Convective instabilities in complex systems with partly free surface

Dietrich Schwabe
1. Physikalisches Institut der Justus-Liebig-Universität Giessen, H.-Buff-Ring 16, 35392 Giessen, Germany
E-mail: Dietrich. Schwabe@physik.uni-giessen.de

Abstract. Experiments and observations and some selected theoretical studies of thermocapillary instabilities are reviewed and presented together with new unpublished work. We start with simple idealized model systems of pure thermocapillarity and add to them more complex features like gravity for ces, temperature gradients inclined to the free surface, static and dynamic surface deformations, solutocapillary effects and reacting or moving crystal boundaries (like during unidirectional solidification). Many effects and instabilities are demonstrated in video clips which can be downloaded from http://meyweb.physik.uni-giessen.de/1_Forschung/crystalgrowth/video/homepage.html. We try to point out the relationship of thermocapillary instabilities in the more complex systems with those in theoretical studies where the names of these instabilities have been coined.

1 Introduction
1.1. Outline
This paper deals mainly with surface-tension-driven instabilities like those of thermocapillary flow or the formation of cellular convective structures. It is motivated by the geometries and boundary conditions found in crystal growth from the melt. Therefore, convective effects induced by the large temperature gradients connected with these crystal growth methods are in the centre of this paper although solutocapillary effects are touched, too. The geometrical and thermal boundary conditions in crystal growth systems can be rather complex, including curved free surfaces like menisci, the combined action of surface tension forces and buoyancy, the presence of moving boundaries (e.g. the growing crystal interface) and the interaction of buoyant-thermocapillary convection with forced convection by crystal rotation in the Czochralski technique. The systems used in melt crystal growth are partly bounded by walls. Therefore, end-effects and boundary layers need to be considered with their special localized instabilities. Nevertheless, we will start with a review and discussion of unbounded simplified model systems and their instabilities before discussing more complex situations, - adding complexity step by step.

This paper is mainly experimental. The instabilities are demonstrated in video clips and some measurements, but almost never fully characterized because not yet numerically simulated.

The literature on liquid interfacial systems, on their phenomena and instabilities, is vast and not even the special field of this paper can be covered completely. We apologize for having not cited some important work of our colleagues. The reader is referred to the following books [1-6], the review articles [7-8] and the cited original literature which can serve a guides to further problems and further literature.
1.2. Simple models

We consider liquid configurations like layers or liquid in vessels with free upper surface or liquid bridges (LBs) and define the following nondimensional groups, e.g. for the case of horizontal liquid layers.

| Group                        | Expression |
|------------------------------|------------|
| Aspect ratio                 | \( \frac{A = \text{lateral extension } L}{\text{liquid depth } d} \) |
| Prandtl number               | \( \text{Pr} = \frac{\nu}{\chi} \) |
| Rayleigh number              | \( \text{Ra} = g \cdot \beta \cdot \Delta T \cdot d^3 \cdot \nu^{-1} \cdot \chi^{-1} \) |
| Horizontal Marangoni number | \( \text{Ma}_h = \left| \frac{\partial \sigma}{\partial T} \right| \cdot \left( \frac{\Delta T_w}{L} \right) \cdot d^2 \cdot \eta^{-1} \cdot \chi^{-1} \) |
| Vertical Marangoni number    | \( \text{Ma}_v = \left| \frac{\partial \sigma}{\partial T} \right| \cdot \Delta T_v \cdot d \cdot \eta^{-1} \cdot \chi^{-1} \) |
| Dynamic Bond number          | \( \text{Bo}_{dyn} = \frac{\text{Ra}}{\text{Ma}} \) |
| Surface tension number       | \( \text{S} = \rho \cdot \sigma \cdot d \cdot \eta^{-2} \) |
| Capillary number             | \( \text{Ca} = \frac{\sigma^{-1} \cdot \left| \frac{\partial \sigma}{\partial T} \right| \cdot \Delta T \cdot L^{-1} \cdot d}{\chi} \) |

with kinematic viscosity \( \nu \), dynamic viscosity \( \eta \), thermal diffusivity \( \chi \), surface tension \( \sigma \), temperature dependence of surface tension \( \partial \sigma / \partial T \), volume expansion coefficient \( \beta \), density \( \rho \), Earth gravity \( g \), temperature difference between the boundaries \( \Delta T \). In certain cases one can replace \( \Delta T_w / L \) by \( \text{grad}_b T \).

We denote the bifurcation point by the superscript "c"; e.g. \( \text{Ra}^c \) is the critical Rayleigh number. We consider first unbounded (\( \infty \)-extended) liquid layers with free surface a) heated from below and cooled from above
b) subject to a horizontal temperature gradient

**Figure 1.** Hexagonal convection pattern in a liquid layer heated from below and cooled from above, visualized by a shadowgraph technique. From [1].

**Figure 2.** Stability diagram for the Bénard-Marangoni-Instability including the action of gravity. From [11].

The basic state of case a) is a stagnant layer, showing a transition to the well-known hexagonal cellular convection pattern [1, 9] of the Bénard-Marangoni-Instability (BMI) (figure 1). Pearson [10] analyzed the case excluding gravity and Nield [11] included gravity. The Marangoni effect and buoyant forces reinforce each other, lowering the critical \( \text{Ma} \) or \( \text{Ra} \), respectively and influence the pattern (figure 2). We find cellular patterns of different type in deep vessels compared to the one in thin layers and again different ones under microgravity (pure Marangoni effect) (figure 3) [12].

We can consider for case b) a linear flow profile without return flow (figure 4a) and thermocapillary flow with return flow (figure 4b) like Smith and Davis [13]. The basic states of these
cases are dynamic layers. The stability analysis of these dynamic layers indicated as most dangerous instability for the linear flow the so called linear rolls (roll-axis aligned with the basic flow) and, as most dangerous instability of thermocapillary flow with return flow the so called hydrothermal waves [13]. The wave vector of the hydrothermal waves is inclined to the applied temperature gradient, the inclination growing with decreasing Pr.

**Figure 3.** Convection pattern of the pure Marangoni-Instability in a small circular layer with diameter D with free upper surface under microgravity ($A = D/d = 5, Ma = 7 Ma^3$). Visualization with fine aluminum flakes. The up- and down-flow regions are dark. From [12].

**Figure 4.** Basic state flow profiles of dynamic thermocapillary layers. A temperature gradient in the free surface or a temperature difference between the lateral walls drives this flow.

(a) linear flow profile of thermocapillary flow (without return flow)
(b) thermocapillary flow with return flow

These flow profiles can be modified by the action of gravity.

Hydrothermal waves (HTWs) in a thin layer of silicone oil (on a metal plate which is impressing the temperature gradient and driving thermocapillarity) in rectangular symmetry is shown in figure 5 [14]. The HTWs travel from "cold" to "hot" in this experiment and for this Pr as predicted by [13]. For radially oriented temperature gradients like in cylindrical annuli, the HTWs become Archimedian spirals (figure 6) because of the constant angle between wave vector and temperature gradient [15].

Figure 7 shows an example of HTWs in an open cylindrical annulus with $A = 1$ and $d = 20$ mm under microgravity [16]. The corresponding numerical simulation is shown in figure 8 [17]. The HTWs are found to be travelling azimuthally but at higher Ma as well to form standing waves or waves travelling from one or different sources in opposite direction and interfering (see examples in figure 25 in [18]).
Figure 5. Hydrothermal waves in a layer from silicone oil with $d = 1.0$ mm, travelling under an angle of $24^\circ$ between temperature gradient and wave vector from "cold" to "hot." (From "left" to "right"). Visualization with IR camera. From [14].

Figure 6. Hydrothermal waves in an open annulus with a small-diameter inner cooled wall have the form of an Archimedian spiral. From [15]. Shadowgraph technique ($d = 1.9$ mm, $\Delta T = 14.5$ K, $Pr = 10$).

Figure 7. Hydrothermal waves under microgravity in an open annulus with $d = 20$ mm and $A = 1$ heated from the outside and cooled in the centre (silicone oil, $Pr = 6.8$). An IR camera shows the temperature distribution at the free surface. From [16]
(a) five wavetrains at $Ma \approx 2 \cdot Ma^c$ travelling azimuthally (experimental)
(b) nearly chaotic at $Ma \approx 4 \cdot Ma^c$ (experimental)

The open annulus investigated in [16] and [17] can be taken as a model for a Czochralski crystal growth system, where a crystal (cold cylinder) is grown and pulled out from the free surface of the melt in a crucible (figure 9). The main differences to crystal growth are:

− the cold side in the centre is a crystal with the solid-liquid interface at approximately the level of the melt
− the thermal conductivity of the crystal is comparable to that of the melt
− gravity is acting because of large temperature (density) differences
− the free surface is normally strongly cooled by radiative heat loss because of the high working temperature (melting points up to 1900° C).

The much higher complexity of the Czochralski system will be discussed later.
Figure 8. Simulated surface temperature distribution for the same annular gap as in figure 7 with Bi = 0. From [17]
(a) slightly above Ma
(b) at approximately 3 \cdot Ma
The five wavetrains (m = 5) rotate clockwise and m does not change when increasing Ma to 5Ma.

Figure 9. Sketch of the Czochralski crystal growth technique and the thermocapillary open annulus investigated in [16, 17]. From [17].

An example of travelling hydrothermal waves in the meniscus attached to a cooled dummy crystal is shown in figure 10 [19]. The HTWs develop only in the range of the cooled crystal because of the sufficiently large radial temperature gradient there. They travel well ordered in azimuthal direction. The attempt to damp their amplitude and/or azimuthal travelling speed by counter rotation of the dummy crystal failed; the HTWs simply change their rotational direction to that of the crystal for not to large counter rotation speeds.

The HTWs do not travel in radial direction in Figs. 7 and 10 compared to figure 6. Their travelling direction is only perpendicular to the temperature gradient and not inclined to it as in figure 5. The reason for the suppression of the HTW-component parallel to the temperature gradient is the confinement of the HTW to the region near the cold thermal boundary layer with a sufficiently large temperature gradient. Only if the temperature gradient is impressed by a metal bottom with a
sufficiently large gradient, HTWs can develop in the whole area of this metal bottom with both wave-components. The HTWs will develop (for Pr > 1) only near and in the thermal boundary layers in case of side-heating and side cooling, and with only one wave component suppressed. This has been observed in side-heated shallow annular pools with thermally insulated bottom [20] shown in figure 11. We can call these HTWs near thermal boundary layers "degenerated" or "frustrated" HTWs because one wave component – the one in the direction of the applied temperature gradient – does not develop; the degenerated HTWs travel parallel to the heated or cooled wall.

**Figure 10** HTWs in the meniscus at the cold dummy crystal (copper) with \( r = 4 \) mm, \( \omega_{\text{crystal}} = 0 \) in a crucible with \( R = 20 \) mm, liquid height \( H = 30 \) mm, \( \Delta T = 25 \) K. Observation under diffuse illumination through the crucible bottom. The small inner grey circle is the face of the dummy seen from below. A thermocouple (black wire) comes from above and white schlieren (generated by the jet of cold liquid falling down from the cold dummy) are visible on the left side of the grey dummy-face. The HTWs are only visible in the video clip on the web page given in the abstract under “HTW in the meniscus” [19].

**Figure 11.** HTWs in a shallow liquid layer (Pr = 17) in an annular gap near the heated wall with the bottom and outside of the gap cooled. The HTWs deform the free surface and this deformation can be visualized by surface reflection shadowgraphy. (L = 20 mm, \( d = 1.8 \) mm, \( \Delta T = 34 \) K). From [20]

HTWs are as well degenerated if the gap between the hot and the cold wall is smaller than their wavelength component in this direction. In this case they travel azimuthally in annular gaps or cross the temperature gradient in channels because of this geometric restriction [21-22]. Stationary rolls have been observed in these so-called channels (figure 12) at larger depth \( d \) (e.g. \( d > 3 \) mm) [22] with the roll axis parallel to the applied temperature gradient (wave vector perpendicular to the applied horizontal temperature gradient). These stationary rolls (SRs) could be related to the "linear rolls" of Smith and Davis [13]. Buoyancy might be the reason for these stationary rolls though thermocapillarity is dominating (to be discussed later). On the one hand the SRs have not been observed under microgravity for \( 2.5 \) mm \( \leq d \leq 20 \) mm, indicating that gravity is essential. On the other hand these 3D stationary rolls have been observed down to \( d = 3 \) mm, that is, at small gravity influence [23]. The increase of the flow amplitude of these stationary rolls indicates that the transition
from the basic state to the roll state is not a Hopf bifurcation [23]. A bifurcation of 2D buoyant thermocapillary convections to 3D (like SRs) is indicated in numerical calculations [24].

**Figure 12.** (a) Linear rolls by shadowgraphy. View through the channel-like layer ($d = 6 \text{ mm, } \Delta T_x = 6 \text{ K, } L_x = 30 \text{ mm, Pr} = 10$). The cold side is down, menisci at cold and hot side. From [37]

(b) Critical temperature differences for travelling waves (TW) = HTW and stationary rolls (SR) = LR in layers with different height $h = d$ in a rectangular gap with glass bottom with $L = 10 \text{ mm (Pr} = 10)$

Liquid bridges (LBs) represent a special rotationally symmetric gap. The free surface is parallel to the gravity vector, the LBs must be short because of gravity and, the HTWs, which are travelling in azimuthal direction, must obey the periodic boundary condition $2 \pi a = m \cdot \lambda$ with $a = \text{LB-radius, } \lambda = \text{wavelength of the HTWs and } m \text{ an integer.}$ The temperature – and flow oscillations in the LBs have been investigated long before the studies of the related annular gaps [25].

**Figure 13.** The amplitude of temperature oscillations in a liquid bridge with $Pr = 8$ heated from the top by $\Delta T$. The temperature oscillation amplitude increases as $(\Delta T - \Delta T_c)^{0.6}$ above the threshold. The exponent 0.6 is near 0.5 of a Hopf bifurcation. From [25].

**Figure 14.** The squared amplitude of the temperature oscillations $A^2$ in a LB with aspect ratio $A = 15$ under microgravity over the applied temperature difference $\Delta T$ is indicating a Hopf bifurcation. From [26]. Signals from two thermocouples (7Az, 8Az).
It was found that the transition from steady flow to oscillatory flow (that is the case of degenerated HTWs) is a Hopf bifurcation (figure 13) [25]. In a microgravity experiment with an extremely long LB (A = L/a = 5) the degeneration of the HTW was very effectively removed and a large axial wave component was observed besides the azimuthal one [26]. And it was found that the transition from the basic pure thermocapillary flow with return flow in the LBs is a Hopf bifurcation (figure 14) [25].

The advantage of the confined very large temperature gradient near a heated or a cooled metal wall seems to be the possibility to observe the so called "surface waves" predicted by Smith and Davis [27]. Thermocapillary surface waves (SWs) have been observed until now only in connection with such thermal boundary layers [20, 28, 29] at the heated wall. SWs are due to a shear mechanism which is largest at the hot side in the boundary layer with the highest surface flow velocities. The dynamic deformation of the free surface is crucial for this instability whereas HTWs are found by linear stability analysis for the case of a nondeformable free surface. Figure 15 shows a shadowgraphic image of the free surface of an annular gap heated from the inner cylinder (r_i = 20 mm) and cooled from the outside container (r_o = 40 mm) and with insulating bottom. The SWs in figure 15 travel from "hot" to "cold" and azimuthally.

Figure 15. Surface wave (SW) with m = 6 in a 20 mm wide annular gap heated from the inside cylinder with r_i = 20 mm (d = 3.1 mm, \Delta T = 24 K). The free surface in the gap is prepared as totally flat (no menisci at both sides) to allow shadowgraphy of light reflected from the free surface to visualize the SWs. From [28].

Figure 16. Streamlines of multicellular (steady) thermocapillary convection in Pr = 4 fluid heated from the right-side. The cell axes are perpendicular to the temperature gradient. For larger Ma HTWs can travel from "cold" to "hot" (or vice versa depending on Pr) and the multicells will oscillate according to the HTW frequency. From [30]. The multicellular state can serve as basic state for HTWs [14, 16].

Hydrothermal waves (HTWs) [13, 14], surface waves (SWs) [27], stationary rolls or linear rolls (SRs, LRs) [13, 22] and multicells (MCs) [30, 20, 14] (MCs see figure 16) have now been observed most likely in many configurations with different aspect ratios and thermal boundary conditions, but it is not easy to tell which case was observed and to compare the different works. In some experiments submerged heaters have been used [31-35] or the free surface was heated locally from above [36]. The temperature gradient along the free surface can be prescribed as linear like in the theoretical paper [13, 14], or it can be supplied by heated/cooled walls with thermal boundary layers like in liquid bridges [25] and annular gaps [16] but can as well be provided as a gradient in the conducting bottom [30]. The thermal boundary condition at the bottom can have a large influence on the type and threshold of the various thermocapillary instabilities (e.g. depending on the thermal conductivity of the bottom (glass [15, 22, 37, 38], quartz glass [23] or metal [30, 39])). As well the degree of confinement
(channel [30, 22] or LB [25, 40] with strong confinement or more extended layers or LBs [14, 15, 37, 26]) make a difference for the appearance of the instabilities. Interesting is as well the case where the "bottom" is made up by a large or a practically infinite vessel with thermocapillarity acting on top, only [33, 35]. The influence of the thermal boundary conditions on the stability of thermocapillary flow is reviewed only for small Pr in [41] and a short review on experiments on thermocapillary instabilities can be found in [42].

2 Systems with higher complexity
2.1. Adding buoyancy to thermocapillarity
We consider an increasing depth d of the liquid with extension L, with thermocapillarity driven in its horizontal free upper surface. Besides an increase of Ra as well A = L/d will approach a smaller value in most experiments. The experimental systems studied are either rectangular [22, 30, 37, 43-48] or circular, e.g. crucibles or annular gaps (to be discussed later).

In rectangular geometry we have an influence of the sidewall, modulating the travelling direction of the HTWs [22, 30] or by inducing 3-D flow [43-48]. It was found that an increasing influence of buoyancy – e.g. by increasing d – will stabilize the flow against transition to time dependence [30]. Smaller critical Marangoni numbers have been found in experiments with annular gaps under microgravity compared with those under normal gravity [16, 50]. The influence of gravity on Ma c for the occurrence of HTWs is shown in figure 17. Increasing Bo(h) in figure 17 means increase of d. Gravity stabilizes the basic flow with multirolls against the transition to HTWs.

![Figure 17](image)

The 3D-stationary instability of longitudinal rolls found at larger d [22, 23, 37, 47] was not found under microgravity, indicating that it needs gravity influence to develop [16].

At larger d, e.g. d > 10 mm, the convection roll driven by thermocapillarity and buoyancy can separate from buoyancy driven convection in the bulk fluid [43-46]. This type of flow seems to be steady in comparison to that for d ≤ 3 mm which is time-dependent at comparably smaller Ma. The separation has been successfully simulated numerically by use of a rather fine mesh and by introducing temperature-dependent viscosity [49]. Moreover, one has to resolve the buoyant flow in the thermal boundary layers at both end walls and their connection with the thermocapillary flow to get a correct numerical simulation of the flow structure. Hotter liquid is circulating on top of colder liquid in the bulk in this separation of surface flow and its return flow from the bulk [43, 46, 49]. This
separation of a thermocapillary convection roll (or layer) on top of the fluid is demonstrated in figure 18 by streaklines in a vertical light sheet and by holographic interferometry indicating the isotherms. This separation was as well observed in crucibles [50, 51] and its instability will be considered later when the separated "basic thermocapillary flow" is strongly cooled from above.

Figure 18. (a) Streaklines and (b) isotherms by holographic interferometry in a cubic fluid cavity with \( L = H = 20 \text{ mm} \) at \( \Delta T = 4 \text{ K} \) (Pr = 17). The cavity has a flat free surface without any menisci at its side walls. From [46]. Indicated is the separation of a thermocapillary convection roll with hot fluid on top of cold fluid in the bulk. The interferometric fringes indicate isotherms with a distance of 0.079 K.

Turning from rectangular cavities to rotationally symmetric annular gaps or cylindrical vessels (annular gaps with cooled or heated centres) does not change the nature of the HTWs or linear rolls. The advantage of this kind of geometry compared to the rectangular one are avoidance of one pair of sidewalls which could become disturbing due to extra lateral temperature gradients or by reflecting travelling HTWs. We mention work on annular gaps with not too large \( d \) [15, 16, 18] annular gaps with \( A \) up to \( A = 1 \) [33, 34, 16, 52] and crucible-like configurations [51]. For large \( A \) and small \( d \) HTWs are observed [15, 16] which form Archimedian spirals in this configuration because of a large and constant angle of between the wavevector and the temperature gradient (figure 6). The HTWs for the fluid with \( 7 \leq \text{Pr} \leq 17 \) have the travelling direction from "cold" to "hot." For smaller \( A \) some wavetrains are observed to travel mainly azimuthally [16, 33, 34, 52]. In a Czochralski crystal growth configuration HTWs have been observed to travel azimuthally in the meniscus at the cooled crystal dummy [19]. Numerical simulation of Czochralski-growth systems of and oxide melt [53, 54] revealed azimuthally travelling waves as those observed in the model systems of annular gaps. But these HTWs have not yet been reported from real growth systems.

2.2. Temperature gradients inclined to the free surface
The motivation to study this problem is application; we encounter at the same time heating from the side and cooling from the top in many practical situations (e.g. in material processing like crystal growth from the melt by the Czochralski-technique). The possibility of an instability of thermocapillary flow in form of the classical Bénard-Marangoni instability by cooling from above is given. Indeed, small convection cells can be observed on the surface of high temperature melts to drift from "hot" places to "colder" ones (figure 19). The small wavelength \( \lambda \) of this cellular instability at the
surface of oxide melts of considerable depth \( d \) (\( 2\lambda \ll d \)) indicates a cellular instability of a thin layer of the melt, only. We have seen in the preceding chapter that a time-independent surface roll can separate from the flow in the bulk (figure 20). We can take the flow profile of this separated surface tension convection as basic state for a new state when cooling from above. Two flow profiles can be studied as

- separated flow with return flow
- linear flow (only from hot to cold without return flow) because the return flow is deeper in the bulk

![Figure 19. Cellular convection pattern observed on Garnet melt surface (at ~ 1750°C in a 12 mm diameter crucible, melt depth \( d \gg 30 \) mm, drifting rapidly radially inwards to the coldest spot of the surface. Note the linear rolls (steady) near the heated wall also called "spokes", note cross cells between the spokes (drifting) and chaotic Bénard-like cells in the centre. From [55].](image)

**Figure 19.**

![Figure 20. Streaklines in a 40 mm \( \phi \) crucible, liquid depth \( d = 20 \) mm, different \( \Delta T \) between hot crucible and cold crystal dummy, showing the separation of the surface tension driven convection from bulk convection. From [51].](image)

**Figure 20.**

Given a melting temperature of the order of 1800°C and a semitransparent melt (or even a black melt), the surface will be cooled very effectively by radiation. This can induce Bénard-Marangoni cells which will increase the heat transfer from the bulk to the free surface and through the free surface. Thus the heat flux through the free surface is enhanced what will in turn increase the thermocapillary action and the tendency for the above discussed separation. One can assume a Nusselt number \( Nu \) for the Bénard-Marangoni instability well above \( Nu = 2 \) because the value \( Nu = 1.8 \) has been reported for \( Ma = 4 \cdot Ma^2 \) from an experiment under microgravity [12]. One can expect the correct simulation of buoyant-thermocapillary convection in the whole melt in the crucible only for the correct simulation of the heat transport through the free surface and through the crystal. This, in turn, is needed to simulate the correct balance between buoyant – thermocapillary convection and forced convection driven by crystal rotation which is decisive to simulate the crystal radius and the shape of...
The basic ideas are sketched in figure 21. The flow and temperature profile at the melt surface, that can become instable due to cooling from above, can be more a linear velocity profile (Figs. 21a, 4) or one with return flow (figure 21b). The type of drifting cells and their drift speed, that is the type of instability, will be different for the two velocity profiles.

Figure 21. The counterbalance of buoyant-thermocapillary and rotationally driven flow in a high-temperature melt (Pr >> 1). A thermocapillary-driven flow can separate from the bulk flow, balancing the rotational one. And this "linear" flow profile (a) can become instable (to Bénard-like convection cells) due to large radiative heat loss from the melt surface. This heat loss is largest near the centre of the crucible or near the crystal. The instability in form of rolls or cells can reach down to the return flow (b) if the return flow is very near the surface flow due to strong separation.

The stability of thermocapillary flow (with return flow) with inclined temperature gradient was investigated theoretically (figure 22) [59]. Transition to HTWs, to transverse travelling rolls (TTRs) or to square drifting cells and stationary longitudinal rolls (LRs) are observed, depending on Maₐ and Maₐ. HTWs are stabilized by cooling from above (line 3 in figure 22, increase of Maₐ/Maₐ) as well observed in [60, 61]. Stationary longitudinal rolls have been observed by many experimenters mentioned above at increased liquid d though the parameter Maₐ/Maₐ in figure 22 is meant for increasing ΔTv, only (dliquid/dgas was fixed in [59]). The stationary rolls are as well observed in the experiments and numerical analysis changing d and ΔTv/ΔTₐ [61]. Transverse travelling rolls (or square drifting cells) have been observed cooling a layer with thermocapillary flow (Pr = 10) from above in experiments with controlled ΔTₐ and ΔTv [62]. The scenario and tendencies of figure 22 seem to be correct and are found in Czochralski melt growth.

In the situation sketched in figure 21 (a) only the linear flow profile, like that of linear thermocapillary flow, becomes instable due to cooling from above. This has been investigated in the same apparatus used in [39] to study linear thermocapillary flow and its instabilities at larger Maₐ. Figure 23 shows the pattern of convection cells drifting in linear thermocapillary flow cooled from above. The drifting speed of the cells is proportional to that of the flow speed in the free surface. It is almost as high as the velocity of the free surface and it is much larger than the drifting speed of convection cells observed in thermocapillary flow with return flow (as sketched in figure 21 (b)) [63]. Figure 24 indicates the stability threshold in terms of Maₐ. The difference between the experiments with liquid layers on a metal plate with T-gradient [39, 63] and that of separated thermocapillary flow on the top of oxide melts in Czochralski growth are the undefined thermal- and flow boundary conditions at the lower border of the latter. But the physics of the instabilities should be the same.
2.3. Static and dynamic surface deformations

The heat transfer properties of a flat interface and that of a curved one can differ significantly. One example is the meniscus of a liquid at a heated wall when the liquid is fully wetting the wall in comparison to a liquid with flat surface pinned to e.g. the upper rim of the vessel. Thermocapillarity will be of another nature in the meniscus region (e.g. stronger in case of a thermal boundary layer at the wall). A flow instability occurring in the meniscus region can be well separated from the bulk flow.

**Figure 22.** Stability diagram of thermocapillary flow with inclined temperature gradient, expressed by the relation between the vertical Marangoni number $M_a$, and the horizontal one, $M_a$. The line "H-F" marks the transition to oblique hydrothermal waves, line "F-E" that to transverse travelling rolls and lines 1 and 2 that to stationary longitudinal rolls. After [59].

**Figure 23.** Linear thermocapillary flow without return flow cooled from above (sapphire lid with temperature gradient parallel to that in the metal bottom) shows convection cells drifting with the linear flow for $M_a$ above the threshold. Visualization with IR camera, IR illumination and aluminum flakes. ($d = 1.34$ mm, $M_a = 12.6$, $M_a = 132$, $Pr = 10$) [63]. $M_a$ is too small in the right part of the figure (hot) for the cellular instability and linear rolls can be observed there.

**Figure 24.** Stability limits of linear thermocapillary flow cooled from above [63]. For a certain $M_a$ with linear flow we find with increasing $M_a$,
- ○ stable linear flow
- ● longitudinal rolls
- ◊ transition from rolls to drifting cells (guessed)
- ♦ drifting cells
because of its geometric confinement and different thermal boundary conditions. Spontaneous convection can occur in the vicinity of a liquid meniscus in case of solute transfer across the curved interface [64]. Heat transfer across the meniscus interface will change thermocapillary flow, too. Thermocapillary flow is largest near the hot crucible wall in Czochralski systems of oxide materials because of the thermal boundary layer and, in accordance with figure 22, stationary linear rolls (called "spokes pattern" in the crystal growth literature) are observed there. In the regions with smaller horizontal temperature gradient, or stronger cooling from above, the pattern of linear rolls transforms into transverse travelling rolls or travelling cells [65].

Liquid bridges LBs are hydrostatically deformed by gravity in laboratory experiments and their volume V can be less than ideal, e.g. $V/V_0 < 1$ (underfilled or necked-in LB) or more than ideal ($V/V_0 > 1$) (bulging LB). The stability of thermocapillary flow can depend strongly on $V/V_0$ with maximum stability near $V/V_0 = 1$ [66-68] in terms of a Marangoni number $\text{Ma}^* = \frac{|\partial \sigma / \partial T| \cdot \Delta T \cdot \eta^{-1} \cdot \chi^{-1}}{R}$, with the temperature difference $\Delta T$ between the support rods and radius R of the LB. We note that most of the $\Delta T$ applied to the support rods is dropping in the thermal boundary layers on both end walls in the LBs with $Pr >> 1$ investigated until now. Thus the meniscus-shape in a necked-in LB will provide a relatively better "thermal coupling" of inner parts of the necked-in LB to the end walls than that provided for an ideal LB. The smaller $\text{Ma}^*$ for necked-in LBs is understandable, therefore. Heat transfer to the surrounding air can play a role for the critical $\text{Ma}$ as well because buoyancy-driven convective rolls, together with thermocapillary-driven rolls, are alive in the gas-surrounding of the LBs in normal experiments.

Known since Bénards experiments and measurements [9, 1] is the depression of the free surface at the spots of the hexagonal convection pattern where the hot fluid is rising. This hotter fluid has smaller surface tension which generates the depression. Thus it is principally impossible to investigate thermocapillary flow or Marangoni effects in liquids with a mirror-like flat surface. It is possible to prepare such a mirror-like flat free surface without meniscus at the wall of the vessel if no temperature gradients are applied. But a surface depression will be observed as soon as a wall is heated. The surface depression in its vicinity occurs because of uprising hot flow [20]. Time-independent flow and temperature gradients will deform the free surface statically and time-dependent states will generate time-dependent surface deformations. The dynamic surface deformations can be essential for the instability mechanism of thermocapillary flow or they can be only reactions to the flow instability.

**Figure 25.** The amplitude of standing surface gravity waves in a cavity with $L = 20$ mm, $d = 20$ mm, $W = 41$ mm, $\Delta T = 27.5$ K when the underfilling $h$ from a flat free surface is increased up to $h = 1.4$ mm. Onset at $h = 0.8$ mm. From [29].

**Figure 26.** Time-dependent flow in the meniscus at the cold wall ($L = 12$ mm, $W = 41$ mm, $\Delta T = 45.5$ K, $h = 1.8$ mm). Streak-photograph of a light cut from the side. The oscillatory vortex fills only the upper part of the meniscus and secondary vortices are visible below it. From [29].
The HTW in dynamic thermocapillary layers [13] have been derived in a stability analysis of layers with undeformable free surface and their properties compare well with HTWs observed in experiments [14, 15]. We conclude that the surface deformations experimentally observed in connection with the occurrence of HTWs are only reactions of the free surface to time-dependent flow. The same is true for the degenerated HTWs in liquid bridges; here experiments and numerical simulations of LBs with undeformable free surface show satisfactory agreement. As well the HTWs observed at the heated cylindrical inner wall of an annular gap [20] and those in the meniscus of a cooled dummy crystal in a Czochralski configuration [19] are small and of secondary importance for the occurrence of this instability.

An interesting case of surface oscillations was observed in the meniscus at a cold wall when – and only when – the height of this meniscus was in a certain range and was pinned to the upper rim of this wall [29]. The experiments have been performed in a rectangular cavity with \(A = 1\) (e.g. \(L = 20\) mm, \(d = 20\) mm, \(w = 41\) mm) filled with ethanol (\(Pr = 17\)) with not too much underfilling below the normal volume \(V_0 = 20 \times 20 \times 41\) mm\(^3\), such that the menisci at all walls were still pinned to the sharp upper rim. Macroscopic standing gravity-surface waves with amplitudes up to 0.25 mm have been excited in the surface of the liquid due to resonance with oscillations in the meniscus at the cold wall. The macroscopic gravity surface wave disappeared when the meniscus at the cold wall was no longer pinned but able to slide at the cold wall. It was concluded that the flow in the meniscus at the cold wall became instable to oscillations of the type of surface waves [27]. The frequency spectrum of these surface waves was tracked as spectrum of temperature oscillations in the meniscus and some peaks of this spectrum are found to be near the frequency peaks of gravity surface waves in this cavity. Tuning the frequency peaks of the gravity surface waves by changing e.g. \(L\), the resonance could be reinforced or suppressed (e.g. with \(L = 12\) mm instead of \(L = 20\) mm). Figure 25 shows the amplitude of the standing wave in direction of the applied temperature gradient \(\Delta T/L\) (special mode) as function of the underfilling \(h\) (measured from the top rim of the cavity) for a fixed \(\Delta T = 27.5\) K. The onset of the surface gravity waves is for \(h = 0.8\) mm, and for \(\Delta T^c \sim 11.5\) K at \(h \sim 1.5\) mm with frequencies of \(\sim 7\) Hz (depending on mode and \(h\)). It was concluded that time-dependent flow in the meniscus is exiting the standing gravity waves (figure 26). The resonance is possible if the frequencies of the exiting and the exited oscillations are not too far apart and if at least one has a large enough half width in frequency (figure 27). We have an example of the excitation of macroscopic standing gravity waves by small amplitude surface oscillations by oscillatory thermocapillary flow in case of resonance.

3 Systems of technical relevance
3.1. Solutocapillarity and dirt films
We will give some examples of thermocapillary flow or solutocapillary flow in systems of technical relevance. We leave the model substance "silicone oil" and turn to metals or crystal growth melts.
The surface tension and its temperature dependence are normally changed by impurities. Solutocapillary effects are often much stronger than thermocapillarity. Moreover, many liquids contain impurities which are surface active; e.g. these impurities can accumulate in the free surface as a film. Such a film can suppress thermocapillarity totally [44]. Water and mercury are examples of such liquids. Thermocapillarity and its instabilities can be studied reliably only in liquid systems without such surface films. Either high purity or very low surface tension of the liquid are used to reach this goal. Only a few liquids with higher surface tension seem to make no problems (e.g. NaNO₃-melt). If the surface is only partly covered by an impurity film, this film will be concentrated at the cold side and will be deformed by surface pressure of the flow [35] or the flow direction is changed [69]. A flow-reversal is known from arc welding where sulphur-impurities are known to produce this and the depth of the weld-bed can increase significantly by this flow reversal (from "cold" to "hot" in the free surface).

The surface tension of oxides is normally significantly smaller than that of the pure metal and solutocapillary effects can override thermocapillary ones. One example is given by a liquid Sn-droplet covered with a SnO₂-skin in high vacuum, bombarded with an ion beam on one spot [70]. It was sufficient to bombard only this spot to remove the whole SnO₂-skin because solutocapillarity is driving any SnO₂-skin towards the (heated) bombarded spot with its higher surface tension where the SnO₂ is sputtered away. Strong solutocapillary flow and flow instabilities including surface vibration have been observed in melt pools of Cu or of Sn blowing locally oxygen onto the melt surface [71].

The experiments to study the onset of oscillatory thermocapillary flow in LBs from mercury suffered from the presence of amalgame films [72] whereas careful experiments on LBs from tin in high vacuum and with a getter (Ti at 600° C) to reduce the oxygen partial pressure, yielded reliable results of Ma˚ and the oscillation frequency [73]. The same is true for the studies of the instability of thermocapillary flow in LBs from silicon melt [74-75] or that in melt pools like in Czochralski crystal growth [76]. Many attempts to measure Ma˚ in silicon melt have been undertaken because of the technological importance of this material. It is not easy to compare the measurements and to predict the flow-state in industrial systems because of the high solubility of oxygen in Si-melts and the volatility of SiO at the melting temperature of Si; the surface tension of Si-melt shows a strong dependence on the oxygen partial pressure [77, 78] and the systems are not easily characterized.

Figure 28. The measured critical Marangoni numbers in liquid bridges and layers (symbols) are generally larger than the theoretical ones ( lines from[13]) but with the correct tendency. The reason for the discrepancy might be uncontrolled surface tension properties for the metallic systems (oxide impurities) and for the high-Pr liquids the “consumption” of the largest part of the applied temperature gradient in the thermal boundary layers. The critical values measured at liquid layers with temperature gradient in the metal bottom is nearest to theory, therefore.

We have the impression that experiments on "easy" model systems with liquids of high Pr are rather reliable because they can be characterized well enough and the physical parameters are known. Fundamental aspects of thermocapillary instabilities can be studied especially if the liquids are transparent. The critical Marangoni numbers measured at LBs from high Pr-liquids are rather reliable
and the extrapolation of the data to smaller Pr was successful. Figure 28 shows the measured critical Marangoni numbers in LBs with different Pr in comparison to theory for infinitely extended liquid layers. Only the tendency over Pr is satisfactory.

3.2. Thermocapillarity and solutocapillarity in interaction with moving crystal boundaries

Crystal growth is a nonequilibrium process. In melt growth we have a moving temperature gradient in front of the crystal-melt interface as the driving potential for the advancement of the solid interface. Thus we expect thermocapillary flow towards the crystal grown from the melt with free surface (floating zone technique, Czochralski technique). This flow transports heat towards the crystal. On the other hand, the melt is never pure or it is even doped deliberately and, impurities or dopants are not incorporated into the crystal as easily as the main constituent. The dopants or impurities are normally enriched in the residual melt and the crystal is more pure than the melt. Defining the concentration of an impurity in the melt as $c_m$ and that in the solid crystallized with growth speed $v$ as $c_s(v)$ we can define the segregation coefficient or distribution coefficient $k(v) = c_s(v)/c_m$. The effective distribution coefficient $k(v)$ is smaller unity if the crystal contains less impurity and this impurity is enriched in front of the crystal growing with speed $v$. We note that the impurity source is ceasing for zero growth speed. The impurity source provided by the growing crystal ceases as well for $v \to \infty$ because then $k(v) \to k(\infty) = 1$. The strength of the impurity source provided by a growing crystal is limited; therefore, we have thus the possibility to use the growing crystal as a source of rejected impurities and, if the impurity is e.g. lowering the surface tension, solutocapillary forces can drive flow in the free surface away from the growing crystal.

Different cases can arise

a) time-dependent thermocapillarity (or any other type of time dependent flow towards the crystal interface) modulates the growth speed of the crystal and thus the amount of incorporated impurity.

b) solutocapillarity and thermocapillarity are directed parallel and reinforce each other

c) solutocapillarity is antiparallel to thermocapillarity and is weakening it

d) solutocapillarity is strong and counteracting thermocapillarity. New instabilities are possible.

e) the solute is surface-active, forming a film at the free surface, but partly soluble with solutocapillarity counter thermocapillarity. New instabilities are possible.

f) thermocapillary flow and forced flow by crystal rotation are counteracting and both are shaping the crystal interface, but differently, where the rotational action depends on the crystal shape. Instable situations for the flow and the interface are possible.

Some of these cases have been treated or observed.

a) A floating zone of Si-melt doped with Sb was crystallized under microgravity from one side with 1 mm/min and the temperature oscillations by time-dependent thermocapillary flow have been measured together with growth rate oscillations [80]. From the good correspondence of both signals it was concluded that the growth speed of the crystal was reacting on the flow - and temperature - oscillations of thermocapillary flow. A quite similar result was obtained by other authors in laboratory experiments [78].

c)-d) A thin sheet Bi-crystal was grown by directional solidification at high growth velocities $v$ from a Sn-0.11 at % Bi melt-layer of 2 mm thickness with free surface under a temperature gradient of approximately 10 K/cm. The authors could observe a coordinated back and forth motion of the meniscus line at the growth front [81]. For the doping of Sn with Bi a flow reversal was observed for some concentrations c (e.g. $c < 0.5$ at % Bi) and solutocapillary flow can oppose thermocapillary one. This work seems to be an example of the active involvement of the growing crystal in a thermocapillary-solutocapillary instability.

d)-e) This active participation of the growing crystal in the instability has been observed in LBs from NaNO$_3$ which have been faintly doped with the surface-active impurity CH$_3$CH$_2$COOK and recrystallized from below [82]. The surface tension of NaNO$_3$ is lowered dramatically by the impurity and the impurity is rejected by the crystal. We thus have solutocapillarity opposing thermocapillarity. In contrast to the well-known passive growth oscillations discussed above
under a) the growth and remelting of the crystal is actively taking part in this instability. The full mechanism of this new type of instability is outlined in this paper [82]. We outline one part of the instability-cycle: “The crystal is a source of impurity, increasing solutocapillarity during growth. This decreases the convective heat transport to the crystal because thermocapillarity is weakened. The convective heat transport to the crystal is weakened. Thus the growth speed is increasing more and more. This is limited by the applied temperature gradient and by $k \rightarrow 1$.” The reverse cycle happens once remelting starts because pure substance is remelted, weakening solutocapillarity (strengthening thermocapillarity). The impurity CH$_3$CH$_2$COOK might form a film on NaNO$_3$-melt with the consequence of a 3D-structure of thermocapillary flow.

Solutocapillarity as analogue to thermocapillarity in a LB was studied numerically [83]. The source for the solute gradient was the growing crystal and the solute distribution differs from the temperature distribution for thermocapillarity, therefore. The authors found for certain conditions a Hopf bifurcation from the basic flow to HTWs.

**Figure 29.** The counterbalance between buoyant-thermocapillary and rotationally driven flow of a flat interface ($K/r = 0$) and a conically deflected one ($K/r = 1$) (crystal radius $r$, stagnation point at $r_s$, rotational Reynolds number $Re = 2 \frac{\pi r^2}{\nu}$).

a) Experimental result of the stagnation position for two different $K/r$ at constant buoyant-thermocapillary flow; the flat interface needs smaller $Re$ to shift the stagnation point to $r_s/r = 0.9$.
b) Numerical result on flow and stagnation positions (SP) for two interface shapes (flat on the left side, conical on the right side) for else unchanged boundary conditions for the flow.

Though the boundary conditions of the experiment are used for the simulation, the simulation does not show the separation of the thermocapillary flow observed in the experiments. The separation of the thermocapillary flow could increase the observed difference in the action of a flat and a conical interface. From [58].

An interaction between buoyant-thermocapillary flow and forced flow by crystal rotation with dramatic consequences was observed in Czochralski growth of high melting point oxides [58]. The crystal has a strong conical interface deflection during the beginning of growth because of large radiative heat transport through it. The rotationally driven convection roll is less intensive near the free melt surface for a conical interface than for a flat interface (figure 29). The rotation of the crystal is chosen by the crystal grower in a way that the balancing point (stagnation line) between forced convection and buoyant-thermocapillary convection is near the crystal periphery to control the crystal radius. This situation is instable during the increase of the crystal radius (figure 29) because of the following reaction chain: “a small increase of the crystal radius $r$ at a certain critical crystal radius will increase the intensity of the rotationally driven convection. This in turn will melt-back a part of the
crystal cone below the surface and this increases the strength of rotational convection near the interface. The stagnation point is shifted outwards because of this and the convective heat transport towards the crystal by thermocapillarity is weakened, resulting in further increase of the crystal radius. Thus the crystal radius will increase uncontrollable and the flow near the crystal interface changes from "buoyant-thermocapillary dominated" to "rotationally dominated." “The reason for the significance of thermocapillary forces in this flow transition is the fact that both, thermocapillarity and forced convection by a flat rotating crystal interface are acting and counteracting near the free melt surface. Radial outwards convection, driven by a rotating conical interface below the surface, has another velocity profile than that driven by a rotating flat interface near the free melt surface.

4 Conclusions
Thermocapillary convection with its instabilities and effects in melt processing (mainly crystal growth from the melt) has been reviewed from an experimental point of view. We propose to interpret the thermocapillary instabilities observed in the more complex systems as derivable from the instabilities found in the model systems, like hydrothermal waves or linear rolls or Bénard cells in infinitely extended layers. We found tendencies supporting this view although the conclusions are not unambiguous.

The presented material covers mainly liquids with Pr > 1. Metallic liquids with Pr << 1 are only touched and their larger difficulties with defined surface properties (e.g. oxide films) are pointed out.

The literature cited in the article can by no means be complete but can serve as a source to cover the complete field.

Crystal growth from the melt is an prospective area to apply the knowledge about thermocapillary (Marangoni) instabilities because of two reasons:

1. The melt adjacent to the growing crystal has a free surface in the Czochralski technique, in the floating zone technique and in directional solidification in horizontal geometry. The free surface is mandatory to avoid heterogeneous nucleation and is thus mandatory for the growth of perfect single crystals. All ingredients for thermocapillary flow and Marangoni instabilities are given.
2. Large temperature gradients near the growing crystal interface are mandatory to avoid constitutional supercooling with its catastrophic effect for crystal quality.

We thus have liquid systems in melt growth with all ingredients for strong thermocapillary flow and its instabilities. Moreover, the growing (moving) crystal interface can react to flow oscillations (heat transport fluctuations) and can incorporate them as dopant striations. Solutocapillary effects can be introduced by the segregation connected with crystallization. The crystal can even play an active rôle in coupled thermocapillary – solutocapillary instabilities.

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