strips. Figures 13 (*a*) and (*b*) show that the sense of rotation changes when the angle between the heat flow direction and the axis of the boat

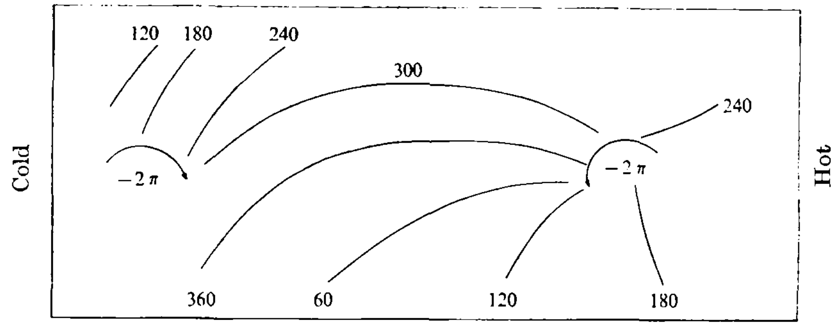


FIGURE 12. Spatial variation of the phase of the oscillations on the top surface of the melt.

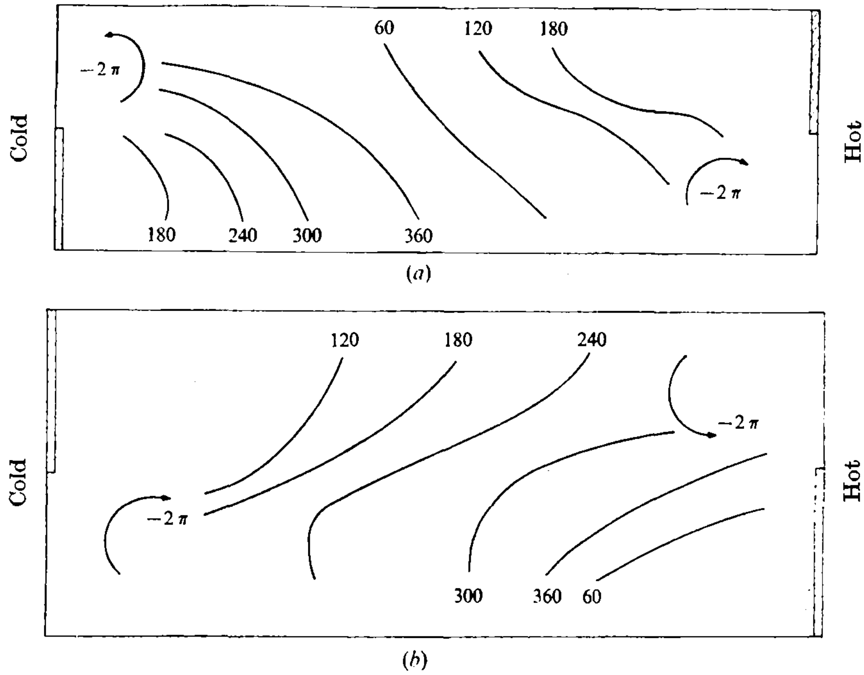


FIGURE 13. Spatial variation of the phase of the oscillations, showing the effect of off-axis heat flow.

changes sign. The use of an off-axis heat flow gave rise to very stable oscillations which were not readily destroyed by mechanical vibrations, draughts or other external disturbances. This observation appears to account for our early experience that the more carefully the experiment was set up the less successful we were in obtaining stable oscillations!

4. Discussion

The three-dimensional isothermal maps (figure 11) show that the time-averaged thermal distributions at temperature gradients above the critical value for oscillations (figure 11 *c*) are of the same form as those below the threshold for oscillation (figure 11*a*). Clearly then, the oscillations are a perturbation on the basic flow rather than the onset of an entirely different mode of flow. Additional evidence for this is provided by the fact that the onset of oscillations does not produce a significant increase in the Nusselt number. The basic flow is, from figure 11, a simple convection loop with liquid metal rising at the hot thermode, flowing along the boat and descending at the cold thermode. The curvature of the isotherms in horizontal planes is due to the insulating vertical side walls of the boat. Off-axis heat flow produces the expected distortion of the thermal field but does not appear to alter the basic mode of circulation.

The above suggests that a critical velocity of circulation has to be established in order to achieve oscillation and it may be, therefore, that the system is better described by a critical Reynolds number rather than by a critical Rayleigh number. The effect of the application of a transverse magnetic field is to damp the circulation as shown by the more equally spaced isotherms of figure 11 (*b*).

Perhaps the most significant pointer to the physical mechanism giving rise to the oscillations comes from the phase measurements (figure 12). Apart from the 'vortex' phenomena, which are almost certainly end effects, the phase lines run, rather surprisingly, parallel to the side walls of the boat. Thus the temperature (and the time-dependent component of the flow) exhibits a phase change of roughly 2π across the boat. A theory which is consistent with this observation is developed in the companion paper by Gill (1974). This predicts a neutral-stability curve which reduces in the low Prandtl number limit to

$$R \sim \frac{4\pi^4(1+\gamma^2)}{\gamma}\left(\frac{2}{3-\gamma^2}\right)^{\frac{1}{2}}, \tag{1}$$

corresponding to the oscillation period

$$\tau \sim \left(\frac{d}{2\pi}\right)^2 \left[\frac{2(3-\gamma^2)}{\nu\kappa}\right]^{\frac{1}{2}} \frac{1}{1+\gamma^2}, \tag{2}$$

where
$$\gamma = 2ld/mW, \tag{3}$$

l and m being positive integers, d the boat depth and W its width. The critical Rayleigh number of 1030 is achieved for $\gamma^2 = 0{\cdot}29$. This may be compared with the experimental values of R_c, which range from 410 up to 8600. The oscillation period predicted by (2) is also in fair agreement with the observed values of a few seconds. (For example, for typical values of d (1·2 cm) and W (1·3 cm) and with $l = 1$ and $m = 2$, we obtain, from (2), $\tau \sim 2{\cdot}1$ s, which compares with experimental values of from 4·8 s at a boat length of 2·6 cm up to 9·3 s at a boat length of 3·7 cm.) The ratio $(2d/W)^2 > 0{\cdot}29$ for all our experiments, so that we might expect to set $l = 1$ in (3), corresponding to one wavelength being contained in the boat width. This is consistent with the phase measurements. Since from (1) and (2) solutions are possible only for $\gamma < 3$ there is a further restriction on m, which may well be equal to one in some cases and two or more in others. This could explain the anomalous dependence of frequency on depth and width which we report, but quantitative comparisons of experiment and theory are not likely to be useful owing to the limitations of the theoretical model. For certain ranges of depths and widths one might expect regions of monotonic

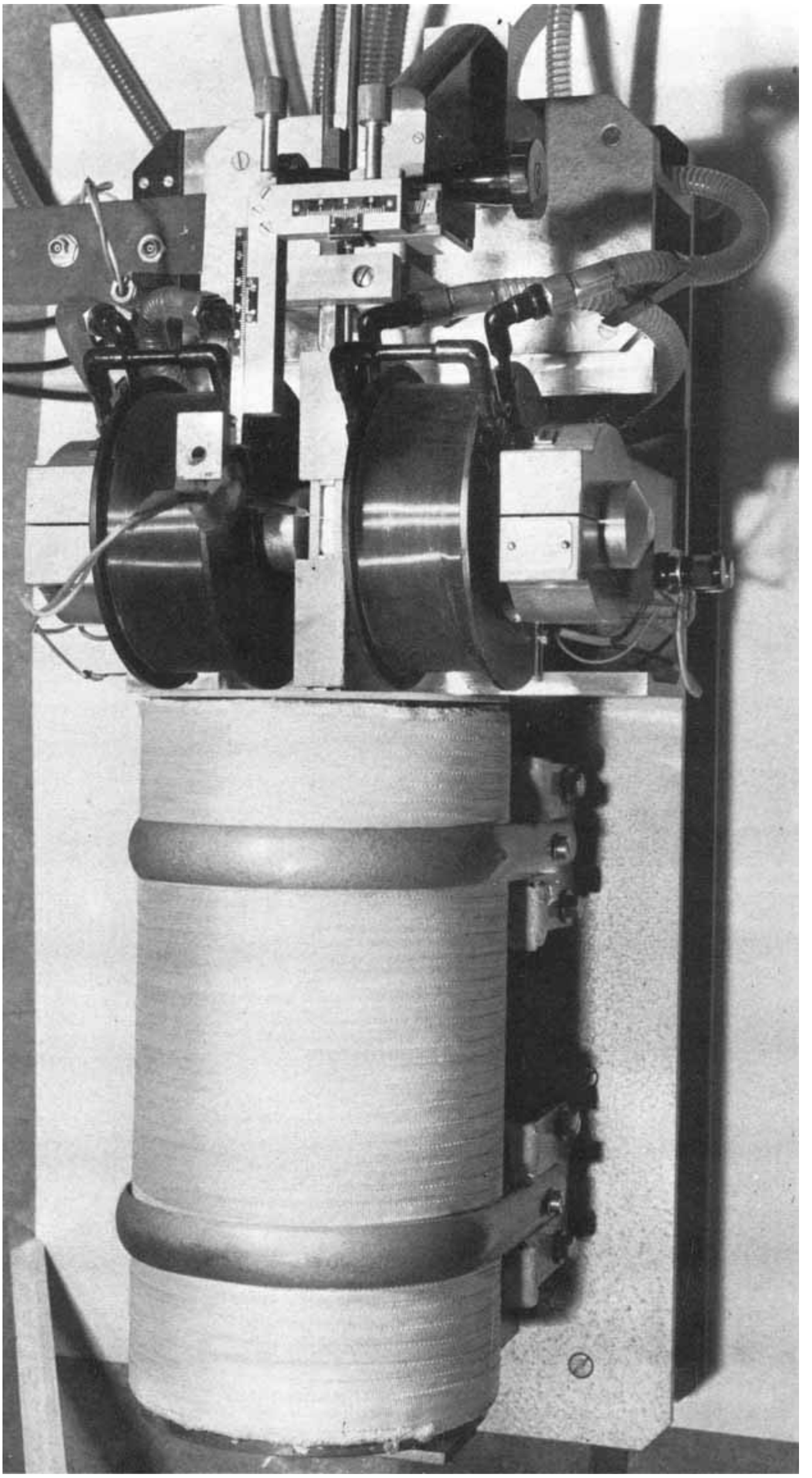


FIGURE 1. Photograph of the convection cell. (Plan view.)

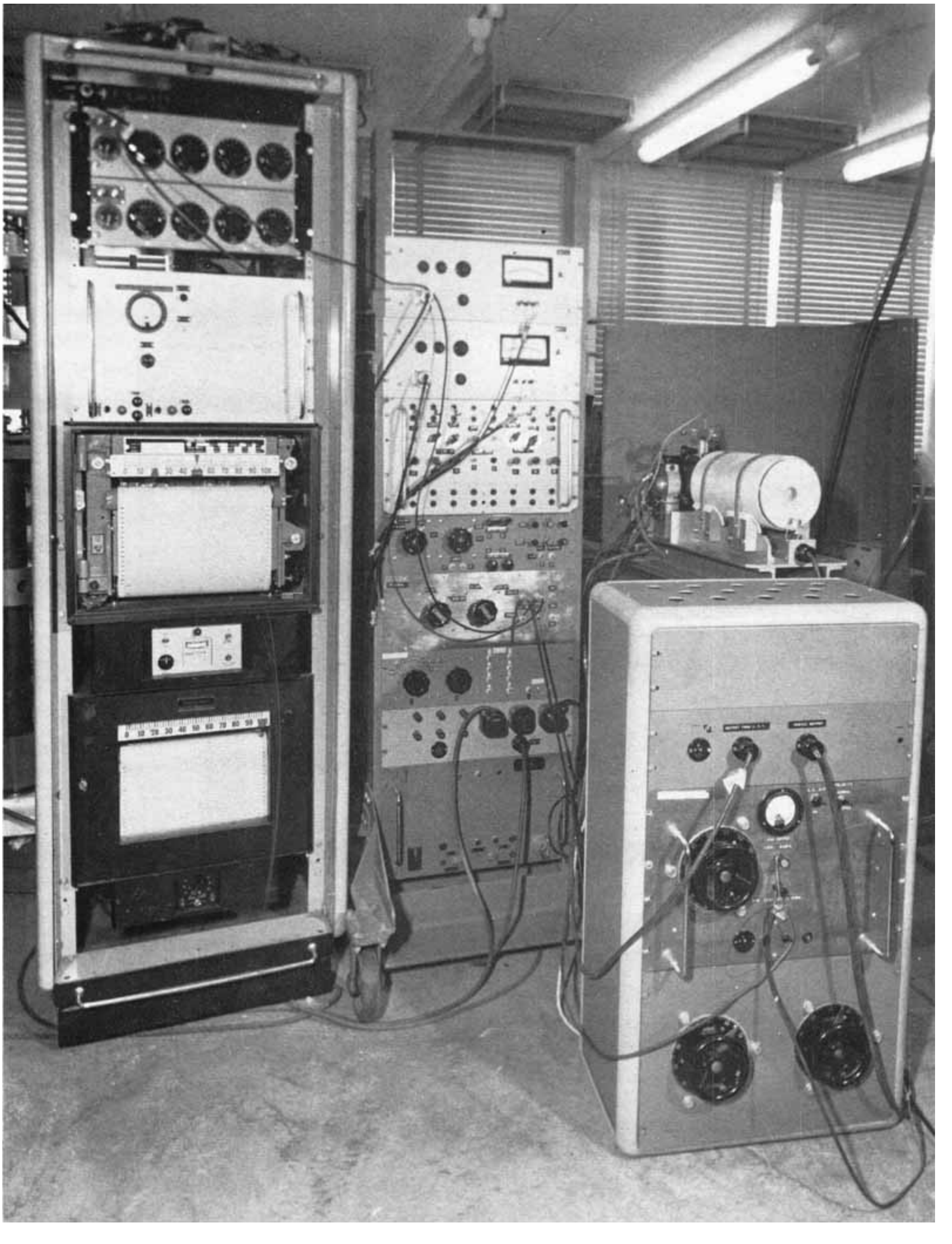
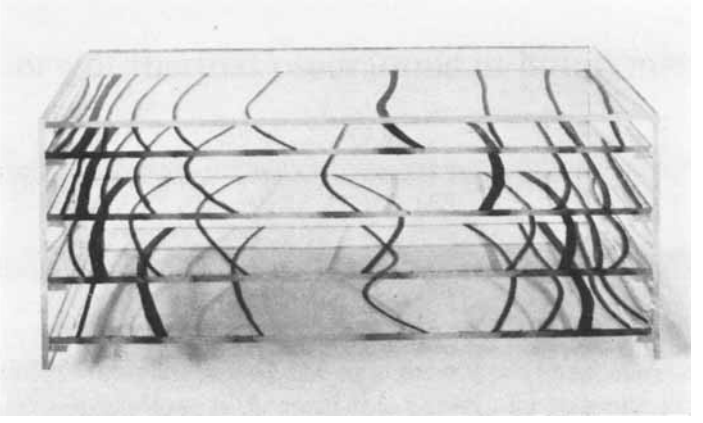
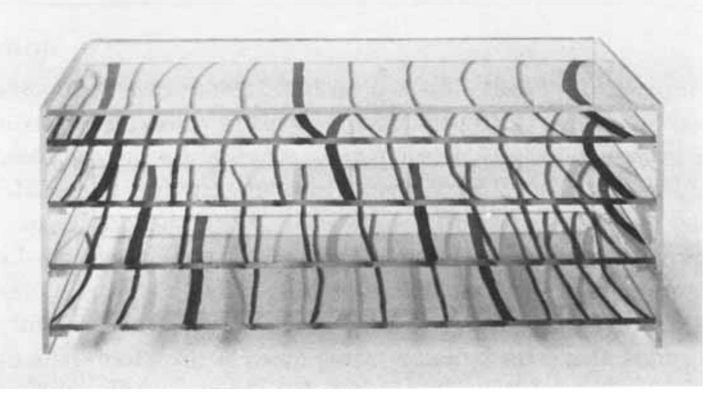


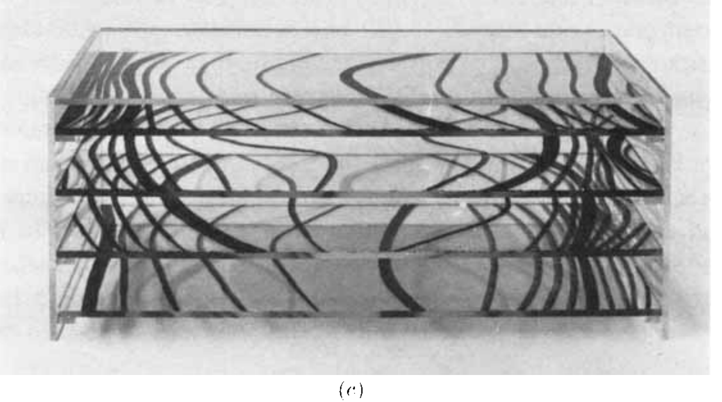
FIGURE 3. Photograph of the apparatus.



(*a*)



(*b*)



(*c*)

FIGURE 11. Perspex models of the temperature distribution in the liquid cell. (*a*) ΔT below threshold for oscillations. (*b*) ΔT below threshold with transverse field of 3800 gauss. (*c*) ΔT above threshold for oscillations. The hot thermode is on the right-hand side and the perspex box defines the extremities of the melt.

behaviour between mode jumps. In such regions the dependence of *R and* τ on these dimensions should be given by (1) and (2). The results presented in figure 8 appear to be consistent, with this type of behnviour.

Although Gill's theory cannot. predict effects which arise owing to the finite length of our containing boat, and in particular the linear dependence of τ on length, it is clear from the above discussion that it is consistent, with several significant observations which we have reported.

Finally, to return to crystal growth; we have argued that the effect of a transverse magnetic field is to clamp the oscillations by reducing the Reynolds number for the basic flow to below some critical value. Our objective in crystal growth is to remove the oscillatory components. (The steady flow produces a desirable mixing of any solutes present. and minimizes the possibility of the occurrence of constitutional supercooling (Hurle 1961).) Clearly, then, the optimum magnetic field to be used in horizontal crystal growth is that field which is just sufficient to damp out the oscillations.

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