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Fluid Physics under Microgravity: Status Report after the German Spacelab D1-Mission

This paper reviews the research areas of fluid physics under microgravity conditions. The relevant experiments performed within the German Spacelab Mission D1 are used as the guideline. Firstly, there is the field of fluid handling and sloshing, whose importance is largely increased in a microgravity environment. Strongly linked with this field is the field of stability and oscillations of fluid interfaces. The terrestrial experience that a fluid is located on the bottom of a container blocks our imagination on what is going on under reduced gravity. The oscillations of fluid volumes often are the clue for judging the tolerability of g-gitters during experiments in the fields of material sciences and live sciences. Research into Marangoni convection, which is generally masked by buoyancy on ground, has taken strong advantage of microgravity conditions. Sounding rocket and Spacelab experiments with liquid columns, on the transition from steady to oscillatory and turbulent Marangoni flows, on the migration of drops and bubbles, and on solutal Marangoni convection are reported. Eventually, transparent model experiments with organic liquids on the separation of monotectic metallic alloys, on convective phenomena arising at solidifications fronts, and microgravity experiments on the propagation of chemical waves are reviewed.

1 Introduction

This report deals with the task of fluid physics under microgravity. The aims and specific results of the D1-experiments serve as a guideline. They are commented in context with their preparation and testing in parabolic flights, TEXUS experiments, and GAS missions. For all D1-experiments performed in the Fluid Physics Module (FPM) it is true, that the wetting conditions between the fluids and the solids used or else some technical details have been tested in two campaigns of KC-135 flights in Houston/Texas.

The tasks of fluid physics under microgravity are

- to study the behaviour and handling of fluids
- to promote basic research on fluids
- to test computer programs on shape, stability, oscillations
- to learn about fresh interfaces
- to learn about thin liquid layers on solids
- to study edge effects
- to investigate wetting and spreading effects
- to study coalescence
- to observe thermal and solutal Marangoni convection
- to look into oscillatory and turbulent modes
- to study heat and mass transfer
- to perform transparent model experiments
- to assist crystal growth experiments
- to assist production of fine-disperse emulsions of metals
- to study microconvection during cellular growth, dendritic growth, etc.
- to assist experiments in the feld of life sciences
- to analyse g-level tolerability

2 Fluid Handling

The first area of research is fluid sloshing, the behaviour and the handling of fluids in tanks.

This was the aim of *Vreeburg's* experiment WL-FPM-08. In order to assure stability and liquid outflow from a spacecraft tank on demand, location and momentum of the liquid must

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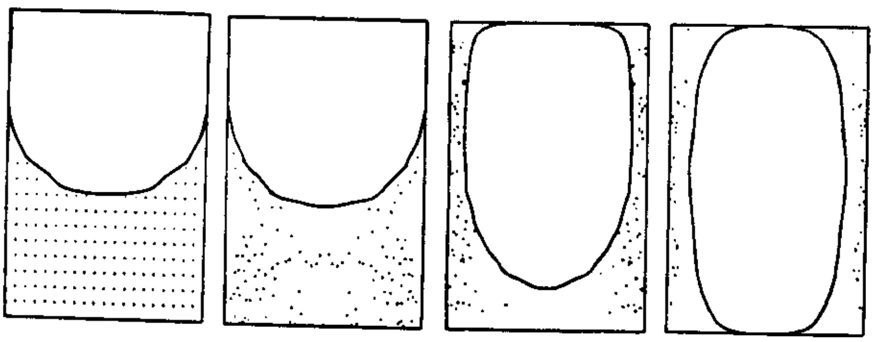
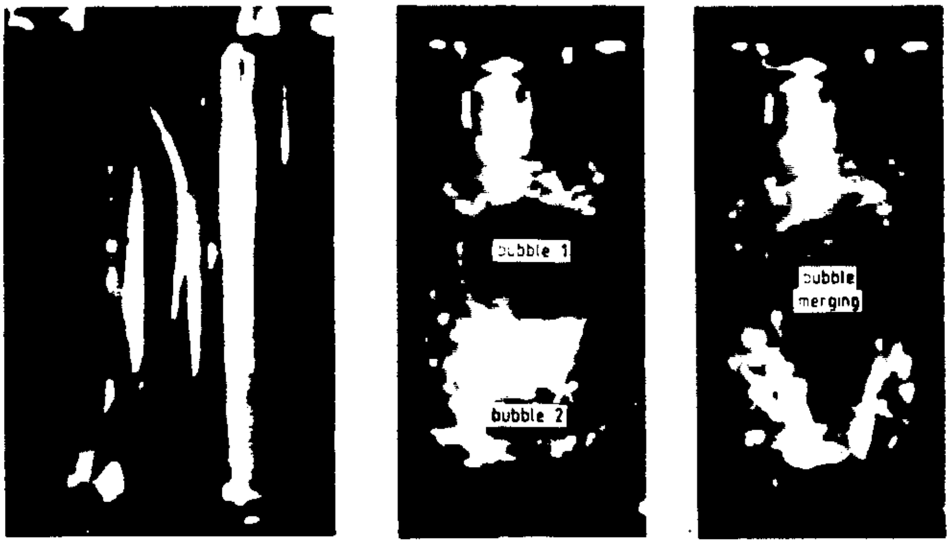


Fig. 1. The drainage of a liquid to the periphery, when a cylindrical tank containing a large bubble is spun up (from [1])



a b c

Fig. 2. a) The shape of a bubble in a rotating thin annulus. b), c) Merging of two bubbles in a cylindrical container

be well known. Fig. 1 shows the calculated behaviour of a large bubble, when a cylindrical tank is spun up [1]. Due to rotation the less dense medium, i.e. the bubble moves to the centre and is expected to take an oval shape.

In Spacelab-1 five different containers have been used. In Spacelab-D1 three liquid containers of different shape were investigated. A cylindrical annulus containing a bubble has been rotated with up to 100 rpm. Although the liquid (silicon oil) was free to move, the surface assumed an asymmetric shape [2], Fig. 2a.

Fig. 2b shows two bubbles in a cylindrical cavity with hemispherical ends. About 1 s later, in Fig. 2c, the bubbles merge, contact the container and are drained under one meniscus. The meniscus is stuck to the wall, since the contact angle between the materials used (water + perspex) is different from zero.

It is not only the shape of the static interface, which must be known, but also its oscillations. If there is resonance with some external structure, large liquid amplitudes will be excited. This makes difficult handling of fluids and perturbs intended experiments on fluid interfaces and all other experiments requiring low gravity levels.

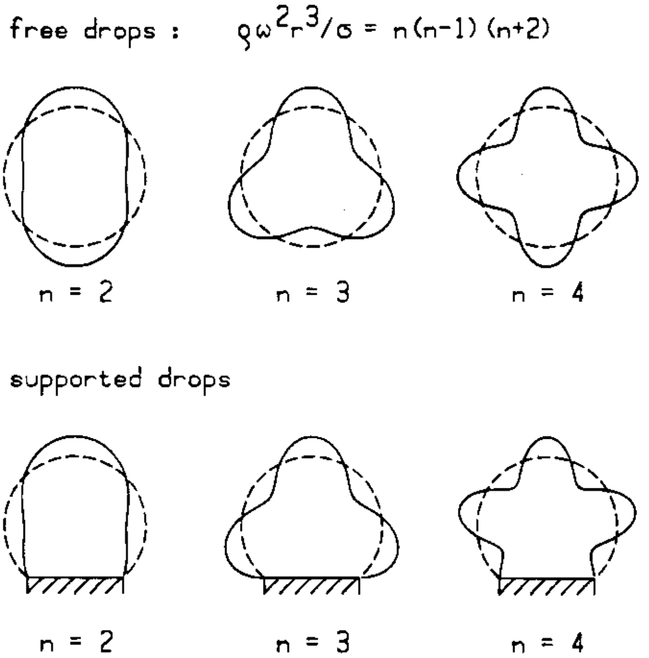


Fig. 3. Resonant oscillation of free and supported liquid drops exhibiting n = 2 to 4 nodes along the surface (inclusive of the node along the edge of the supporting disk)

3 Stability and Oscillations

The next topic to be treated are oscillations of liquid volumes. In the US program the Drop = Dynamics Module (DDM) has been developed for observing the oscillations of acoustically levitated drops. It has been flown by the Principle Investigator *T. Wang* in Spacelab-3. In Spacelab-1 investigations on the oscillations of supported drops were performed by *Rodot* and *Bisch* [3], Fig. 3.

In the European program attention has focussed on liquid columns. They are considered for assisting crystal growth experiments and for reasons of basic research as well. Stability limits and diagrams have been calculated in dependence on volume, height, surface tension, rotation frequency, and gravity (Bond number) [4, 5, 6], see Fig. 4.

Stability diagrams can tell, whether a liquid column between two coaxial disks is stable or breaks into a hanging and a sessile drop, whether an axisymmetric meniscus or that shown in Fig. 2a arises in a cylindrical container, and whether the bottom of a rectangular container is covered with liquid or else a dry spot arises, see Fig. 5.

The properties of long liquid columns were studied by *Da Riva* and *Martinez* already in Spacelab-1 (1ES 331). The columns broke earlier than expected from the reference experiments based on density-matched liquids (Plateau simulation). Therefore a TEXUS-12 experiment was performed on the maximum allowable injection rate [7]. For rapid filling the momentum of the liquid became so high that the cylindrical column broke during being built up (when the stretching rate was 6 mm/s for a disk radius of 30 mm and a final height of 80 mm).

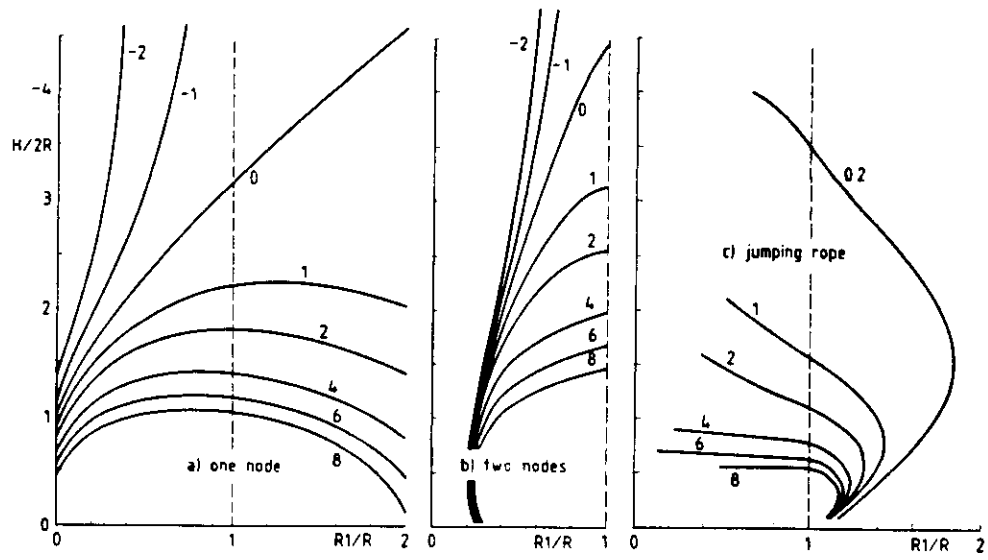


Fig. 4. The stability limits of rotating liquid columns at zero Bond number. The parameter shown is $W = \Delta\rho\omega^2 R^3/\sigma$
a) The first antimetric instability (amphora-mode)
b) The first symmetric instability
c) The jumping-rope instability (c-mode)

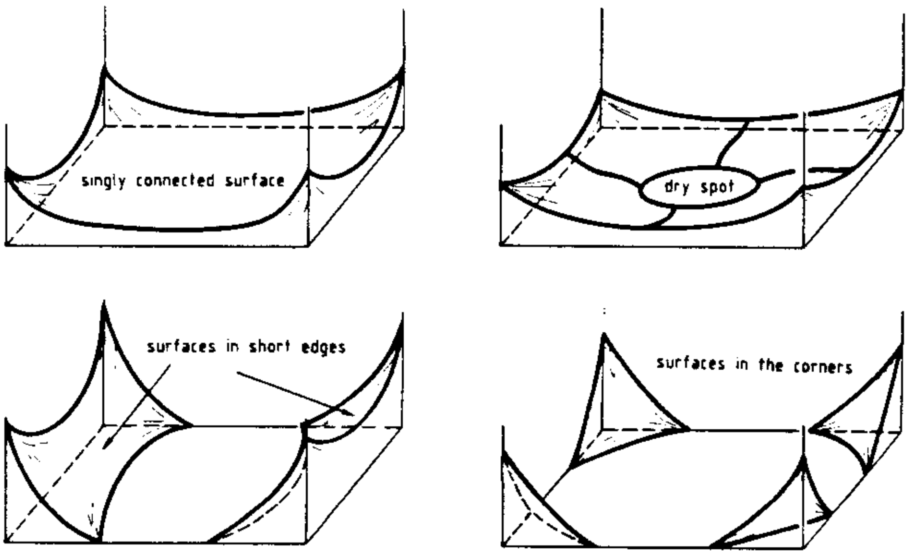


Fig. 5. Liquid surfaces in a rectangular container. With decreasing liquid volume first a dry spot arises. The dry spot then extends over the long edges and eventually the short edges

In the D1-experiment WL-FPM-04 the oscillations and the breakage of long liquid columns and the appearence of the jumping rope mode due to rotation have been studied (Fig. 6). Three rotating columns with shapes close to the transition from the amphora-like instability to the jumping-rope instability all broke according to the former mode. This is ascribed to residual accelerations.

Figs. 7 and 8 show the result of ground based experiments on the stability and oscillations of small liquid columns using a large magnification. (*Heide* and *Maul*). Liquid columns have been produced between disks with radii $R = 1$ mm and $R = 2.5$ mm. They have been lengthened at constant volume until breakage. The height H at breakage fits well with the theoretical value for the Bond number $B = g\rho R^2/\sigma = 1.46$. This value of the Bond number has also been found by adapting the shape of the columns by means of a computer program solving the Gauß-Laplace equation. Fig. 7 shows the first four resonance modes of such columns, which correspond to n = 1, 2, 3, 4 nodes of the respective vibration. The resonance frequencies ω_n shown in Fig. 8 have been determined by column lengthening at constant frequency and liquid volume, too. They appear to satisfy the relation $\rho\omega_n^2 R^3/\sigma = c_n(H_n/H)^2((H_n/H)^2 - 1)$, where H_n is the maximum stable height of the column with respect to the surface deformation exhibiting n nodes.

The stability limits considered are stationary limits, i.e., they do not depend on the viscosity of the liquid used.

4 Solid/Liquid-Attraction

The D1-experiment WL-FPM-06 by *Padday* explored the properties of liquid bridges between disks of unequal radius and used them to examine the forces acting in a wetting layer formed at the perimeter of the meniscus at the larger (lower) disk [8]. In Run A five different catenoids (A_1 to A_5 in Fig. 9) were formed without adjacent wetting layers. Each zone behaved as a perfect Laplace shape. In Run B two different zones (B_1 and B_2) were formed, but this time with extensive wetting films on the large disk. In these experiments only the liquid volume forming the bridge did possess a Laplace shape.

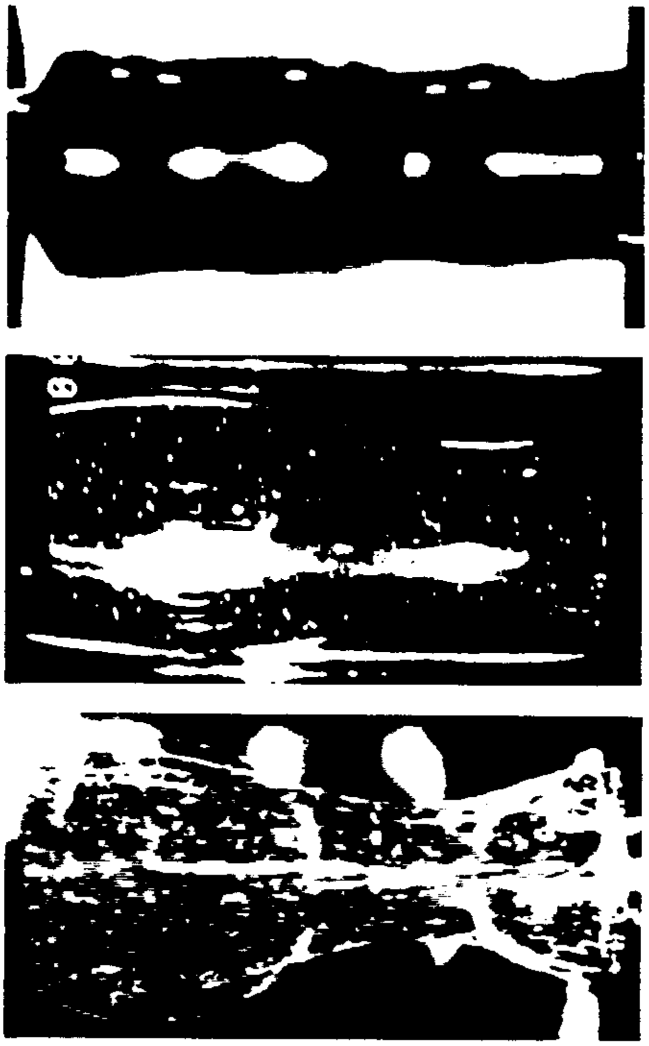


Fig. 6. Oscillations and breakage of a column of silicon oil with 35 mm in diameter and roughly 100 mm in height (from WL-FPM-04)

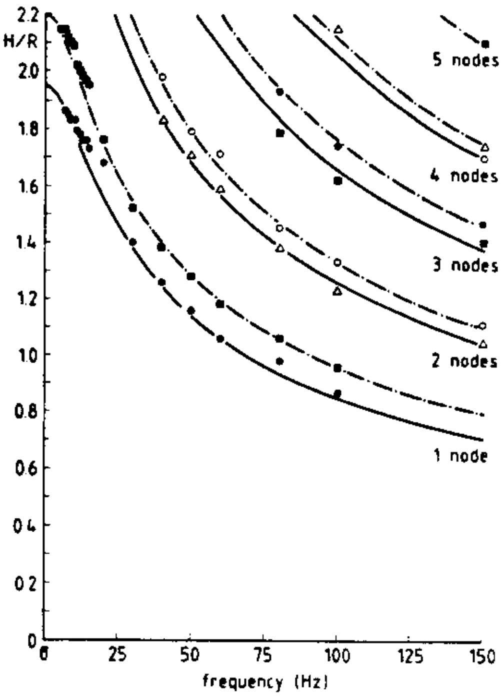


Fig. 8. The resonance frequencies of liquid columns with constant volume and increasing height. The full and the dash-dot lines correspond to the liquid volumes V1 = 45 mm³ and V2 = 63 mm³, respectively

The wetting film was not flat but was found to be much thicker than anticipated. The increased volume of the film entailed an earlier breakage of the bridge. In Run C a variety of non-catenoid zones (C_1 to C_4 in Fig. 9) were stimulated, first by rotation and then by vibration and lengthening together. Rotation at 5 rpm caused nearly no change in shape, but vibration increased the stability to lengthening well beyond the theoretical limit.

During several breakages the formation of a satellite drop has been observed (Fig. 10). The satellite drop moved forth and back along the symmetry axis and was repelled by both

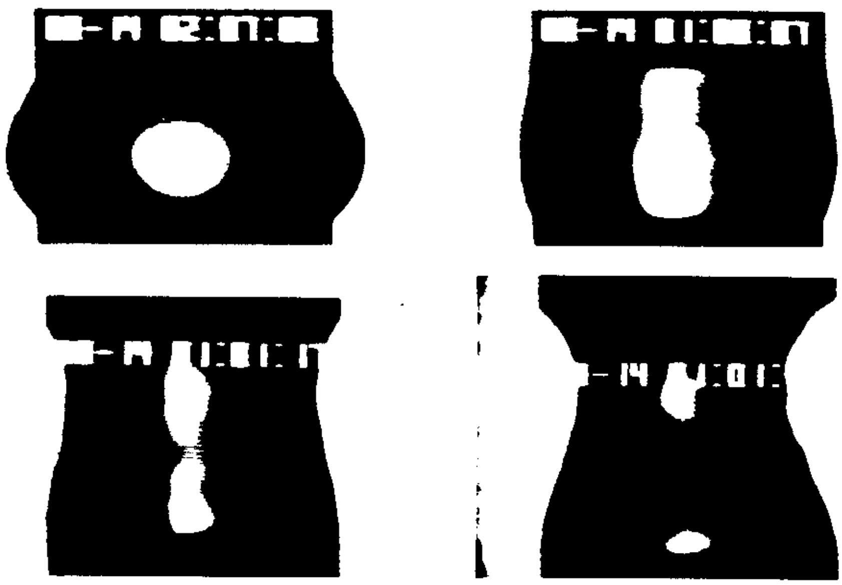


Fig. 7. Resonance modes of water columns between disks with 5 mm in diameter

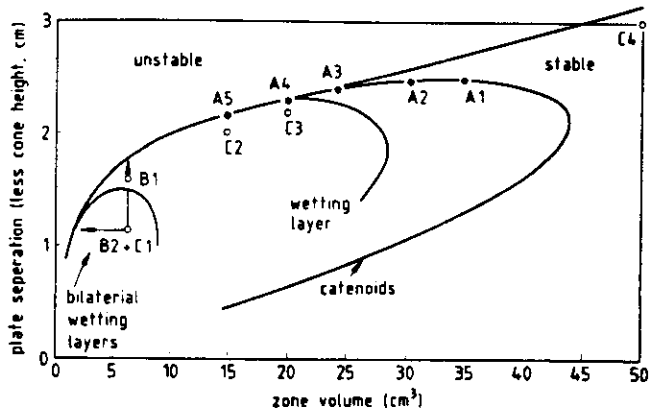


Fig. 9. Stability diagram for zero-g liquid zones between coaxial disks with radii 1.5 cm and 0.5 cm and experimental points in WL-FPM-06 (Courtesy of J. Padday)

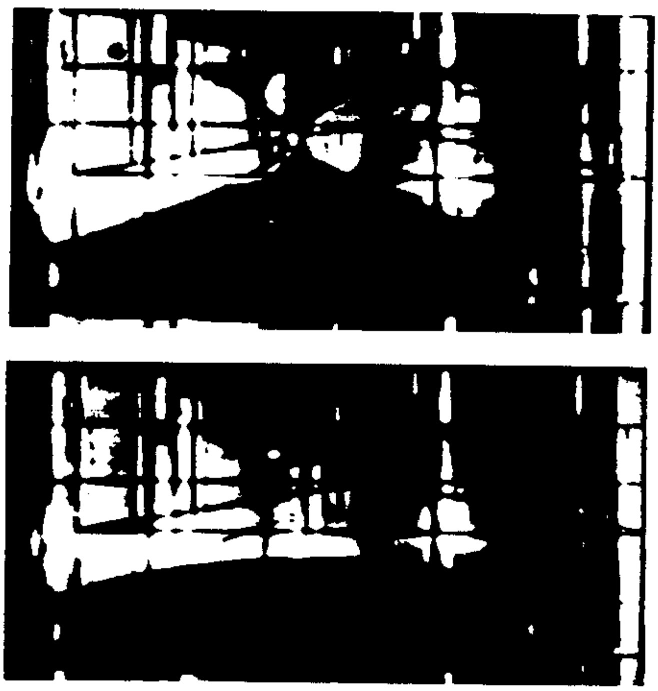


Fig. 10 A nearly-catenoid shape before breakage and a satellite drop after breakage. The latter movel forth and back along the symmetry axis without picking up speed in radial direction

the liquid volumes on the supporting disks. Whereas axial repulsion may be caused by opposite electrical charging, it is difficult to find an explanation for the extremely slow radial drift.

5 Marangoni Convection

Let us now turn to Marangoni convection. It is known since the last century, and is attributed to *Lord Rayleigh* and *Marangoni* as well. If along a fluid interface there are regions with higher and lower interface tension, a shear force from the latter to the former regions results, which gives rise to a corresponding fluid flow. Marangoni convection is the cause of dirty particles to move rapidly from the centre to the edge in a burning candle and the reason for cognac to creep up a nicely shaped cognac glass.

There is thermal and solutal Marangoni convection, i. e. the driving difference in interface tension may be caused by temperature gradients or by concentration gradients along the interface. One should not forget, however, that there are further causes for differences in interface tension like electric fields (electrocapillarity). One may, for instance, put a ring electrode around a liquid column and use the supporting disks as counter electrodes. With this arrangement two symmetric convection rolls may be expected.

6 Liquid Zones

Three lines of research on Marangoni convection under microgravity conditions have been followed. Firstly, there is a group of investigators, represented by *Chun, Monti, Napolitano, Schwabe*, whose effort is directed towards Marangoni convection in liquid zones. Such zones are used for crystal growth from molten zones or solvent zones. They are also favourable objects for basic research, since the axial symmetry is well suited to theoretical modelling. Under microgravity even perfectly cylindrical zones between two supporting coaxial disks can be established.

The disks are heated to different temperatures and the resulting convection is observed by means of tracers or else by analysis of the temperature field. For low temperature differences between the supporting disks usually an axisymmetric flow is found. With increasing temperature difference oscillatory modes arise, which are non-axisymmetric and appear to rotate around the axis (Fig. 11).

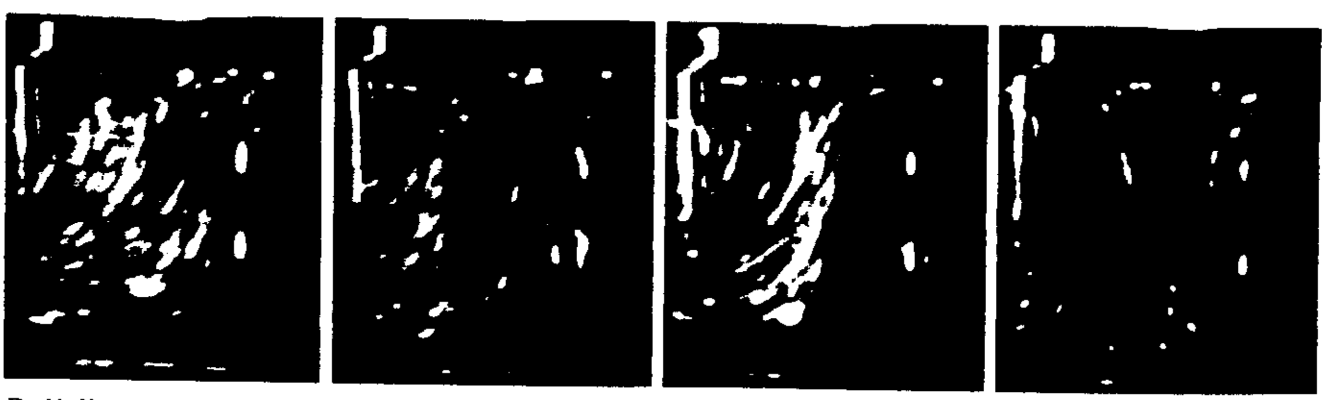


Fig. 11. Non-axisymmetric, oscillatory Marangoni convection during Run A of WL-FPM-07

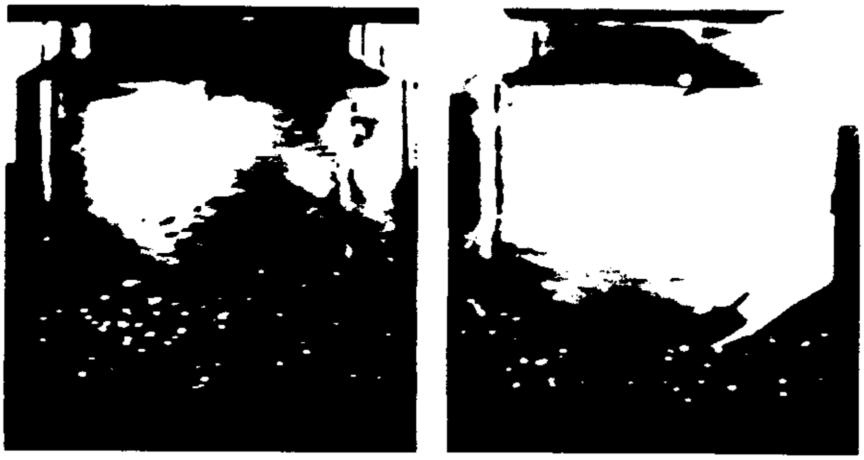


Fig. 12. The distortion of the interface between silicon oil and d-octylphthalate during Marangoni convection in Run B of WL-FPM-07

Within the TEXUS-3b mission *Chun* et al. have investigated Marangoni convection at a zone of silicon oil, whereas *Schwabe* et. al. studied Marangoni convection at a zone of sodium nitrate. *Chun* et. al. used meridian plane illumination and camera observation. The tracer particles, due to being heavier than the silicon oil, clearly moved out of the vortex centres of the flow [10]. *Schwabe* et. al. by inserting thermocouples into the zone were able to measure the transition to oscillatory flow conditions. The periods of the oscillations were nearly equal to those observed in the reference experiments on ground [11]. The investigations on silicon oil have been repeated and extended in the mission TEXUS-7, those on sodium nitrate in the missions TEXUS-5 and TEXUS-8 and eventually in the MAUS mission DG-306 [12, 13].

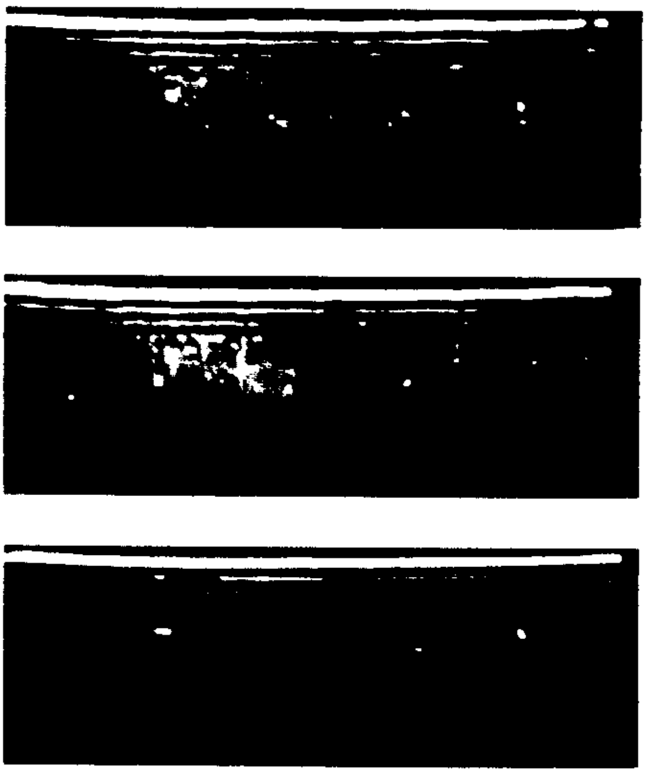


Fig. 13. Two convection rolls in the same direction, which evolved during WL-FPM-06

Monti et. al. in the TEXUS-9 mission observed the temperature profile and the flow velocity in a zone of silicon oil. Agreement with the computer model developed was found to be good in the laminar, non-oscillatory regime [14].

In the Spacelab-D1 experiment WL-FPM-07, *Napolitano* et al. [15] extended these investigations to non-cylindrical columns, to vibrating and rotating columns and to columns made up by two immiscible liquids. Surprisingly, the interface between the two liquids turned out not flat and tilted (Fig. 12). The shape of the interface was only slightly changed by rotation and vibration, but strongly adapted to and in turn determined the flow lines of Marangoni convection. Like in the experiment FPM-06, the formation and oscillations of a satellite drop has been observed during breakage of the zone.

7 Rectangular Containers

Another group of investigators is favouring rectangular containers with a flat free surface of the liquid. This approach is taken by *Legros*, *Lichtenbelt*, and again by *Schwabe*. *Legros* and coworkers investigate the surface tension minimum, which several aqueous solutions of alcohols exhibit. If the heating and cooling temperatures are chosen just above and below the surface tension minimum, a flow along the free surface from the centre to both sides results. This effect has been

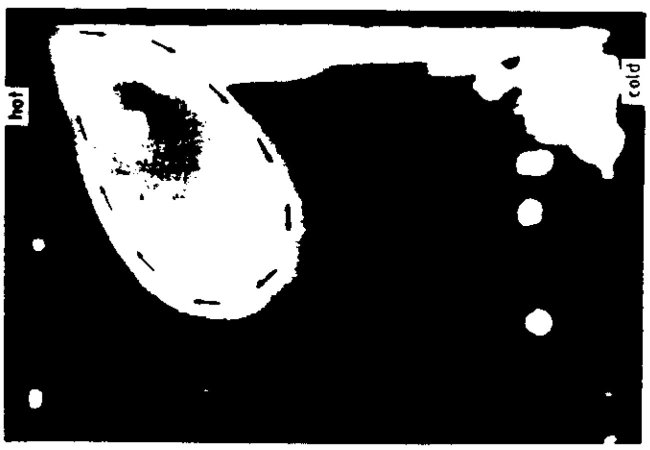


Fig. 14. Marangoni convection at the hot side of the rectangular container used in PK-MKB-00

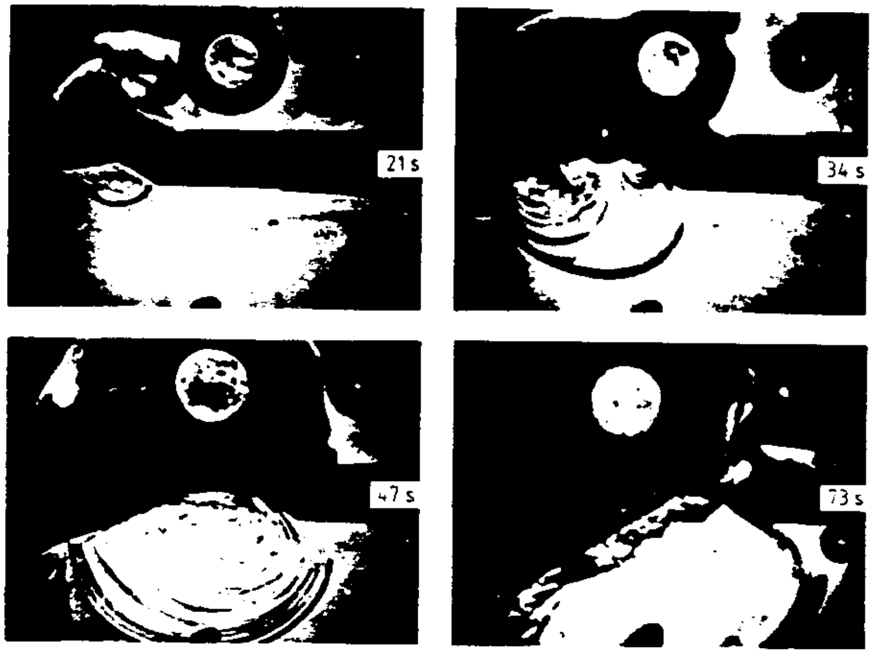


Fig. 15. The building-up and fading-out of convection rolls during diffusion of aceton from water to toluol during the TEXUS-8 experiment TEM 06-1

investigated in missions TEXUS-8, TEXUS-9, and TEXUS-13 using tracers and differential interferometry as well [16].

In the D1-experiment WL-FPM-05 the mixtures water/hexanol and water/heptanol have been used and a temperature ramp ought to be applied. Temperature control worked insufficiently, but two convection rolls in the same direction could be observed [17]. Despite careful searches, no counter convection between the two rolls has been found, see Fig. 13.

In *Schwabe's* experiment PK-MKB-00 Marangoni convection in tetracosane has been investigated [18]. The temperature difference applied was 30 K and strong Marangoni convection confined to the hot side has been observed. The tracer particles tended to stay in the vortex centre (Fig. 14).

Lichtenbelt et al., in the D1-experiment WL-FPM-01, used the rectangular container for investigating solutal interface convection. This experiment has much in common with earlier TEXUS experiments by *Brückner*. *Lichtenbelt* has studied diffusion of aceton from water to air [19, 20]. *Brückner* has investigated Marangoni convection during diffusion of aceton from water to toluol [21, 22]. Both experiments were aimed at detection of a diffusive-convective instability:

If due to a fluctuation the concentration varies along the interface, the interface tension changes accordingly and interface convection to or from the respective position results. Depending on the relative magnitude of the diffusion coef-

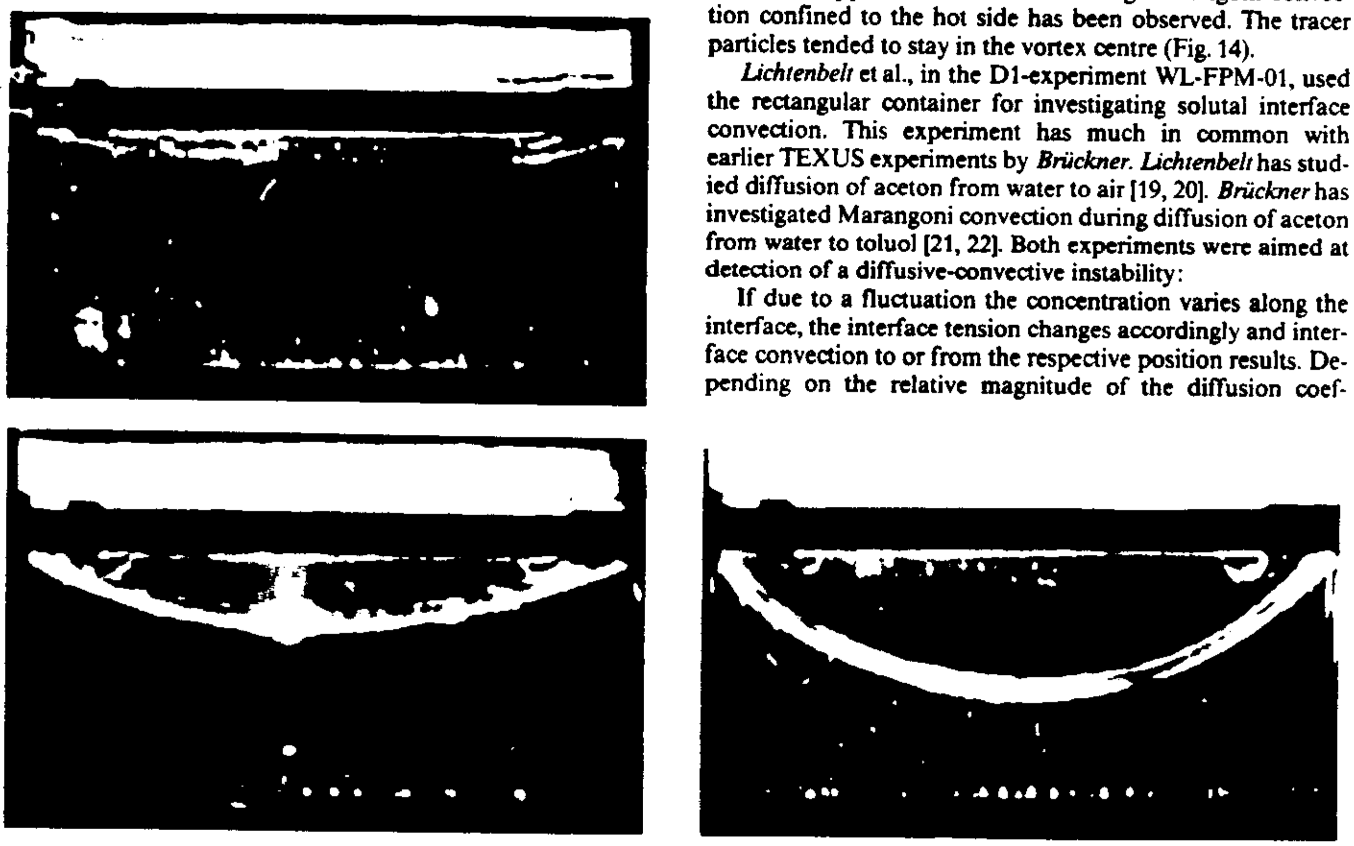


Fig. 16. In the experiment WL-FPM-01, the number of convection rolls during diffusion of aceton from water to air increased with the curvature of the fluid surface

ficients and the viscosities of both liquids interface convection stabilizes or destabilizes the initiating fluctuation in concentration.

In both experiments solutal Marangoni convection could be observed. *Brückner* found convection rolls, which usually are initiated at the edge of the piston used for keeping the liquids apart before the experiment starts (Fig. 15). In the TEXUS-11 experiment, when a larger cell was used, convection rolls were initiated also along the free interface [22]. In the D1-experiment WL-FPM-01, *Lichtenbelt* found no interface convection at the particular concentration used as long as the liquid interface was flat, and an increasing number of convection rolls, when the liquid volume was reduced, such that the interface became curved (Fig. 16). Thus, in both cases interface convection appears to be caused by geometrical reasons. It is not a fluctuation in concentration at the interface, but a systematic change in concentration due to local differences in species supply from the bulk, which initiates interface convection.

8 Drop and Bubble Migration

A third line of research is directed towards Marangoni migration of drops and bubbles in liquid matrices exhibiting temperature or concentration gradients. This research is mainly stimulated by microgravity experiments on not transparent liquids like monotectic alloys, crystal growth, etc.

Nähle et al. and *Neuhaus* studied Marangoni migration of bubbles and drops in silicon oil, WL-FPM-02 and PK-HOL-03 [23, 24]. The D1-experiments have been preceded by many experiments in parabolic flights. Different containers filled with different silicon oils were used. A temperature gradient was established in a heater fixed to the aircraft during normal flight conditions. Before each parabola one of the containers was mounted onto a platform. A bubble then was injected and its motion was observed, when the platform was free-floating (Fig. 17). In the experiment PK-HOL-03 the bubbles in the three containers were produced in the isothermal mode, before switching on the heater. Bubble migration during building-up and fading-away of the temperature gradient was recorded by holograms. The bubble velocity was zero in one of the containers and differed in the two others. Since the oils were similar with respect to viscosity, thermal conductivity, and surface tension, this has been ascribed to their differing chemical radicals [24].

Sounding rocket experiments on Marangoni migration of droplets have been flown in missions TEXUS-7 and TEXUS-9 [25, 26]. It was intended to show Marangoni convection of droplets in a transparent immiscible system. Rather than requiring droplet injection the droplets grew by cooling down the liquid system into the miscibility gap. The temperature gradient thus caused droplet growth and migration as well. Very strong Marangoni convection could be observed of cyclohexan droplets in methanol (TEXUS-7) and of methanol droplets in cyclohexane (TEXUS-9). This strongly touches all the space experiments on monotectic alloys by *Ahlborn, Fredrikson, Löhberg, Walter*, etc. All these experiments turned out actually to be experiments on capillarity spreading, thermal and solutal Marangoni convection, and droplet migration.

9 Transparent Model Experiments

The investigations into the mechanisms causing separation of monotectic systems under microgravity have been continued with the D1-experiment WL-FPM-03. In contrast to the TEXUS experiments mentioned, a free fluid surface has been chosen. A free surface generally reduces heterogeneous nucleation, but may give rise to additional Marangoni convection. A free liquid column between coaxial disks has been established and the mechanisms contributing to mixing during active heating and to separation during passive cooling have been observed [27, 28]. The main effects observed were fog

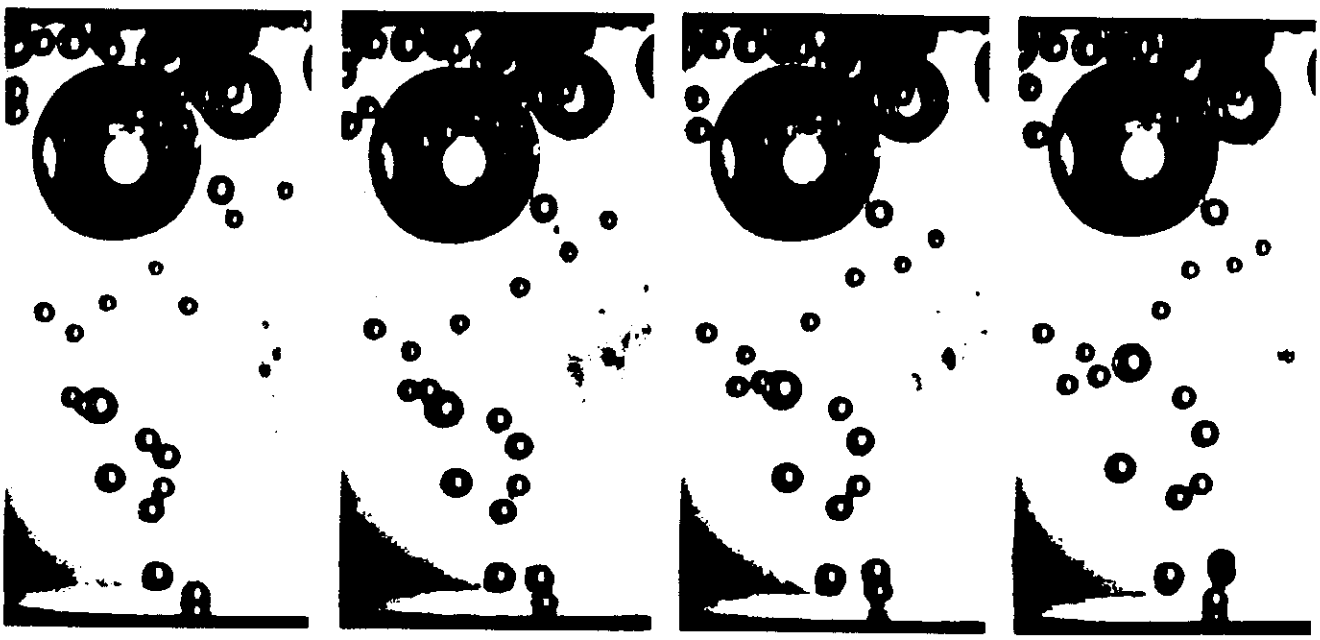


Fig. 17. Marangoni convection of bubbles during a KC-135 flight. The convective flow of the bubbles to both sides is superposed by a slow upward Marangoni convection. The time interval between two consecutive pictures is 8 s (Courtesy of D. Neuhaus)

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Fig. 18. The spreading of benzylbenzoate along the upper, heated disk in Run A of WL-FPM-03

Fig. 19. The breakage of the inner liquid column of benzylbenzoate in the outer liquid column of paraffin oil due to a Rayleigh instability in Run A of WL-FPM-03

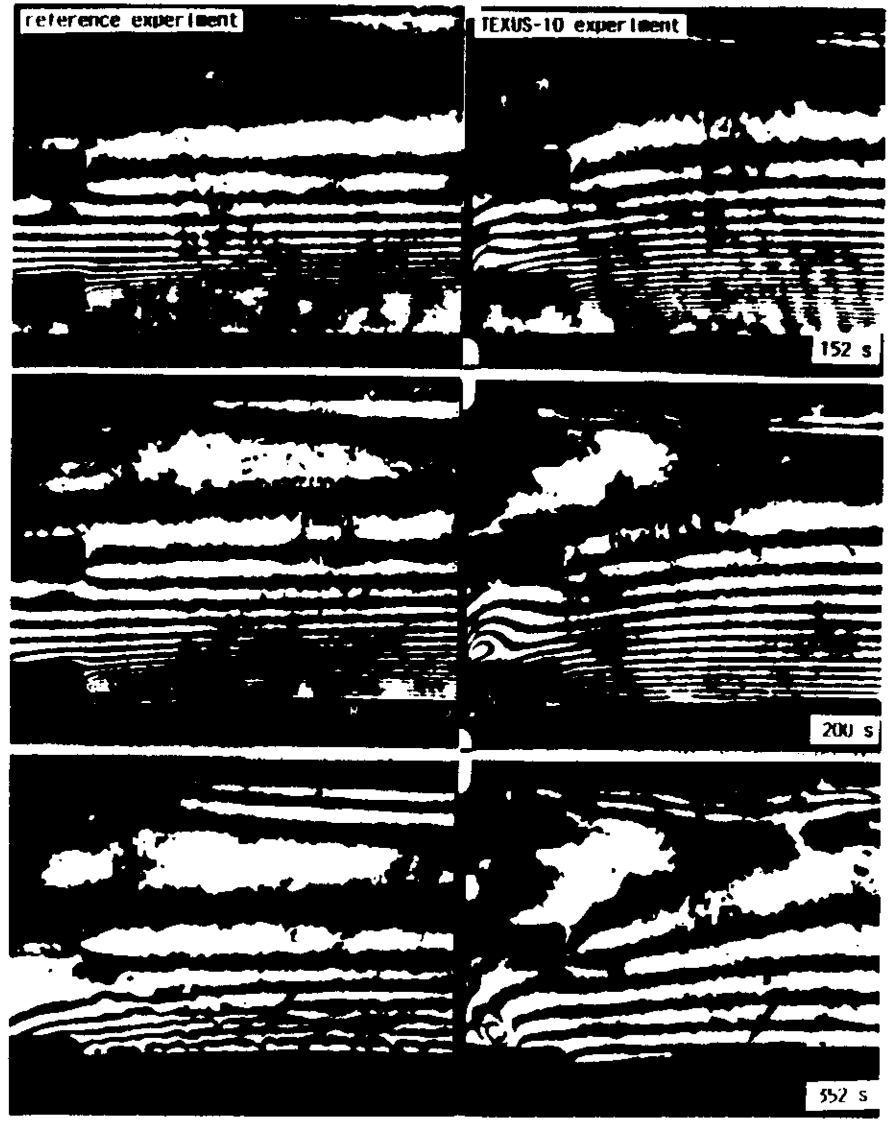

Fig. 20. Comparison of the interferograms of the TEXUS-10 experiment TEM 06-6 (right) and of the reference experiment on ground (left), demonstrating the differences in fluid flow (Courtesy of A. Ecker)

formation, thermal and solutal Marangoni convection along the free fluid surface, bubble migration and extrusion, the spreading of one component along the heated disk (Fig. 18), and capillary effects like the breaking of the inner of the two coaxial columns due to a Rayleigh instability (Fig. 19).

In another D1-experiment, in PK-HOL-01 by *Bewersdorff*, Marangoni migration of bubbles was used as the means of observation. They follow the propagation of a chemical wave [29]. Due to some unexplained failure of the holograms only a few bubbles could be observed, whose motion is in rough agreement with the expectations.

In the experiment PK-HOL-04 it was intended to observe the temperature and concentration fields and their interaction with the flow field during dendritic growth in the transparent monotectic system succinonitrile/ethanol [30]. It has been preceded by a TEXUS-10 experiment, which provided knowledge about heat and mass transport (Fig. 20). Two cells containing differing concentrations of the two liquids were unidirectionally cooled and temperature and concentration during solidification were recorded by thermocouples and differential interferometry.

Owing to malfunctions of the film transport and the laser no holograms of Run A were taken. From a second run using video recording it could be proven that the cell hardware was correctly working.

Acknowledgements

Various of the experiments performed in the Fluid Physics Module during the D1-Mission strongly profited from the high scientific and technical skill of the payload specialists R. Furrer, E. Messerschmid and W. Ockels. The team of experimenters and technical support has expressed great appreciation for their exceptionally fine performance of the seventeen experimental runs. - Many of the experimenters cited in this paper contributed valuable suggestions on the draft manuscript sent to them.

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