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Abstracts

Evaporation and thermocapillary flow near apparent contact lines on heated surfaces

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In the studies of evaporating thin films and sessile droplets the contact line is often represented as a transition region between a solution corresponding to the macroscopic shape and an ultra-thin adsorbed film in which disjoining pressure is important [1]. The standard approach to modeling the latter, based on the theory of London - van der Waals forces, is not adequate for describing many liquids used in heat transfer applications, most notably water and liquid metals. The presence of ions in water and free electrons in metals implies that the electrostatic component of disjoining pressure is also important. The focus of the present work is on incorporating realistic two-component models of disjoining pressure into studies of both steady and moving evaporating contact lines. These two-component models correspond to the pseudo-partial wetting case, i.e. they predict finite contact angle and the existence of adsorbed film in an isothermal system. An extension of the classical Frumkin-Derjaguin theory to the case of evaporating liquids is proposed. For moving contact lines, Tanner's law turns out to be followed surprisingly well even for relatively strong evaporation, although some deviations are observed as the contact line speed is suddenly changed. Marangoni stresses are shown to result in an increase of the apparent contact angle compared to the case when they are neglected. Finally, the development of fingering instability in gravity-driven evaporating liquid films is studied. It has been previously shown that evaporation can suppress this instability [2]. However, increasing the apparent contact angle by adjusting the disjoining pressure parameters or increasing the relative significance of the thermocapillary stresses both turn out to have a destabilizing effect.

^[1] Ajaev, V. S., J. Fluid Mech., 528, pp. 279-296, 2005.

^[2] Klentzman, J., Ajaev, V. S. Phys. Fluids, 21, Art. 122101, 2009.

Thermocapillary Ratchet Flows along Walls with Asymmetric Topography

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A critical need exists in developing multi-functional microfluidic devices that enable researchers to carry out complex, high-throughput analyses of fluidic samples. In many practical situations, the operational pattern is unknown a priori and, therefore, adaptable, dynamically reconfigurable devices could greatly facilitate biochemical studies and assays. Open microfluidic devices in which fluids are placed on open surfaces and are, thereby, exposed to the surrounding air could be especially useful in creating such highly adaptive microfluidic systems, where the operational sequence can be modified on demand, a property that is often difficult to achieve in enclosed systems which are typically in use today [1]. Furthermore, these free-surface or open devices are advantageous in those particular situations where a direct interaction with a gas phase is required. That includes bio-sensing, molecule manipulation, explosives detection, microchip cooling, and others [2]. What is needed, however, is an effective way to direct and regulate flow of fluidic samples in open microfluidic devices.

Recent experiments have demonstrated that fluids exposed to another fluid, e.g. gas, at their interface can be successfully transported along substrates with asymmetrical topographic grooves by simply heating these substrates [3]. The reason for the net flow in this case is the presence of non-uniform stresses at the free surface due to a non-uniform temperature in the film, which is in turn imposed by the asymmetrical substrate profile. Normally, surface tension of fluids decreases with increasing temperature and, thereby, temperature gradients at the fluid-gas interface give rise to thermocapillary stresses that entrain the entire fluid bulk into motion by viscosity.

Here, we use direct numerical simulations of the full continuity, Navier-Stokes and energy equations along with the analysis based on the long-wave approximation to examine the dynamics of thin films on substrates with periodic asymmetrical topography and heating. Using these two modeling techniques we probe how the periodical substrate and local thermal effects can be harnessed to induce and regulate directed fluid flows along the substrate. We examine emerging flow structures and characterize the fluid flow in terms of relevant dimensionless parameters. Furthermore, we study the stability of these solutions to identify the parameter range providing robust and safe operation of open microfluidic systems involving thermocapillary effects.

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[1] Whitesides, G. M., Nature, 442, pp. 368-373, 2006.

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[3] Stroock, A.D., Ismagilov, R.F., Stone, H.A. and Whitesides, G.M., *Langmuir*, 19, pp. 4358-4362, 2003.

Mixing generated by Faraday instability

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The generation of waves near the interface of one or two liquid layers that is subjected to vertical vibrations is known as the Faraday instability (see, e.g., [1, 2]). This instability occurs on account of a resonance that is set up when there is a tuning of the imposed frequency with the natural frequency of the free surface which possesses surface potential energy. Now if the free surface was removed by completely confining the container then no such instability could occur unless potential energy was introduced in some other way, say via density gradients. In this regard, we have recently shown experimentally and numerically that Faraday type of instability can also occur between two miscible liquids with different densities [3]. The amplitude of the instability grew during the experiments which then led to the mixing of the liquids (Fig. 1). The waviness of the interface finally disappeared once the two liquids were fully mixed over a volume, considerably larger than the initial diffuse region.

Here, we report on experimental and numerical study of Faraday instability used as a mixing tool. In particular, we investigate the mixing efficiency of the instability by measuring the size of the volume where the two liquids were fully mixed with the corresponding time under different external vibration parameters. An effective diffusion length based on the experimental and numerical observations is proposed. This study is numerically extended to the case where gravity is absent with potential space applications.



FIG. 1: Snapshots of Faraday instability between two miscible liquids: pure and brine water.

^[1] Benjamin, T. B. and Ursell, F., Proc. R. Soc., London A 225, pp. 505-514, 1954.

^[2] Kumar, K., Tuckerman, L., J. Fluid Mech., 279, pp. 49-68, 1994.

^[3] Zoueshtiagh, F., Amiroudine, S., Narayanan, R., J. Fluid Mech., 628, pp. 43-55, 2009.

Thermocapillary motion of viscous liquid and binary mixture in a tube domain

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We consider the motion of immiscible viscous heat conducting liquid and binary mixture with an interface. Suppose that the densities ρ_j , j=1,2, the kinematic viscosities v_j and the thermal diffusivities χ_j of the mixture and liquid are positive constants. We also assume that there are no external mass forces acting on the liquids. Under these assumptions, the equations of continuity, momentum, energy and diffusion in the cylindrical coordinates admit two-parameter subgroup of transformations corresponding to the generators

$$\frac{\partial}{\partial z} + A_j \frac{\partial}{\partial \theta_j} + B \frac{\partial}{\partial c} - \rho_j f_j(t) \frac{\partial}{\partial p_j}, \quad \frac{\partial}{\partial \varphi},$$

where A_j , B are constants and $f_j(t)$ are functions of time. The invariant solution should be sought in the form

$$u_{j} = v_{j} = 0, \quad w_{j} = W_{j}(r,t), \quad p_{j} = -\rho_{j}f_{j}(t)z + P_{j}(t),$$

$$\theta_{j} = A_{j}z + T_{j}(r,t), \quad c = Bz + K(r,t).$$
(1)

Solution (1) can be interpreted as follows. Suppose that on the interface r = a between mixture and liquid the surface tension linearity depends on the temperature and concentration: $\sigma(\theta, c) = \sigma_0 - \kappa_1 \theta - \kappa_2 c$, where $\kappa_1 > 0$ and $\kappa_2 < 0$ are constants. Initially, the mixture and liquid are at rest and occupy the cylindrical domains 0 < r < a and a < r < b respectively. At t = 0 the temperature field $\theta_j = A_j z$ and concentration field c = Bz in the mixture are created instantly in the whole domains. The thermoconcentration effect and pressure gradients $f_j(t)$ induce the motion of mixture and liquid. In this motion, the interface is represented by the cylindrical surface r = a and the trajectories are straight lines parallel to z axis. The functions W_j , T_j , K can be called the perturbations of the quiescent state.

Substituting expressions (1) in the governing equations and taking into account the conditions on the interface r = a, we obtain the three initial boundary value problems for unknowns (W_1, W_2) , (T_1, T_2) and (K). These problems can be solved successively.

The following results were obtained:

- 1) the functions W_j , T_j , K tend to the stationary state $w_j^0(y)$, $T_j^0(y)$, $K^0(y)$ as $t \to \infty$ when the pressure gradient in the mixture tends to constant;
- 2) the volume flow rates through the domains are found and inverse problem with respect to pressure gradients is solved;
- 3) the influence of thermoconcentration effect and domains thickness on the flow type is investigated numerically.

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Spreading of drops on a hydrogel

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We study the spreading of pure water drops or water drops with a surfactant (*surfactine*, produced by the bacteria *Bacillus Subtilis*) on hydrogels (Agar/Water gel). We find that water drops do not spread indefinitely on hydrogels, but remain in a state of partial wetting. Eventually the liquid diffuses into the gel on a time scale short with respect to evaporation times. The contact angle hysteresis is strong, and hardly depends on the gel concentration. We discuss what this implies for various physical effects expected near the contact line. The drops containing surfactant show a complex dynamics: at first the spreading velocity decreases, until the front stops and starts receding at about constant velocity. Concurrently, a second front detaches from the rim of the drop if the agar concentration is sufficiently low, and continues to move outwards. This dynamics is reminiscent of the spreading of totally wetting liquids in the presence of strong evaporation.[1]



FIG. 1: Wetting experiment. Left: sketch of the experimental set-up; drops are deposited on a hydrogel and filmed from above against a striped back-ground. Middle: $1 \mu \ell$ water drop on a 0.5% Agar gel. Right: evolution of the drop. The center line from the middle picture has been plotted for different times (top to bottom). The drop maintains a constant radial extension for around 90 s after deposition, then recedes and disappears. The gel around the drop can be seen swelling.

^[1] Banaha, M., Daerr, A. and Limat, L., Eur. Phys. J. ST, 166, pp. 185-188, 2009.

Faraday Interfacial Instability in Immiscible Layers

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Faraday instability is the manifestation of surface undulations when a fluid bilayer is subject to an oscillatory acceleration. These oscillations can be perpendicular or parallel to the interface. Understanding this instability is important to microfluidic mixing, bubble disintegration in microgravity capillary loops and even in the measurements of interfacial tension.

The instability can be traced to a trade-off between two effects. The first is the force due to the imposed periodic acceleration which causes the instability. The second effect is stabilizing and is made of pressure differences between crest and troughs which in turn are due to gravity and interfacial tension, the latter acting on short wavelength disturbances . The two effects can be in perfect resonance when frictional damping due to viscosity is ignored.. Otherwise they can only come into "near resonance". It is this resonance that is at the heart of the physics of Faraday instability.

The goal of this study is to develop the theoretical understanding of the physical effects that determine the appearance of the instability and to go beyond by extending this understanding to more refined aspects of the phenomena such as multiple frequency forcing, the interaction of multiple excited interfaces, and the appearance of the instability in a microgravity environment. It is the last of these that is perhaps the most exciting aspect of the work as Faraday waves are an important phenomenon to understand in the application of microgravity experiments, the mechanics of bubble breakup, and the design of microfluidic mechanical systems where mixing is pivotal in microgravity on account of the absence of buoyancy.

To determine the critical state that characterizes the instability a linearized stability theory can be employed. In this work a spectral method is used to model a three dimensional system with realistic no-slip boundary conditions, and the predictions are verified with experiments. It is further shown for a Darcy fluid the instability is not observed except for extremely high critical accelerations. The results are qualitatively compared to the Bénard problem subject to a double frequency forcing acceleration.

Marangoni Instability with Evaporation : a Transient Approach.

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A transient Rayleigh-Bénard-Marangoni convection due to cooling by evaporation is investigated. The stability of these time-dependent basic temperature profiles is studied by means of a general approach which can easily be extended to other unsteady basic flows. The method is based on a non-normal technique, and predicts the onset of instability, critical wavenumber and time. Numerical results as well as theoretical scalings for the critical parameters as function of the Biot number are presented for the limiting cases of purely buoyancy-driven and purely surface-tension-driven convection.

Comparison of the non-normal approach with the frozen-time approximation shows similar results for the critical Marangoni or Rayleigh numbers and the critical wave numbers. Moreover, the "blurriness" inherent in any transient problem is analyzed as a function of the amplification threshold values and perturbation types. It is shown that the transition region is thin compared to the large domain of Rayleigh or Marangoni numbers covered by the analysis.

Finally, a comparison with experimental results has been performed, where convection is induced by solvent evaporation during the drying of polymer solution. A good agreement was indeed found between the present theoretical study and experimental observations.

Dynamics of a thin film binary mixture: Influence of evaporation

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Surface instabilities of thin fluid layers heated from below were previously investigated in the case of a nonvolatile binary mixture [1]. Here, the Soret effect and the surface tension dependency on both concentration and temperature were considered. Recently an improved model using a systematic long wave approximation was derived. The two non dimensional variables related to the film height ξ and concentration ϕ obey to a set of equations written in a conservative formulation:

$$\partial_t \xi = -\nabla \vec{j}_{\xi} \tag{1}$$

$$\partial_t \phi = -\nabla \vec{j}_\phi \tag{2}$$

Usually, the volatility of at least one component cannot be neglected. Therefore the next step is to include the evaporation in the model, in a similar way with previous attempts [2–4]. As a first approximation the influence of the evaporation can be taken into account by additional terms representing the mass flux through the free surface, roughly estimated by a Hertz-Knudsen law. The mass flux is a function of film height and concentration and the simplest possibility is to consider a linear dependence $J = J_0 - E_1 \xi - E_2 \psi$. If one chooses the initial condition so that for a flat film one has no evaporation one has $J_0 = 0$ and then, for the linearized equations one can write:

$$\partial_t \xi = -a_1 \partial_x^2 \xi - a_2 \partial_x^2 \psi - a_3 \partial_x^4 \xi - \rho_r \left(E_1 \xi + E_2 \psi \right) \tag{3}$$

$$\partial_t \phi = -b_1 \partial_x^2 \xi - b_2 \partial_x^2 \psi - b_3 \partial_x^4 \xi + (E_1 \xi + E_2 \psi) \tag{4}$$

From the linear analysis (Figure 1) one can observe that by including the evaporation one avoids coarsening. Regular stationary or oscillatory structures are expected from fully nonlinear computations.



FIG. 1: Growthlength as function of wavelength from linear stability analysis of a system (a) without and (b) with evaporation.

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Transitions from fast to delayed coalescence: A phase field modelling

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We present a phase field approach for studying the coalescence of two sessile droplets on a highly wettable horizontal solid substrate. The drops consist of different perfectly miscible liquids and there are connected through a precursor film. For this problem we introduce two order parameters. One is the density which controls the phase of the system (liquid or gas). The second one is the relative composition which distinguishes between the miscible liquids.

Increasing the surface tension gradients between the approaching drops, different behaviors are observed. For low surface tension differences the droplet fusion occurs fast (Fig. 1-a). At higher surface tension gradients the Marangoni flow through the bridging film becomes strong enough to keep the drops separated for some seconds. The two drops start to run one after other from lower surface tension to higher surface tension, the droplet fusion is delayed (Fig. 1-b).

Using 2D phase field simulations we propose a criteria to distinguish between fast and delayed coalescence regimes. In this aim we analyze the temporal behavior of the precursor film between the drops at different viscosities and surface tension gradients. A planar two-dimensional formulation cannot catch the dynamics and the droplet curvature in the transverse direction. For a complete description quantitative computer simulations in three dimensions will be done.



FIG. 1: Phase field simulations on coalescence of two sessile droplets connected through a precursor film with Marangoni effect at the liquid-gas interface: (a) fast coalescence; (b) delayed coalescence.

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Droplet displacement and deformation induced by surface acoustic waves

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Acoustic waves generated at the surface of a solid substrate can induce deformation, displacement and even atomization of partially wetting droplets [1-3]. The characteristic time scales associated with the droplets response strongly differ from the acoustic period, suggesting the existence of nonlinear coupling between acoustic waves and droplets dynamics. If different behaviors have been observed in different experimental conditions (droplet size, acoustic wave frequency and intensity, wetting properties of the liquid), the underlying physics remains unclear. To understand it, a parametric experimental study has been performed at a fixed frequency of 20 MHz, by varying the droplet size, the liquid viscosity and the acoustic wave intensity. In these experiments, the free surface of the droplet is modified in three different way: first a breaking of its symmetry, second global oscillations of the droplet and finally small amplitude and higher frequency "trembling" modes. To explain all these deformations, two classical nonlinear acoustic driving can ve invoked: first the radiation pressure and second the acoustic streaming. The relative importance of these nonlinear phenomena strongly depends on the frequency considered. At 20 MHz, the acoustic wave is multiply reflected into the droplet and therefore the acoustic radiation pressure plays an important role. At higher frequencies, the acoustic wave hardly reaches the surface and the radiation pressure plays no role. With our experiments, we show that while both acoustic streaming and radiation pressure can induce the asymmetry of the droplet, only the latter one can explain the existence of Rayleigh-Lamb oscillation mode. We therefore exhibit for the first time the role played by the acoustic radiation pressure on droplets dynamics in a certain frequency range. The comprehension of these phenomena is of fundamental importance to enhance SAW induced deformation, displacement and mixing of small droplets.



FIG. 1: A sequence of drop deformation subjected to SAW. The drop moves from right to left at about $2 cm s^{-1}$

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Star-drops formed on an air cushion and on a vibrating surface

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In the seek for the dynamical properties of liquids drops, which are more and more used for discrete microfluidics applications, various studies have been dedicated to the determination of their eigen modes with respect to the response to external forces. In the linear regime and for invicid spherical drops, the resonance frequency has been theoretically predicted a long time ago by Rayleigh and Lamb [1]:

$$f_n = \left(\frac{n(n-1)(n+2)\gamma}{3\pi\rho V}\right)^{1/2} \tag{1}$$

in which f_n denotes the resonance frequency of the nth mode of oscillation, V is the drop volume, γ and ρ the liquid surface tension and density.

A common way to access these dynamical properties is to put a sessile drop onto a mechanical shaker, subjecting the liquid to an oscillatory inertial force [2–5]. This geometry allows for the observation of a host of phenomena from linear to strongly non-linear behavior, i.e. from surface wave undulations to strong deformations up to atomization [2]. However, an additional degree of complexity is brought by the presence of the contact line (CL), which accounts for the liquid-solid interactions at the microscopic scales. Most of the viscous dissipation occurs in the vicinity of the CL, and the combination of roughness and chemical heterogeneity of any real surface leads to a pinning force, quantified by a contact-angle hysteresis. The eigen-mode of a drop is strongly influenced by this pinning force [3], but it is possible to remove this complexity by using a low-friction hydrophobic substrate [4]. In this case the drop's eigen frequencies follow a law similar to (1), with a corrective geometrical pre-factor [5, 6] strongly depending on its shape (from a flattened sphere to a puddle) and on the contact-angle.

A drop weakly pinned on its substrate can experience a shape transition from axisymmetric to that of finite azimuthal wave-number, giving it *the shape of a star* (see Fig. 1). This transition happens in various situations, but *we restrict our study to three specific cases*: (a) when the drop sits on a hydrophobic and weakly adherent substrate that is vertically vibrated; (b) when the drop sits on a warm plate of temperature T much larger than the boiling temperature of the liquid, hence experiencing a boiling crisis - so-called 'Leidenfrost effect' - with a thin vapour layer insulating the liquid from the substrate and keeping the drop in levitation [7–10]; (c) when the drop is sustained by a gas stream going from underneath [11]. In the two last cases, the drop exhibits *spontaneous* oscillations, as the levitating flow is not time-dependant. Drops levitated on an air cushion have numerous applications: for example in lens manufacture, drops of molten glass can be prevented from contact with a solid substrate [12] by levitating the glass above a porous mould. It is also employed as a viscosimeter for harmful or high-temperature liquids [13].

Although it is now admitted that the star shapes come from a parametric Faraday-like[18] instability, originating from a periodic acceleration field, the mechanism for periodic spontaneous oscillations of the drop is not yet understood. Mechanisms invoking thermal effects are invalidated by the observation of star drops on a ambient temperature air cushion [11, 14], and an analysis only based on viscous lubrication and capillarity is unable to reproduce sustained oscillations [15]. The stability of this air cushion versus vertical oscillations is also involved in the non-coalescence of droplets on a vibrated bath



FIG. 1: (a) Star drops obtained on a shaken hydrophobic substrate. (b) Star drops obtained on an air cushion formed above a porous mould.

[16, 17]. It is also still unclear why when put on an oscillatory substrate, a sessile drop responds at the frequency of excitation whereas a non-adhesive drop undergoes a parametric instability with a response at half the frequency of excitation.

In this paper, we carry out a comparative study of puddle drops shaken on a super-hydrophobic substrate and of drops levitated onto air cushions that develop similar star shapes. By comparing our results to that in the literature (mostly obtained in the Leidenfrost situation), we investigate experimentally and numerically the possible mechanisms for the coupled dynamical and morphological transitions of the drop.

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Observation of Marangoni flow in ordinary and self-rewetting fluids using optical diagnostic systems

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As discussed in previous papers [1] dilute aqueous solutions of alcohols with a high number of carbon atoms (such as butanol, pentanol, exanol, heptanol or octanol) can be considered as "self-rewetting" fluids due to their properties associated to an anomalous dependency of the surface tension with temperature in some ranges of concentrations. According with previous work [2] for dilute aqueous solutions of these alcohols the surface tension, as a function of the temperature, goes through a minimum and there is a range of temperature in which the surface tension increases. In addition, since these solutions are in non-azeotropic compositions, alcohol preferentially evaporates in the course of liquid/vapor phase change. The surface tension gradient, caused by both temperature and concentration gradients, is direct responsible for spontaneous liquid inflow directed from cold side to hot side along a liquid-vapour interface. The expression "self-rewetting" fluids comes from such particular surface tension behaviour.

More recently, self-rewetting fluids have been proposed and investigated as new heat transfer fluids for advanced heat transfer devices, e.g. heat pipes or heat spreaders for terrestrial and space applications [3]. Within two phase heat transfer device, like heat pipe systems, liquid condensed is spontaneously driven to the hot evaporator improving the thermal performances and increasing the maximum heat transport rate.

In this paper research activity focused on numerical simulation and laboratory experiments of the behaviour of a thin layer of liquid subject to a horizontal thermal gradient. The investigate liquids include both, ordinary liquids and water/alcohols mixtures. The liquid cell has been designed to favour 2D flows and presents a configuration close to heat transfer applications. The cell also allows good thermal boundary condition and minimization of the liquid meniscus.

Flow visualization has been obtained using optical diagnostic system. In particular, in order to analyze the velocity field, preliminary experiments have been carried out adding tracer to the liquid; then, interferometric measurements have been carried out using a two wavelength Mach-Zehnder interferometer (Fig.1a).

Numerical simulations are presented to illustrate the role of the surface tension gradient. The results are obtained with computational fluid dynamics simulations of two-phase flow with a liquid–vapor interface. Model is based on the solution of the continuity, Navier–Stokes and energy equations for a viscous incompressible liquid; the boundary conditions at the liquid–vapor interface are the continuity of the mass flux and the surface balance equation of tangential momentum.

Physically properties measurements are also presented. In particular, surface tension measurements have been carried out with pendant drop technique; index of refraction has been measured using an Abbe refractometer.

A nearly 2 D flow is observed. From interferometry the thermal field is quantified and compared to the numerical results (Fig.1b). In addition, temperature and concentration distribution measurements have been carried out for the water/alcohol solution. Results highlighted self-rewetting behavior of such alcohols solutions and for the first time thermal and concentration effect has been simultaneous investigated.





(a) Experimental set up.

(b) Laboratory and numerical result.

Fig. 1 Marangoni convection in a shallow cell observed with a Mach Zehender interferometer.

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Two-phase hydrodynamic model for air entrainment at the moving contact line

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The moving contact line problems are challenging because they involve multiple length scales. One interesting case arises when an advancing liquid of high viscosity entrains the surrounding phase, such as air. In this presentation, we introduce a hydrodynamic model that generalizes the lubrication theory in order to take into account the velocity fields of the two phases. Assuming that the curvature of the interface is small we derive a differential equation for the interface profile at stationary state. We found that there is a critical capillary number above which no steady meniscus can exist and instability will occur. For example, air bubbles will be entrained into the liquid at the advancing contact line. However, we found no instability when neglecting the viscosity of the surrounding phase, illustrating the two-phase nature of the problem.

Mass transfer enhancement by Marangoni convection in a sessile droplet¹

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Marangoni convection caused by variations of surface tension with interfacial concentration is an important phenomenon in gas-liquid systems, by which the mass transfer coefficient can be intensified. The purpose of this work is to study the Marangoni effect and its influence on interfacial mass transfer in ethanol-water binary sessile droplets.

Marangoni convection in an ethanol-water droplet is qualitatively observed by a digital microscope. The vaporization of ethanol induces concentration gradient near the interface of the droplet and this gives rise to local surface tension gradients. As a consequence, Marangoni convection occurs due to the surface tension instability at the interface. The Marangoni convection on the surface of the droplet is found to be in a transient state and the velocity distribution varies with time. To further characterize the Marangoni convection flow patterns and their influence on interfacial mass transfer, experiments are also conducted to investigate instantaneous Marangoni convective velocity by micro Particle Image Velocimetry (PIV). Instantaneous velocity profiles in a slab near the top of the sessile droplet are mapped and statistically analyzed. The results by the PIV measurements show that vigorous convections occur near the droplet free surface and convections appear to be disordered and irregular in space and time. The results of the statistical analysis indicate that spatial average velocities fluctuate, decreasing with time in a certain oscillation frequency. Marangoni convection may bring the fresh liquid in the bulk to be in exposure to the gas phase and increase the opportunity for surface renewal, so that intensify interfacial mass transfer. Mass transfer coefficients for ethanol vaporization in different ethanol concentration conditions are measured by a Dynamic Contact Angle Analyzer. The analysis of the results demonstrate that the mass transfer coefficients are fluctuating with existence of Marangoni concentration in a same fluctuation periodic according with the velocity; and the ethanol mass transfer rates with existence of Marangoni concentration are found to be significantly greater than that for the pure ethanol. The conventional mass transfer correlation is also improved by incorporating the Marangoni number, Ma, a dimensionless number defined basically by the ratio of surface tension and viscous force to indicate the degree of Marangoni instability.



Fig. 1 Velocity vectorgraph of Marangoni convection in a droplet, ethanol 10wt% Fig. 2 Liquid mass transfer of ethanol in a droplet vs time

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The discrete orbits of a self-propelled wave-particle association on a rotating liquid interface

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It has been shown recently that a droplet bouncing on a vertically vibrated liquid interface can become dynamically coupled to the surface waves it excites. It thus becomes a self-propelled "walker", a symbiotic object formed by the droplet and its associated wave⁽¹⁾.

We used several experiments to investigate the properties of these walkers. We found^(2,3) that in all cases (diffraction, interference, tunneling etc...) where the wave is split, a single droplet has an apparently random response but that a deterministic behaviour is statistically recovered when the experiment is repeated. The truncation of the wave generates an uncertainty on the drop's motion. We showed that these properties result from a "path memory" effect of the walkers. The wave-field of the walker is not instanteneous but results from waves that keep being emitted by the points of the interface recently visited by the droplets.

In the present talk we will report new results on the orbiting motion of walkers when they are placed on a rotating interface. We first show that, in the rotating frame, the modulus of their velocity is practically unchanged by rotation. As expected, they have an orbiting motion due to the Coriolis effect. However when the rotation rate is increased., the radius of the orbit, instead of decreasing continuously, undergoes succesive abrupt transitions between discrete values (Fig. 1). This effect can be understood as resulting from the wave induced path-memory. As shown on Fig.2, the orbiting droplet is in interaction with all the waves it generated on its own path. Only those trajectories generating a coherent wave-field are stable.

The relation of these trajectories with the quantized Landau orbits of a charged particle in a magnetic field will be discussed.



Fig. 1 The evolution of the orbit radius as a function of increasing rotation rate.



Fig. 2 The wave field of an orbiting walker

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Capillary rising of self-rewetting solutions in laser engraved micro-channels

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The main objective of the activity presented in this paper is the development of an aluminium ultralight spreader for space applications. The main concept is a combination of grooved heat pipe, filled under vacuum with special heat transfer fluids, integrated with a thin radiator panel. Commercial heat pipes for electronic devices or for other heat transfer applications are typically filled with pure liquids, e.g. Water or Ethanol, for which surface tension driven-flows are generally directed from the higher temperature to the lower temperature regions, i.e. in the opposite direction of spontaneous capillarydriven flow, moving condensed liquid towards the evaporator thank to the grooved structure. If the direction of the flow becomes inverse, as in the case of "self-rewetting fluids, i.e. fluid mixtures with a non linear dependence of the surface tension with temperature, the situation can be drastically changed.

Self-rewetting fluids include dilute aqueous solutions of high carbon number alcohols (e-g- Butanol, Pentanol, Exanol, Heptanol. etc.) that show surface tension minima versus temperature, and the temperature dependence turns out to be positive in the higher temperature region beyond the minima. Since these solutions are in non-azeotropic compositions, alcohol preferentially evaporates in the course of liquid/vapor phase change. The surface tension gradient nearby the three-phase interline, caused by both temperature and concentration gradients, is responsible for spontaneous liquid inflow directed towards hot spot or dry patch on heater surfaces, justifying the expression of "self-rewetting" behavior [1].

The technique used in this work for manufacturing grooved micro-channels on aluminium panels is the laser engraving. The fiber laser (Yb:YAG) operating at the fundamental wavelength of 1064 nm, is characterized by a mean power of 30 W, a pulses duration of about 50 ns, pulse repetition frequency up to 80 kHz, single pulse energy up to 1 mJ. In the laser head the beam was first expanded, then directed towards two galvanometer mirrors (which determined the feature geometry to be produced) and finally focused by a "flat field" lens with a focal length of 160 mm onto the workpiece.

Two different geometries of grooves have been realized and investigated: triangular and trapezoidal sections (Fig. 1). The research activity has been focused on the experimental study of capillary rising in both types of grooves, using pure and self-rewetting liquids with well characterized surface tension and contact angle properties. In particular, self-rewetting fluids exhibit improved wettability, especially on Aluminium surfaces, in comparison to water; this is a very important property to guarantee inflow into micro-channels [2]. Using theoretical models available in the recent literature [3] capillary rising has been predicted in the capillary structure for of the different fluids, including Ethanol and self-rewetting solutions of Water-Butanol at several concentrations.

The experiments carried out using Ethanol show a good agreement with the theoretical predictions; on the contrary, in the case of self-rewetting solutions, higher values of experimental capillary rising eights are obtained. In the latter case, when evaporation occurs, the concentration of the more volatile component is reduced and the surface tension increases (Fig.2). This effect may be the main responsible of the higher capillary rising of the self rewetting solutions. In addiction, the difference in surface tension between the top and bottom of the liquid column, caused by evaporation, causes a Marangoni flow towards the region with a lower Butanol concentration (top). The coupled effect of increasing capillary pressure at liquid-vapour interface and the inverse Marangoni flow could justify the experimental results. Numerical simulations are in progress to explain the different phenomena.



Fig 1: Images of trapezoidal grooves (a) and triangular grooves (b) captured by optical microscope.

Water-Butanol Mixture



Fig 2: Surface tension measurements of water-butanol solution versus butanol concentration in water.

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From Chemo-Marangoni Cells towards Large-scale Interfacial Deformations

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Transport of chemical species across interfaces is crucial to many processes in nature and chemical engineering. Frequently, chemical reactions affecting the physical properties of the fluid are involved. In that case, the chemistry can cause hydrodynamic instabilities unexpected in non-reactive systems. The understanding of the complex spatiotemporal dynamics of such reactive fluids is much less advanced as compared to non-reactive fluids. To get deeper insights into the interplay between chemistry and interfacial-tension-driven hydrodynamic instabilities, we combined fluid dynamic and kinetic experiments, conducted on ground, with experiments performed under microgravity during ESA's Maser 10 mission.

The system studied was a quasi two-dimensional configuration of two immiscible liquid phases in contact along an initially plane interface. At this interface, a chemical reaction takes place which produces in-situ a strong surfactant. Most of the experiments were conducted in novel Hele-Shaw (HS) cells into which the two fluid chambers for the two solvents were integrated [1]. The flow patterns in the HS cells were analysed by shadowgraphy, differential interferometry and particle image velocimetry.

In the presentation we discuss the nonlinear evolution of the Marangoni convection which starts with a pattern of numerous small Marangoni cells along a nearly plane interface. In parallel with their rapid growth, the interface starts to deform in a periodic manner. With growing time, large-scale interfacial deformations develop, termed as *tongues*, which periodically move from the organic phase towards the aqueous one. An explanation of the phenomenon is proposed taking into account results of kinetic studes of the chemical system.



Fig. 1: Periodic formation of large-scale interfacial deformation in a liquid-liquid system (hexanewater) with an interfacial reaction.

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The lattices of bound states of wave interacting particles: Structure and excitation modes

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Droplets can be sustained on vibrated liquid bath. At each bounce, they emit waves on the surface. These waves mediate a long-range interaction: a droplet bouncing on an inclined surface feels its slope and moves accordingly. When they are placed close one to each other, two droplets bind. Because of the nature of the interaction through waves, the possible distances of binding of nearest neighbors are multi-valued. For large amplitude of the forcing, the bouncing becomes sub-harmonic and the droplets can have two different phases. This effect increases the possible distances of binding and the formation of various polygonal clusters is observed. From these elements, it is possible to assemble crystalline structures related to the Archimedean tilings of the plane, the periodic tesselations which tile uniformly the 2D plane with convex polygons [1]. Eight of the eleven possible configurations are observed. They are stabilized by the coupling of two sub-lattices of droplets of different phase, both contributing to sustain a common wave field.



FIG. 1: 3 snapshots of droplets crystalline aggregates with various symmetries.

Increasing the forcing acceleration, the droplets aggregates show more complex behaviour: over an onset value, they present a global and spontaneous optical vibration mode. We have investigated experimentally these vibrations in triangular and square lattice clusters and shown that there is a long range selection amongst the modes. As with the secondary instabilities of a periodic pattern, a time symmetry breaking leads to the growth of this unstable mode. A model, based on a mass-spring chain approximation and taking into account the physical properties of the interaction between the droplets explains the mode selection.

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Flow Stability in Liquid Bridges at High Prandtl Numbers

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The first goal of the paper is to extend flow stability analysis in liquid bridges for small and moderate Prandtl numbers [1] to a region of high Prandtl numbers up to 200. The second goal is to predict onset of oscillations by linear stability analysis (LSA) for MEIS-2 spatial experiment (Japan, 2009) with 5 cSt silicone oil (Pr=68).

Consider thermocapillary convection in a cylindrical half-zone liquid bridge of the height H and the diameter D. The governing equations consist of incompressible fluid flow equations in zero gravity and heat transfer equation supplied by boundary conditions. The boundary conditions are no-slip and imposed temperature difference between top and bottom rigid discs, thermocapillary stress balance and zero heat flux at free surface. The non-dimensional numbers are the aspect ratio A=H/D, the Prandtl number Pr and the thermocapillary Reynolds number Re; the Marangoni number is Ma=Re*Pr.

The linear stability analysis is applied to study critical conditions of hydrothermal-wave instability. The problem is reduced to calculation of an axisymmetric basic state solution and a generalized eigenvalue problem for three-dimensional disturbances assumed to be normal modes in axisymmetrical direction [1, 2]. The steady state solution is calculated by matrix Newton-Raphson method and the generalized eigenvalue is treated by inverse iterations.

Since the IMA-2 numerical benchmark shows difficulties in calculation of stability limits even for low Prandtl number liquid bridges [2] a code validation is quite important. The code validation includes different set of thermocapillary flows and presented in [2, 3]. The intensive validation was made for numerical results of [1]. A good agreement has obtained for H/D=0.5 liquid bridges for wide region of Prandtl number from zero to 4, however there are some discrepancy of critical Reynolds number upon aspect ratio for small Prandtl numbers. To validate high Prandtl number calculations the experiment with NaNO₃ (Pr=7) [4] was used. The linear stability analysis data has demonstrated good correspondence to the experiment and monotonic behaviour of critical Marangoni number.

Fig. 1 presents critical Marangoni number and Hopf frequency upon the Prandtl number from 4 to 200 for the cylindrical liquid bridge with the aspect ratio H/D=0.5. For high Prandtl number flows the eigen values are quite separated. The most dangerous azimuthal modes H/D=0.5 for are m=2 for lower Prandtl numbers and m=1 for higher Prandtl numbers. The modes exchange occurs at Prandtl number about 28, that corresponds to LSA results of paper [5]. Surprisingly, the Hopf frequencies of both modes are very close to each other's at the intersection point. One could see, that Hopf frequency for the m=1 mode is much sensitive to Prandtl number then for the m=2 mode. Also, Hopf frequency for the m=1 mode has monotonically decreasing behaviour with growth of Prandtl number.



Fig.1. Critical Marangoni number and Hopf frequency upon Prandtl number for H/D=0.5

The critical Marangoni number and Hopf frequency can be approximated as $Ma=6700*Pr^{0.53}$ and $freq=200*Pr^{-0.7}$ for Prandtl numbers more or equal to 20 (black curves in fig. 1). It should be noted,
however, that the most of experimental data are fitted by $Ma=2000*Pr^{0.6}$ for $10^{-2}<Pr<10^{2}$ that is much lower (see, for example, [6] for analysis of critical Marangoni number behaviour upon Prandtl and aspect ratio). The difference in the LSA and the experimental critical Marangoni numbers could be explained, most probably, by a non-zero (and non-uniform) heat flux through the free surface, an influence of the gravity and a non-cylindrical shape of the free surface of liquid bridge.

Fig. 2 presents the critical Marangoni number and the Hopf frequency upon aspect ratio H/D for cylindrical half-zone liquid bridge of 5cSt silicone oil (Pr=68). That correspond to some set of data of MEIS-2 spatial experiment (Japan, 2009). The blue data corresponds to LSA analysis and the red data corresponds to the mentioned experiment. Both data correspond to the m=1 azimuthal wave number. The reason to apply LSA was a non-monotonical behaviour of the experimental critical Marangoni number. The comparison of experimental and LSA data is quite amazing. Some set of the critical Marangoni numbers correspond quite well to each other (H/D=0.875; H/D>1.2). But, for other set of aspect ratio the difference in the experimental and the LSA critical Marangoni numbers the difference is very large. Namely, for H/D=0.5, corresponding to many experimental and theoretical studies, the difference is about 3 times (remember theoretical fitting Ma=6700*Pr^{0.53} and experimental one Ma=2000*Pr^{0.6}). For aspect ratio H/D from 0.5 to 0.7 the difference in critical Marangoni numbers are even higher. Nevertheless, the behaviour both theoretical and experimental critical Marangoni numbers exhibit two extremums. That might be a key circumstance to support "two-extremum" behaviour of the experimental neutral curve from a side of LSA stability analysis and a way to further understanding based on conjugate models of half-zone liquid-bridge stability analysis. The Hopf frequency upon aspect ratio has one extremum and a gap at another point. In might be used for additional identification of branches of neutral curve. The origin of instability is identified by evaluation of aposteriori energy balances.



Fig.2. Critical Marangoni number and Hopf frequency for MEIS-2 experiment and LSA

The linear stability analysis for MEIS-2 spatial experiment is done within collaboration with Prof. H. Kawamura lab.

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Longwave Marangoni instability in a binary mixture under the action of vibration

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We consider a longwave Marangoni convection in a layer of a binary mixture with the Soret effect. The surface tension at the free nondeformable surface depends on both the temperature and solute concentration. The heat flux at the free surface is governed by the Newton's law of cooling. The lower solid boundary is subject to vertical vibration; a constant heat flux is specified at the substrate. In the absence of vibration both monotonic and oscillatory modes were found [1].

We assign the vibration period to be large enough, so that it is comparable with the characteristic time of the evolution of the system found in [1]. The amplitude of vibration is large with respect to the layer thickness, which ensures the vibration impact to be finite. The thermal expansion is taken into account only in vibration-induced term, whereas the gravity-induced buoyancy convection is neglected. In order to suppress the Faraday instability [2], we assume the reference surface tension to be rather large. The Biot number is small, which warrants the emergence of the longwave instability.

This problem admits a base state which corresponds to linear distributions of both the temperature and solute concentration in motionless liquid. We study the linear stability of this state with respect to the longwave perturbations.

By means of lubrication approximation, the set of equations for small perturbations reduces to the following amplitude equation:

$$\ddot{\rho} - \alpha(\tau)\dot{\rho} - \gamma(\tau)\rho = 0, \tag{1}$$

where ρ is the density perturbation, the dots denote the derivatives with respect to the rescaled time τ and both the coefficients α and γ are cumbersome functions of parameters, including the rescaled wavenumber K and Biot number β . The term $A \cos \Omega \tau$ enters both above-mentioned coefficients, i.e. the amplitude equation is reminiscent to the well-known Mathieu equations. (Here A and Ω are the rescaled dimensionless amplitude and frequency of vibration.) Later on, we use the conventional classification of perturbations: synchronous perturbations oscillate with the period of vibration, subharmonic perturbations have a doubled period, and for quasi-periodic mode the perturbations are the superposition of two signals with incommensurable frequencies.

The amplitude equation was solved numerically by means of the Floquet theory, several limiting cases were treated analytically. At small amplitude, both subharmonic and synchronous modes emerge, when oscillatory mode takes place in the absence of vibration. The resonance condition in this case serves for the selection of the critical wavenumber. The correction to the critical Marangoni number is always nonpositive and it is proportional to A for the subharmonic mode and to A^2 for the synchronous mode. Therefore, subharmonic mode is critical for small A. In "longwave" approximation, $K \ll 1$, for $\beta = 0$ the Marangoni number coincides with the case of A = 0 [1], which means that the vibration can only destabilize the layer. The first nontrivial correction is proportional to K^4 in this case. In "shortwave" limit, $K \gg 1$, Wentzel-Kramers-Brilloin method was applied. It should be noted that because the analysis is done in the framework of the lubrication approximation, the case $K \gg 1$ corresponds to an intermediate asymptotics rather than a true short-wave asymptotics. A full stability problem for arbitrary values of unscaled wavenumber k will be a subject of a future separate analysis. The limit $K \gg 1$ provides an essential (but only sufficient) condition of the instability. Let us start the discussion of the numerical results with the negative values of the Soret number χ , when the oscillatory instability takes place in the absence of vibration [1]. As it has been stated above, at small *A* the subharmonic mode is critical. With further increase in the vibration amplitude, substantial decrease in the critical Marangoni number is observed. This decrease is well-pronounced for both the synchronous and subharmonic modes, which start to compete, see the lines in the upper part in Fig. 1. It is clear, that both these modes are resonant.

Above a certain amplitude of vibration, the vibration-induced mode appears, see the lines in the lower part in Fig. 1. This mode is synchronous and it takes place even in absence of surface-tension-driven convection.

The increase in β leads to the stabilization of the base state with respect to the both resonant modes, see Fig. 1 (left), since the energy dissipation grows as β increases. On the contrary, for the vibration-induced mode with growth in the rescaled Biot number, the stability domains shrink. This effect is rather unexpected, since additional loss of heat amplifies the convection.



FIG. 1: Neutral stability curves for $\chi = -0.05$. Red, blue, and black lines correspond to synchronous, subharmonic, and quasi-periodic modes, respectively. Left: solid line $\beta = 0$, dashed line $\beta = 0.2$ at A = 0.6. Right: solid line A = 0.4, dashed line A = 0.6 at $\beta = 0.2$. Domains of stability are marked with "S."

In Fig. 1 (right) the neutral stability curves are plotted for two different values of the vibration amplitude at fixed value of β and for the same value of negative Soret number. It is seen that increase in A amplifies the Marangoni convection, destabilization of the layer with respect to both resonant perturbations and vibration-induced synchronous mode takes place.

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Stability of a two-layer quasi-incompressible binary-fluid system with a diffuse interface

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A system of two layers separated by a diffuse interface, which is created due to a phase separation in a binary liquid in the gravity field, is considered. The system is bounded by a planar solid substrate from below and by a gas phase from above. The deformable liquid-gas interface is considered as a sharp boundary.

The goal of the present work is the investigation of the linear stability of two-layer base solutions with respect to long-wave disturbances. In a contradistinction to previous works on that subject [1-3], the dependence of the density on the mass fraction is taken into account, which is significant under the action of gravity. In order to investigate this problem under non-equilibrium conditions, a mathematical model, which consists of the continuity equation, Navier-Stokes equation and modified Cahn-Hilliard equation governing the motion of quasi-incompressible fluid, is formulated. This model extends the model suggested in [4].

The linear stability analysis of the system is carried out in the general case, and a longwave limit of the general dispersion relation is derived. The influence of gravity, solutocapillary effect at the free boundary and Korteweg stresses inside the diffuse interface on the stability is studied. Parameter regions of long-wave instabilities are found.

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Orbital motions of a drop levitating around a circular hydraulic jump.

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We have investigated the structure of a hydraulic jump formed by impacting with a viscous liquid a horizontal or slightly inclined disk, and explore the dynamics of a liquid levitating drop, of the same liquid trapped around the jump. Depending on the conditions (plate inclination, drop diameter, liquid properties) different regimes are observed: static drop [1], drop oscillating around the equilibrium position [2], drop rotating around the jump [3-4], chaotic oscillations [4]. We have built a reasonably simple model of this motion by treating the drop as a spherical rotating solid, with two points of contact with the liquid. In the case of a horizontal jump, we show that, rather surprisingly the air thickness disappears from the equations, the period of the orbit being proportional to the quantity $Q/(rR^2)$, built on the flow rate Q, the drop radius r and the jump radius R. In our conditions, the jump radius R seems to be close to the value predicted by Bohr et al. [5], and we also provide a simple model to recover this scaling law in our conditions.



Fig. 1: A typical chronophotography of the drop motion, showing the orbital case (a) for a horizontal jump, and the equivalent spatio-temporal diagram (b); (c) chaotic case, in which the drop motion becomes irregular.

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Short- and long-wave Marangoni instabilities in liquid films on walls with spatially-periodic temperature distribution

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Liquid films on heated substrates are subject to short- and long-wave modes of Marangoni instability. The short-wave instability leads to the development of convection cells, whereas long-wave instability is one of the primary causes of the film rupture [1].

In many practical situations Marangoni convection and interface deformation in liquid films are induced by nonuniformities of the temperature at the liquid-gas interface, which arise due to the wall topography [2-4], due to the local heating of the wall [5] or due to the nonuniform thermal properties of the substrate [6]. These phenomena significantly affect the film stability and pattern formation in the film, as well as concomitant heat and mass transport [4]. However, the mechanisms of the influence of wall/liquid interface on Marangoni instability are not completely understood. Until now, only the effect of long-scale wall topography on long-wave Marangoni instability has been studied theoretically and numerically [2, 4].

In the present work the effect of spatially-periodic wall temperature distribution on the short- and long-wave Marangoni instabilities in a liquid film is studied by solving the Orr-Sommerfeld equation. The spatially-periodic boundary condition corresponds to the condition of the local heating of the wall and to the heated wall having spatially-periodic thermal properties of the substrate [6]. It is expected that the stability properties of the film with spatially-periodic wall temperature distribution are closely related to those of films on walls with periodic topography.

We show that the effect of the wall temperature distribution on the Marangoni instability depends on the period of the temperature variation and on the Marangoni number. If the period of the temperature variation is much larger than the film thickness, the long-wave Marangoni instability is reinforced. This result is illustrated in Fig. 1 showing a significant increase of the disturbance growth rate due to long-scale periodic variation of the wall temperature.



Fig. 1 Disturbance growth rate Ω as a function of the wave number K for a thin liquid film on a substrate with long-scale periodic temperature variation. A denotes the dimensionless amplitude of the temperature variation

We have also found that in a certain range of parameters the Marangoni instability can be suppressed by applying the wall temperature variation with a period comparable with the film thickness.

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Instability invoked by gas flow along thermocapillary interface

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Gas-liquid flows induced by gas stream in annulus and Marangoni convection on free surface are analyzed for fluids in large range of gas flow rate and temperature difference. The gas enters into the annular duct and entrains initially quiescent liquid. The problem is solved numerically in complex geometry, which corresponds to a liquid bridge axially placed into an outer cylinder with solid walls. The internal core consists of solid rods at the bottom and top, while the central part is a relatively short liquid zone filled with viscous liquid and kept in its position by surface tension. The solid rods have a complex geometry; they contain grooves and protrusions at the bottom and top parts closest to the liquid.

The behavior of the liquid flow under mechanical stresses produced by co- or counter flow of the ambient gas is one of the points which were studied (see Fig. 1). Depending on gas flow rate and temperature difference between rods four different flow regimes were classified including oscillatory. For each flow regime the heat fluxes through the surface have been analyzed.

The aim of this study is concerned to space experiment JEREMI (Japanese European Space Research Experiment on Marangoni Instabilities) which is devoted to the study of the threshold of hydrothermal instabilities in two-phase systems in cylindrical geometry. The experiments is planned to be performed in 2012 in the Japanese module on ISS using the dedicated FPEF (Fluid Physics Experiment Facility).



FIG. 1: Temperature distribution for downstream (left) and upstream (right) gas flow.

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Liquid entrainment by gas flow along the interface.

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Abstract. We report the results of numerical and experimental study of two-phase flows in annulus. The geometry corresponds to a cylindrical liquid column coaxially placed into an outer cylinder with solid walls. Gas enters into the annular duct and entrains initially quiescent liquid. The internal column consists of solid rods at the bottom and top, while the central part is a liquid zone filled with viscous liquid and kept in its position by surface tension. Silicone oil 5cSt was used as a test liquid and air and nitrogen as gases. An original numerical approach was developed for study the problem in complex geometry. The flow structure in the liquid is analyzed for wide range of gas flow rates.

1 Introduction

Heat/mass transfer on the moving gas-liquid interface is an important subject directly related to many industrial applications from crystal growth to cooling of electronic devices. In the present study, the attention is focused on flows in cylindrical geometry. There exists a large number of studies of two-phase flow through the circular pipe. Another well studied problem is two-phase flow in annuli when liquid films covered internal rigid wall of a cylinder and gas stream is moving along the center of the channel. In present study liquid occupies the central part of the channel and gas is moving in duct between liquid core and rigid wall. Such systems have a large field of applications in the production of highest quality semi-conductors [1], fibers, micro-jets [2], etc.

The best quality semi-conductor crystals are grown by floating zone technique. Studies on the mechanisms governing the flow dynamics and heat transfer are particularly helpful in controlling the crystal growth process and thereby the quality of its products. The scientific model, which is used to mimic industrial floating zone technique, is called a liquid bridge. During last decades, a significant progress has been achieved in understanding nonlinear regimes of buoyant-thermocapillary convection in liquid bridges. However, only recently it was recognized that the flow in surrounding gas phase and heat/mass transfer at the interface are important factors for the stability of flow inside liquid bridge. The experimental results were the first to indicate the extreme sensitivity of flow stability in liquid bridges to environmental conditions. Experiments performed in different configurations by Shevtsova *et al.* [3,4], Mialdun *et al.* [5] and Kamotani *et al.* [7], [6] have demonstrated a similar tendency that ΔT_{cr} changes by a factor of two or three by varying the air temperature relative to the cold wall temperature.

The number of publications considering flow both in liquid bridge and surrounding gas is still very limited, see [9], [11]. As a rule, the flow is considered inside liquid, and the boundary conditions on the free interface capture the impact of surrounding gas through the Biot number, e.g. [10]. Note that experiments by [6] have shown that the disturbances of gas flow near the interface produce significant impact on the stability of the liquid flow.

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Fig. 1. Geometry of the problem. (a) Geometry and scales used in numerics (b) Photo of magnified liquid zone from the experiment; (c) Sketch of an overall experimental set-up.

The present study provides insight into the role of shear stresses on the interface. The emphasis is on isothermal, adiabatic two-phase gas-liquid flow in cylindrical geometry without mass transfer in configuration where liquid zone has limited length.

In the numerical part the gravity effect is not included into consideration so the channel orientation is not important. In addition, these results remains valid for flow in the micro- and large nano-channels with cross-sectional diameters of the order 300-500 nm with one-two order of magnitude reduction in the hydraulic diameter, see [8].

2 Geometry of the problem

During the experiments, external gas is blown into the cylindrical channel through a system of equally distributed nozzles to avoid azimuthal motion of gas around the rod (to avoid kind of vortex tube). It provides quasi-constant velocity at the inlet. The gas at a constant flow rate enters into the annular duct and entrains initially quiescent liquid. Usually numerical studies consider only the flow inside liquid zone. However, experimental set-ups include rods of different lengths and configurations. Main accent in this paper is on numerical study with a geometry as close as possible the laboratory set-up is used. The geometry of the problem and notations used throughout the paper are shown in Fig. 1a. Photo of the central part of the system, i.e. liquid bridge is shown in Fig. 1b, while the sketch of overall experimental design is presented in Fig. 1c.

Let us look at the internal core of the system. Liquid bridge itself, Fig. 1b, is a drop of liquid kept by surface tension between two solid rods. Usually, the system is heated from above. The temperature of the lower rod is either kept constant or decreased to maintain constant the mean temperature in the system. The thermocapillary flow arises in liquid for any temperature difference ΔT between the supporting rods. To prevent fluid from creeping over the edges of the rods, they have to be coated with an anti-wetting-barrier, which depends on the liquid and on the material of rods. To skip anti-wetting coating or to reinforce arrangement, the bottom rod is often processed into a sharp edge, e.g. see [4], [12]. In the experiments with large ΔT , while heating from above, the use of anti-wetting-barrier may be not sufficient for prevention of creeping. In this case, a groove is also made in the upper rod, see Fig. 1a. This type of geometry was used in experiments by Kamotani [6] and in our laboratory experiments. This internal parts are surrounded by co-axial cylindrical tube of radius R_{out} .

3 Mathematical model

To study the influence of shear stresses imposed on the interface, various gases are blown in the cylindrical channel with different velocities. Gas of constant flow rate Q_{in} flows through annular duct of outer diameter $r = R_{out}$ and inner diameter $r = R_0$, see notation in Fig. 1a. Reaching the liquid zone after distance H from the tube inlet, it creates flow in quiescent liquid due to balance between viscous stress and shear stresses. After passing the liquid zone of length d, gas continues to flow through annular duct of inner radius $r = R_0$ over the distance H up to the tube outlet. The total length of the tube is L = 2H + d. The radius of liquid zone is R_0 . The geometrical scales are given in Table 1. In addition, size of triangle grooves is 0.5mm in horizontal and vertical directions. The gas is assumed incompressible in consistency with the low pressure drop between the tube ends. Hereafter, the hydraulic diameter,

$$D_h = 2\,R_{out} - 2\,R_0,\tag{1}$$

will be used as characteristic length scale.

The dynamics of system in the geometry of Fig. 1a is described by the momentum and continuity equations for an incompressible Newtonian fluid in gas $(^g)$ and liquid $(^l)$ in cylindrical coordinates (r, z):

$$\mathbf{V}^{\mathbf{g}} \cdot \nabla \mathbf{V}^{\mathbf{g}} = -\frac{1}{\rho^g} \nabla P^g + \frac{\mu^g}{\rho^g} \nabla^2 \mathbf{V}^{\mathbf{g}}$$
(2)

$$\nabla \cdot \mathbf{V}^{\mathbf{g}} = 0, \tag{3}$$

and

$$\mathbf{V}^{\mathbf{l}} \cdot \nabla \mathbf{V}^{\mathbf{l}} = -\frac{1}{\rho^{l}} \nabla P^{l} + \frac{\mu^{l}}{\rho^{l}} \nabla^{2} \mathbf{V}^{\mathbf{l}}$$

$$\tag{4}$$

$$\nabla \cdot \mathbf{V}^{\mathbf{l}} = 0, \tag{5}$$

where the velocity $\mathbf{V} = [V, U]$ includes the radial V and axial U components; μ is the dynamic viscosity and ρ is the density. Boundary conditions are: on the liquid–gas interface $r = R_0$:

$$\mu^{l} \frac{\partial U^{l}}{\partial r} = \mu^{g} \frac{\partial U^{g}}{\partial r}, \qquad U^{l} = U^{g}, \qquad V^{l} = V^{g} = 0$$
(6)

on liquid bridge axis r = 0:

$$\partial_r U^l = 0, \quad V^l = 0, \tag{7}$$

on the wall of the external tube $r = R_{out}$:

$$U^g = 0, \quad V^g = 0,$$
 (8)

no-slip conditions are also imposed on the supporting rods.

Inlet conditions between cylinders $z = 0, r = R_0 < r < R_{out}$.

$$U = U_0 = const, \quad V = 0 \tag{9}$$

or constant flow rate, calculated as $Q_{in} = U_0 \pi (R_{out}^2 - R_0^2)$.

Outlet conditions at z = L, $R_0 < r < R_{out}$. The flow velocity and pressure at the outlet are not known prior to solution of the flow problem. The zero flux conditions applied at outflow boundaries are approached physically in fully-developed flows, i.e. flows in which the flow velocity profile is unchanging in the flow direction.

$$\partial_z U^g = 0, \quad \partial_z V^g = 0 \tag{10}$$

The question whether these conditions allow perturbations to penetrate back inside the tube have been investigated. As a result, the simulations were performed with extended upper part,

Table 1. Geometrical scales used in calculations, unless otherwise stated.

R_{out}	R_0	D_h	H	d	L
10 °m	10 °m	10 °m	10 °m	10 °m	10 °m
5	3	4	20	3	43

z = L + H/2, to minimize artificial boundary effects in the results obtained. However the results below are analyzed for initially prescribed length, 0 < z < L, and extended zone is not under discussion.

Commercial solver FLUENT v.6.3 (laminar steady) was used for computations of governing Eqs.(1)-(11). Computations in multi-domains require a complicated computational mesh, which was generated by commercial code GAMBIT. The solution method and code validation procedure one may find in [14].

4 Results and discussion

For a two-phase system of a liquid entrained by gas, different internal flow structures can occur depending on the size or shape of the flow channel, the magnitudes of the gas flow parameters, and on the physical properties of the two phases. Two gases were used in the experiments: air and nitrogen. Silicone oil 5cSt was used as a test fluid. Since the fluid is transparent and colorless the optical observations were performed using two sources of light. Back white light was used for illumination of the entire liquid bridge with a purpose to measure the interface shape. He-Ne laser equipped with cylindrical lens was used for producing of a light sheet aimed at particle tracking velocimetry. The shape of interface was measured according previously developed technique [13]. The details of particle tracking velocimetry will be published elsewhere.

The relative volume in the experiment was kept close to unity, $V/V_0 \leq 1$, $V_0 = \pi R_0^2 d$. Even in this case the liquid bridge interface is deformed, see photo in Fig. 1b. On one hand we are familiar with the commercial code for the flat interface. On the other hand we have in-house made code for liquid bridge with deformable interface in single phase (liquid), see [15]. Our expectations was that flow in the liquid entrained by gas will be relatively weak and interface deformation will not strongly affect the velocity field in fluid. It will allow us to use commercial code in complex geometry for direct comparison with the experimental results.

To clarify the role of surface deformation on the strength of the velocity field we perform a dedicated study in two steps. In our experiments gas enter to the annular gap through the top. In some way there is analogy between the actions of Marangoni force and gas flow.

$$\mu^{l} \frac{\partial U^{l}}{\partial r} = \mu^{g} \frac{\partial U^{g}}{\partial r}, \quad (\text{shear}) \qquad \mu^{l} \frac{\partial U^{l}}{\partial r} = \frac{\partial \sigma}{\partial T} \frac{\partial T}{\partial z} \quad (\text{Marangoni}) \\ \rightarrow \quad \mu^{g} \frac{\partial U^{g}}{\partial r} = \frac{\partial \sigma}{\partial T} \frac{\partial T}{\partial z} \quad (11)$$



Fig. 2. (a) Interface velocity when fluid is driven by gas $Q_{in} = 2.3ml/s$ (blue curve), and by Marangoni force, Ma = 144 (red curve); $\mu_l/\mu_g = 255$. (b) Radial velocity for straight and deformed interface.

At the first step using both codes for flat interface (in-house and Fluent) we identify ratio of Marangoni force and shear stresses, which produce the similar interface velocity. For example for the considered geometry the Marangoni driven flow for Ma = 144 ($\Delta T = 0.25K$) is similar to that, produced by blown gas with flow rate $Q_{in} = 2.3ml/s$ ($U_0 = 0.045m/s$). For these parameters the distribution of the velocity along the interface is shown in Fig. 2a. The velocity maximum in the case of Marangoni flow (blue curve) is shifted in the direction of the flow, while flow driven by gas (red curve) is almost symmetrical with respect to mid-height of zone.

On the second step simulations were performed in the liquid zone both with straight and deformed interface for Ma = 157 ($\Delta T = 0.35K$) using in-house code. Variation of the axial velocity with radius at mid-height of the liquid zone is shown in Fig. 2b for: (blue curve) straight interface, g = 0; (magenta curve) straight interface, $g = g_0 = 9.81m/s^2$; (green curve) deformed interface, $g = g_0$. In the case of deformed interface the velocity near the symmetry axis is larger than at other cases. Also in this case the interface velocity maximum is located near the mid-height of liquid zone. The difference in strength of the flow between considered cases is relatively small. Thus, performing two-phase calculations in the geometry shown in Fig. 1a with the straight interface of the liquid zone, the error will not be very large in the case of "slim" liquid zones (i.e. V < 1).

The values $\Delta T = 0.25K$, 0.35 were chosen not occasionally. Surprisingly, in the experiments we observed the motion in the fluid at zero gas flow rate without obvious driving force. Our dedicated study showed that temperature difference $0.25K < \Delta T < 0.35K$ arises in the "isothermal" system (liquid) because of working lasers, lights and cameras. The measured velocity, caused by this non-uniform heating in the system, is shown in Fig. 2b by symbols. Originally this range of ΔT was found by using thermocouples. Then the velocity caused by the ΔT was determined by using particle tracking velocimetry and compared with simulations, Fig. 2b. The numerical results display an agreement with experimental one and confirmed the origin of the motion. Procedure and software for particle tracking velocimetry were developed by one of the authors. This technique also allows to follow trajectory of the particles. Well-marked trajectories of a few particles are shown in Fig 3a.

The experiments and corresponding numerical simulations were conducted for wide range of flow rates. The flow entrained with gas at flow rate $Q_{in} = 77 \, ml/s$ is shown in Fig 3b. In the two-phase calculations the additional flow rate $Q_{in} \approx 3 - 4 \, ml/s$ (because of nonuniform heating) were taken into account for comparison with experimental data. An excellent agreement between computed results and experimental data demonstrates that the developed experimental technique and numerical code are capable to capture the main characteristics of the phenomenon studied.

The dependence of liquid velocity upon the gas flow rate is shown in Fig 4. Numerical results are shown by solid curves while the experimental one by symbols. Blue curve presents maximal velocity (absolute value) attained at the interface (see profile in Fig 3b). In the experiments due to liquid zone curvature the velocity is measured not at the interface, but nearby. Correspondingly, the experimental points, dark blue rhombus, lay below the numerical curve.



Fig. 3. (a) Trajectory of the particles moving due to temperature non-uniformity induced by experimental facility. (b) Entrainment of fluid with gas; two-phase calculations (solid curve) and experimental points $Q_{in} = 77ml/s$ (silicone oil 5cSt and air, $\mu_l/\mu_g = 255$)

 $\mathbf{5}$



Fig. 4. (a) Liquid velocity as function of gas flow rate ; $Rm \approx 2.2 - 2.4 mm$.

The axial velocity in the bulk near the sign change is shown by green curve (numerics) at r = Rm = 2.4 mm and by pink cubes (experimental). The larger value of the measured velocity is attributed to the small uncertainty in the radial position of particle due to liquid zone curvature. The most probably that the measures velocity corresponds to particle position at $r = Rm \approx 2.25 - 2.35 mm$. We may conclude that the agreement between numerical and experimental results is good. Both sources display linear dependence of the flow strength with the increase of gas flow rate.

5 Conclusions

An experimental set-up and numerical approach have been developed to study gas-liquid flows in annulus. The geometry corresponds to a cylindrical liquid column co-axially placed into an outer cylinder with solid walls. Gas enters into the annular duct and entrains initially quiescent liquid. The flow structure in the liquid is analyzed for wide range of gas flow rates. Good agreement between computed results and experimental data demonstrates that the developed experimental technique and numerical code are capable to capture the main characteristics of the considered phenomenon. The study to be continued for non-isothermal case.

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Impact of electric fields on the speed of contact line in vertical deposition of diluted colloids

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Colloidal crystals have gained a remarkable attention from research groups across the world due to its simplicity in fabrication and broad range of applications. A number of simple techniques and quick methods (mostly derived from the Langmuir-Blodget technique) are available for preparing colloidal crystals and vertical deposition is one of the simplest [1, 2].

We report the experimental results on the impact of DC electric fields on the deposition of negatively charged micron-sized colloids. We measure the speed of a receding contact line of water-based colloidal suspension in vertical deposition configuration. The obtained speeds are further compared with the dried structures, which reveals their relationship. This further explains the reason behind in obtaining different and varying speeds at a wide range of voltages and concentrations.

The experiments are performed using an evaporation cell at a controlled temperature and fixed humidity. The cell is designed in such a way that two substrates can be placed vertically and the distance between them is of 1 mm. Also, the design allows us to follow the receding contact line which is recorded by a video camera. The electric fields are of the order of 1 V/mm and are applied perpendicular to the ITO coated glass substrates. We measure *in situ* the evolution of the contact line on the substrate where the voltage is applied with respect to the ground.



FIG. 1: Characteristic speed of contact line as a function of applied voltage for 0.3% (w/w) concentration.

In this work we measure the speed of contact line at different applied voltages and also at different concentrations (from 0.1% to 0.5%). For every experiment, we construct a histogram of these speeds for every position and time. The characteristic speeds are calculated by fitting the cumulative histogram with normal distribution functions. The mean value of each distribution represents the corresponding characteristic speed. The standard error of the fitting is the error in obtaining this characteristic speed.



FIG. 2: Optical micro-graphs (Magnifications: First row-50X, Second row-20X) of the colloidal crystals at 0.3% concentration. A.- non-compact (NC) sparse deposit, B1,B2.- non-compact dense (NCD), C.- compact monolayer (CM), and D.- multilayer (ML). Scale bars are 25μ m.

At 0.3% concentration (Fig.1), the characteristic speeds have a varying slope as we decrease the voltage. These speeds mainly depend on local concentrations, forming morphologies, previously deposited structures and dynamical perturbations (unbalanced forces due to pin-depin in the contact line at the counter substrate). All these factors may occur in the same experimental realization and they are interrelated. The morphologies (Fig.2) dominantly observed in these kind of systems are multilayer (ML), compact monolayer (CM), non-compact dense (NCD) and non-compact sparse (NC) deposit. In addition to these structures, vertical non-dense stripes (at lower concentrations) and vertical dense columns (at higher concentrations) are found in 0.3% due to an instability in the local concentration near the contact line [3].

Electric fields affects the conditions at which the deposition takes place (e.g. contact angle, local concentration of particles, ...). At high positive voltages, the number of particles (negatively charged) reaching the positive substrate get increased and initially forms NCD. There appear flows generated due to the capillarity rise into the previously deposited structure. These flows drag more particles towards the depositing region [4]. This results in multilayer or in a dense column which depends on the strength of the flows and on the local concentration. In the negative voltage region, multilayer and monolayer are formed at lower speeds because the particles have been repelled from the substrate. Also at negative voltages, many multilayer or monolayer form in counter substrate which frequently executes pin-depin behavior. This results in a rapid advancement of the contact line in the observed substrate. These type of behavior completely distorts the local parameters that make the system more complex and non linear.

In the full paper, we will discuss these phenomena for several concentrations, and we will provide a global picture of the mechanisms involved in vertical deposition of diluted colloids with applied electric fields.

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Nonlinear dynamics of a liquid coating on an axially oscillating cylinder

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We have investigated the dynamics of an axisymmetric thin liquid film on a horizontal circular cylinder subjected to axial harmonic oscillation. Using the methods of long-wave theory we have derived the nonlinear evolution equation describing the spatio-temporal dynamics of the film. Linear stability analysis has been carried out for both the unforced and forced systems. The results obtained for the unforced system have been compared in terms of the growth rate as a function of the disturbance wavenumber and the cutoff wavenumber with other theories based on either the general axisymmetric Orr-Sommerfeld equations and on various long-wave theories.

The nonlinear evolution equation has been investigated numerically with periodic boundary conditions. The dynamics of the film on a static cylindrical surface leads to drainage into drops and in the long-time limit the film ruptures. The main result is the prevention of film rupture as the forcing parameters such as amplitude and frequency become sufficiently large. We have shown that along the critical curve separating in the parameter space between ruptured and contiguous states of the product of the dimensionless amplitude and frequency of forcing is constant depending on the rest of problem parameters.

In the parameter domain where the continuity of the film is preserved due to forcing, a typical pattern consists of one drop in the periodic domain. We have also found a continuous transformation from the pattern consisting of several droplets in the case of the unforced system to a single droplet near the critical curve. In the forced system where the rupture of the film is prevented, the film dynamics displays a periodic behavior whose period equals to the forcing period. All flows found in the parameter range investigated here have been found to be periodic with period that equals to the frequency forcing. Finally, the variation of the film thickness norm for the forced system has been numerically studied and found to be typical for supercritical bifurcation.

We have also investigated the case of a double-frequency forcing and found a possibility to prevent rupture of the film even if the oscillation parameters of each of the forcing components correspond separately to the domain where rupture takes place.

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Complex fluid interfacial flow induced by evaporation near a contact line

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Drying of colloïdal suspension or polymer solution with a moving contact line is involved in many coating processes. For low Capillary numbers, the velocity and concentrations fields in the meniscus result of a complex coupling between hydrodynamics and drying (evaporative regime). To investigate this regime an experimental set-up has been designed where the contact line velocity and the evaporative flux can be controlled independently and varied on a large range. A capillary growth is performed in a Hele-Shaw cell partially immersed in a tank. The drying is achieved with an air flow whose temperature, humidity and velocity are carefully controlled. The contact line is moved by changing the tank level with a push/pull-syringe and is recorded through CCD camera. The contact line velocity can be varied from 100 nm/s to 2 mm/s. Thanks to the use of the capillary rise phenomenon, we are able to measure with a very good precision the contact line position during the experiment. A posteriori observations with AFM, MEB or optical profilometer give the mean thickness and the shape of the dry deposit.

We present experimental results performed with different systems (colloïdal suspensions or different polymer solutions). For the smallest Capillary numbers tested, a strong periodic patterning resulting from successive pinning and unpinning of the contact line has been observed for some systems. The corresponding pinning force has been deduced from the height deviation between the real capillary rise and that of ideal clean vertical flat plates. When increasing the Capillary numbers (but still below the dynamical wetting regime), the stick/slip fades away and it is possible to derive scaling laws for the mean thickness of the deposit. It has been shown that there exists a large domain of contact line velocity where a universal law is obtained, i.e. the mean thickness does not depend on the system under study, but only on the evaporative flux, solute concentration and contact line velocity. Moreover the mean thickness is proportional to the inverse of the contact line velocity, unlike in the dynamical wetting regime. A simple model is proposed that accounts for this behavior.



FIG. 1: Optical profilometer view of a dry colloïdal suspension - periodic patterning, wavelength $\sim 240 \mu m$, height ~ 3 to $4\mu m$

Marangoni Driven Full-Zone: Magnetic Stabilization, Transition and Energy Analysis at Low Prandtl Number

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The linear stability of the Marangoni driven full-zone is investigated for low Prandtl number fluids. A uniform magnetic field is applied along the axis of the zone to stabilize the axisymmetric base state. Primary transitions to three-dimensional, steady flows are observed up to moderate magnetic field strengths using a vorticity transport formulation and spectral collocation scheme. These instabilities are hydrodynamic in nature, as suggested by the perturbation flow and confirmed by energy analyses.

A full-zone liquid bridge with a parabolic heat flux is studied under microgravity conditions. The surface is assumed non-deformable, and the rigid end walls are taken to be the solidification temperature of the semiconductor melt, as in Houchens and Walker [1]. The thermocapillary driving force is measured by the thermocapillary Reynolds number, Re_{FZ} . Here, a steady magnetic field is applied to damp and stabilize the base flow. The strength of the magnetic field is proportional to the Hartmann number, Ha. Rather than a fourth-order biharmonic formulation used in previous work, here a vorticity transport formulation is used, resulting in second-order partial differential equations. This numerical system is better balanced, allowing for a significant increase in the range of magnetic field strengths that can be studied. Only three-dimensional perturbations are analyzed as, in the case without magnetic stabilization, it has been shown that axisymmetric perturbations are stable to very high thermocapillary Reynolds number [2]. Results are presented for Pr = 0.001, which is thought to represent the $Pr \rightarrow 0$ limit, as shown in the zero magnetic field case [1]. As expected for the low Pr limit, the instabilities observed by normal mode linear analyses are hydrodynamic rather than thermocapillary in nature, as was found for the zero magnetic field case in similar configurations [3].

The critical curves for the onset of the primary instability are shown in Fig. 1. Below the curves the axisymmetric base state is stable. Above, the flow transitions to a three-dimensional steady state with azimuthal wave number indicated by m. Below Ha = 40, antisymmetric instabilities with symmetry



Fig. 1 Critical Re_{FZ} versus *Ha* curves for anti-symmetric (open circles with solid line) and symmetric (dashes with dashed line) disturbance modes for Pr = 0.001, b = 1.



Fig. 2 Perturbation velocity vectors in the $\theta = 0$ plane superimposed on base flow stream lines - green/red indicates where the perturbation reinforces/opposes the base flow, respectively (left). Radial and axial electric current perturbations superimposed on the electric potential contours in the $\theta = \pi/12$ plane (right). All plots for Pr = 0.001, Ha = 50, m = 6, at the point of symmetric instability, $Re_{FZ,cr} = 12,925$.

opposite the base flow are critical. At Ha = 40 and above, symmetric instabilities are dominant. As the magnetic field strength increases, the primary circulation cell of the thermocapillary flow is confined to an ever thinner layer near the free surface. Weaker cells form in the interior. This is demonstrated in Fig. 2 for Ha = 50. The stabilization results in a sharp increase in $Re_{FZ,cr}$ with increasing Ha. As the primary circulation cell becomes thinner, the mode number also increases, indicating an instability pattern which repeats many times in the azimuthal direction, and is primarily confined to the first two circulation cells in the radial extent.

Energy analyses of the steady perturbation flow confirm the hydrodynamic nature of the instability mechanism. This can be observed through computation of the energy transfer from the base state to



Fig. 3 Perturbations of velocity (vectors) and temperature (contours) on the free surface for the symmetric instability at $Re_{FZ,cr} = 12,925$, for Pr = 0.001, Ha = 50, m = 6; hot perturbations: solid contours, cool perturbations: dotted contours.

the perturbation flow. Visualization of the perturbations on the free surface also verifies that the instability is not thermocapillary in nature. From Fig. 3 it is evident that the flow on the surface travels generally from cold spots to hot spots, just opposite of what would occur in the thermocapillary instability. Energy analyses indicate that the primary transfer of energy to the perturbation is from a shear flow instability near the free surface.

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Three Dimensional Flow Instabilities in Thermocapillary and Buoyant-Thermocapillary Driven Flow in an Annular Pool

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A linear stability analysis of thermocapillary and buoyant-thermocapillary flows in annular cavities over a wide range of aspect ratios is conducted for a silicone oil $(Pr=\nu/\alpha=6.7)$ under both 0G and 1G conditions. The cavity is composed of its heated outer wall (Ro=40mm), cooled inner wall (Ri=20mm), adiabatic top surface and adiabatic solid bottom and its depths d ranges from 0.8mm to 20mm (the aspect ratio $\Gamma=(Ro Ri)/d$ ranging from 1 to 25). The details of LSA is found in elsewhere [1]. In this work, we use following non-dimensional variables r=R/d, z=z/d, V=vd/v, $\tau=tv/d^2$, $P=d^2p/\rho v^2$, $\Gamma=\Delta R/d$, $\mathcal{O}=\Gamma(T^*T_c)/\Delta T$ and parameters $Re=\gamma_{T}\Delta Td^2/\mu v\Delta R$, $Gr=g\rho_{T}\Delta Td^4/v^2\Delta R$, $Bo_d=Gr/Re=g\rho_{T}d^2/\gamma_{T}$, where Re, Gr and Bo_d is the Thermocapillary-Reynolds, Grashof and Dynamic Bond numbers. Thermophysical properties of the silicone oil are assumed to be $\rho=760$ kgm⁻³, $\alpha=9.7 \times 10^{-8} \text{ m}^2 \text{s}^{-1}$, $v=6.5 \times 10^{-7} \text{ m}^2 \text{s}^{-1}$, $\gamma_{T}=7.0 \times 10^{-5} \text{ Nm}^{-1} \text{K}^{-1}$ and $\rho_{T}=1.34 \times 10^{-3} \text{ K}^{-1}$. Fig. 1 and Fig. 2 show the thermocapillary-Reynolds number for the incipience of 3-D disturbances as a function of the aspect ratio under 0G and 1G, respectively. The solid keys correspond to the critical condition and the open keys to the neutral stability limits. Some of the perturbation temperature patterns on the surface are also shown in the figures.



Fig. 1 The neutral and critical Reynolds number as a function of the aspect ratio for OG. Surface temperature pattern are shown.

Fig. 2 The neutral and critical Reynolds numbers as a function of the aspect ratio for 1G. Surface temperature patterns are shown.

These results indicate that 1) the buoyancy stabilizes the basic flow field, 2) flow mode changes at certain liquid depths (d^*) , 3) stationary instability becomes dominant in deep pools under 1G. It should be noted that degeneracy of flow modes (two different flow modes with different wave numbers become unstable at the same ΔT) occurs at depth d^* . A LSA result was reported for rectangular cavities of silicone oil (*Pr*=10) under 1G for aspect ratios ranging from Γ =1.2 to 8 [2], however, present work seems the first one for an open annular cavity corresponding to that used in the experiments of Schwabe et al. [3].

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Interfacial Instability from Equilibrium- Why are Cylinders so Interesting?

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There are at least two classes of "**equilibrium**" problems where interfaces contribute to an instability. In the first class, two immiscible phases are separated by an interface. The problems of this class are flow problems; one fluid displaces the other and a typical example is that of the instability of jets. The curvature of the interface determines local pressure variations and these translate into flows, sometimes reinforcing a displacement. The base state is an equilibrium state and cylindrical geometries give the interface two roles to play. There are two competing curvature effects: the stabilizing axial variations of the surface are accompanied by destabilizing diameter variations, viz., the diameter at a crest exceeds the diameter at a trough. These two effects alone are enough to determine the critical point of a jet. This arises from Rayleigh's work principle. The second class of problems are phase change problems, viz., material crosses the interface and solidification and electrodeposition are representative examples. These are diffusion problems. Ordinarily the second class of problems are non-equilibrium in character but in the cylindrical geometry, equilibrium can be attained, the calculations greatly simplify and cheap answers can be obtained.

Curves of growth constants versus disturbance wave numbers are a classical way to study the physics of most instability problems. We will show how and why these two classes of problems have widely different signatures on the growth curves. The beauty of the physics will thus be revealed.

Stability threshold for the flow driven by a rotating disk in a small aspect ratio open cylindrical cavity

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The three-dimensional linear stability analysis of a viscous fluid flow inside a fixed cylindrical tank in which the bottom disk rotates and the top is a free surface has been performed. The aspect ratio G (see figure 1 (a)) is such that the radius of the disk is larger than the fluid height. Such configuration exhibits interesting instability patterns and *Poncet & Chauve* [1] found the critical Reynolds and azimuthal wave number (Re_c and m) as a function of the aspect ratio G.

As a first approach, *Kahouadji & al.* [2] have numerically imposed the free surface to be flat. They have found the same critical azimuthal wave number comparing to the experiment for all aspect ratio G. The critical Reynolds numbers Re_c found numerically are close to the experiment for G ranging between 0.107 and 0.071 but differ significantly for G < 0.0701.

The hypothesis of a flat free surface is likely to be too strong for very small G. We will present the results obtained taking into account the free surface deformation with the code we have developed using curvilinear coordinates.



FIG. 1: (a): Experimental configuration, (b) and (c): are the experimental and numerical visualization of the instability pattern at the free surface for G = 0.0714 and Re = 24630.

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Nonlinear Dynamics in Bounded Thin Liquid-Vapor Layers

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We investigate the nonlinear evolution of bounded thin liquid-vapor layers, in the framework of the model recently developed in Ref. [1]. The first case considered here is that of the liquid (vapor) heated (cooled) and gravity acting toward the liquid (superheated or supercooled case), and the second is of the liquid (vapor) cooled (heated) and gravity acting toward the vapor (Rayleigh-Taylor unstable case).

In the former case, the surface deflection monotonically grows due to lateral vapor pressure fluctuation, until the crest touches the ceiling. We have found neither a rupture of the liquid layer nor a steady state for the realistic parameter values. The left panel of Fig. 1 illustrates the velocity pattern in both layers during the evolution. The liquid is pushed downward by the vapor at the depressed region of the interface, leading to the formation of the liquid crest as a consequence of the conservation of mass. This velocity field is essentially different from that of the two-layer model without phase change [2].

In the latter case, we have found nontrivial nonruptured stationary states, where the stabilizing effect of vapor pressure balances with the destabilizing gravity effect. The right panel of Fig. 1 shows one of such stationary states along with the associated velocity field. There are vortices in the liquid velocity field, which are similar to those of liquid films with the Marangoni effect [3, 4]. However, in the present system the liquid motion is induced by the vapor flow itself, which generates the shear stress along the interface in the same direction as the vapor flow. Note that the emergence of these vortices was not predicted in the study of evaporating or condensing liquid films with an infinitely deep gas layer [5].

In both cases, we have observed that the average vapor pressure and interface temperature deviate from their equilibrium values once the instability of the flat interface takes place. We show that these pressure and temperature variations arise from the combination of incompressibility of the vapor and the effect of local variation of the saturation temperature. Moreover, the thermocapillary effect emerges if the system significantly deviates from thermodynamic equilibrium, because the temperature equilibrating effect of latent heat on the interface is weakened. We also discuss the dependence of the solution on both the initial condition and the initial thickness ratio for the Rayleigh-Taylor unstable case, resulting in different evolution toward stationary states.



FIG. 1: Typical velocity fields in one spatial period in the horizontal (x) direction. The curve indicates the liquidvapor interface, below which the liquid phase is located. Here, the x-coordinate, the x-component of velocity and the vapor velocity itself have been compressed by the factor of 10^{-2} . Left panel: The superheated or supercooled case (non-stationary state); Right panel: The Rayleigh-Taylor unstable case (stationary state).

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Delayed Coalescence of Sessile Droplets with Different but Miscible Liquids

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Due to capillary forces two sessile droplets of miscible liquids will fuse when they get in contact with each other because one large droplet has a smaller interfacial energy than two smaller droplets of the same volume. Usually the droplet fusion proceeds very fast, delayed mostly by viscous forces. However, quite unexpected, it was observed recently [1] that under certain conditions, the coalescence of sessile droplets of completely miscible liquids can be delayed up to several minutes. Instead of fast coalescence, after a first contact of their three phase lines, the droplets remain separated by a thin liquid neck. Often, the drops even push each other across the substrate before they finally merge (see figure 1).



FIG. 1: Left: Experimental setup for studying coalescence of sessile drops [2]. Right: Two sessile drops of different but miscible liquids (view from the top). After a first contact of their three phase lines, they remain separated by a thin liquid neck and travel across the substrate.

For the first time this has been mentioned by Maxwell more than a century ago, although only very briefly in only one sentence [3]. In 1938, Bangham et al. [4] presented for the first time a qualitative description of the delay of the coalescence of sessile droplets of completely miscible liquids. However, they could not really explain the findings, analyzing them only in vague terms such as caused by "certain adsorption effects and molecular transport phenomena through the vapor phase". Recently it has been assumed that the coalescence is delayed by a marangoni convection through the thin film connecting the drops [1]. The convection originating from the differences in the interfacial tension of the liquid causes a dynamic pressure, which keeps the drops separated. This suggests that the effect is quite common. Meanwhile it could also be modeled in the framework of phase field formalism and lubrication approximation [2]. The delayed coalescence may be relevant for technical applications, for instance in the field of microfluidics.

We present new experimental results [5] from imaging the droplet shapes/topologies addressing the influence of the liquid properties on the coalescence behavior during the first few tenths of a second after initial contact. The experiments were conducted as follows: First, one droplet was placed on the substrate (Silicon wafer pieces with natural oxide layer) with a syringe. Then the second droplet was slowly filled through a hole in the substrate (black spot inside the upper droplets in figure 1) until it touched the first droplet. The liquids consisted of various aqueous mixtures of different non-volatile diols and carbon acids with surface tensions ranging from 33 to 68 mN/m, contact angles between 9 and 20° , and viscosities from 1 to 12 cP.

In these experiments two major coalescence modes have been observed: The "fast" mode, where the connecting bridge between the droplets grows fast and reaches a value close to the droplet height within about one second. This behavior is quite similar to droplets from identical liquids [6], although the droplets' center of gravity moves somewhat due to mass transport by marangoni convection. The other case, the "delayed" mode, is qualitatively different. Here, the bridge between the droplets remains shallow (< 20 microns, the experimental resolution) over a long time during which the droplets travel across the substrate (see figure 2) [5].



FIG. 2: Experimentally determined center to center profiles of two sessile drops coalescing fast (left) or delayed (right) [5]. The viscosities of the liquids were around 4 cP. The gray region around zero height indicates the experimental uncertainty.

Surprisingly, a remarkably sharp transition from fast to delayed coalescence is found as the difference in surface tensions $\Delta\gamma$ exceeded approximately 3 - 5 mN/m. Other parameters have also been varied systematically. It is found that the coalescence mode depends almost exclusively on the differences in the surface tensions of the two droplets. Within the investigated parameter space it is independent from droplet sizes (contact areas 6.0 to 87 mm^2), contact angles, and absolute values of the surface tensions. With respect to viscosity, around 5 - 6 cP a slight maximum for $\Delta\gamma$ is found ($\approx 4 - 5 \text{ mN/m}$), compared to 2 - 3 mN/m around 1 cP and 3 - 4 mN/m around 12 cP. Within the errors ($\pm 0.5 \text{ mN/m}$), this trend appears to be not really significant [5]. Resulting from these experimental data we present a phase diagram of the boundary between fast and delayed coalescence. It consists of two clearly distinct regimes: fast coalescence below the $\Delta\gamma$ -threshold, and delayed coalescence above.

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Delayed Coalescence of Sessile Droplets of Completely Miscible Liquids: Tracing Surface Flows with Fluorescent Microspheres

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Due to capillary forces two sessile droplets of miscible liquids will fuse as soon as they get in contact with each other. Usually the droplet fusion is delayed only by viscous forces and proceeds very fast. This is the case for droplets of the same liquid. However, quite unexpected, for sessile droplets of completely miscible liquids it was found recently [1] that the coalescence can be delayed by orders of magnitude (up to minutes). It is observed that after the first contact, the main droplet bodies remain separated. They are however in contact through a thin liquid neck through which they continuously exchange liquid.

For the first time this has been mentioned by Maxwell more than a century ago, although only very briefly in only one sentence [2]. In 1938, Bangham et al. [3] presented for the first time a qualitative description of the delay of the coalescence of sessile droplets from completely miscible liquids. However, they could not really explain the findings, analyzing them only in vague terms such as caused by "certain adsorption effects and molecular transport phenomena through the vapor phase". The experimental data show that this flow originates from the different surface tensions of the two liquids. It is assumed that the droplet separation is indirectly stabilized by marangoni convection through the thin connecting film: The flow induces a dynamic pressure; this pressure balances the capillary pressure and keeps the droplets separated [1]. Meanwhile the experimental observations could also be modeled qualitatively in the framework of phase field formalism and lubrication approximation [4]. This effect has obviously been largely overlooked/ignored up to now, even though it is quite common and may be quite relevant for technical applications, e.g. in the field of microfluidics.

Although some quantitative experimental data are meanwhile available [5], the delayed coalescence behavior it is still very poorly understood. It was observed that in the delayed mode the droplets are connected through a thin liquid bridge and that they remain in this configuration up to minutes while traveling across the substrate. The determining parameter for the coalescence mode is the difference in surface tension between the two droplets, $\Delta\gamma$. There exists a remarkably sharp transition from fast to delayed coalescence as $\Delta\gamma$ exceeds $\approx 3 - 4 \text{ mN/m}$. Yet the the origin of this sharp transition is not completely understood. In view of the importance of the marangoni flow it is crucial for a better understanding to obtain quantitative experimental data on the surface flow field of the system.



FIG. 1: Setup for tracing fluorescent microspheres during drop coalescence.

We introduce an experimental setup designed for tracing surface flows (see figure 1). Droplet 1 is placed on the substrate (Silicon wafers) from above with a syringe. The other droplet is slowly filled

through a hole in the substrate until it touches droplet 1. By this, coalescence of sessile droplets with free liquid-gas interfaces can be studied. The flow field is visualized by tracing small fluorescing polymeric spheres. The particles can be either dissolved in the bulk droplets, or they can be spread over the surfaces of the newly created droplets. The beads are illuminated with a green DPPS laser and observed with a high speed CCD camera from above. A longpass filter and a dichroic mirror reflecting in the laser wavelength range are used to enhance the contrast in the images. With this setup, individual particles of less than 1 micron diameter can be traced at high framerates.



FIG. 2: Flow of fluorescing particles during delayed coalescence of sessile droplets from completely miscible liquids with different surface tensions (view from the top, each image: eight frames oversampled, time delay between first and last frame 0.130 s).

We present for the first time quantitative experimental data on the flow fields of two sessile droplets during the delayed coalescence process. Figure 2 shows three images obtained by oversampling eight successive frames recorded with the described setup. Each oversampled image covers a time interval of 0.13 s. The resulting data provides insight to the flow velocities in the bulk and at the surfaces of the droplets. By varying the particle sizes, a threshold can be determined for different liquid combinations at which the particles are able to cross the liquid neck between the droplets. This gives an estimate for the height of the neck.

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Terrestrial Simulation of Drop Saturation by Surfactant under Microgravity Conditions

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It is well known that diffusion plays an appreciable role in mass transfer only in the case of weak gravitational convection. However, even under such conditions the use of a surfactant as a diffusing substance essentially complicates the mass transfer processes. The appearance of gas or liquid inclusions in the surfactant solution causes the development of solutocapillary motion on their surface, which may become a generator of the large-scale flows in the surrounding liquid, especially in microgravity conditions.

The paper presents the results of terrestrial simulation of such flows during saturation of the drop of weakly soluble fluid by a surfactant from its water solution forming a thin (~1 MM) horizontal layer. In our experiments, we used chlorobenzene and water as the basic fluids of the drop and the surrounding medium, respectively, and isopropyl alcohol - as a surfactant. The initial concentration C_0 of the alcohol in the solution ranged from 5 to 50%. This lent specific feature to the saturation process, manifesting itself in the fact that at concentration higher than 25% a mutual dissolution of water and chlorobenze began to increase.

Visualization of flow structures and concentration fields showed that in laboratory conditions even at maximum suppression of the gravitational convection the saturation of the surfactant is a rather complicated process specified by the initial surfactant concentration in the solution and by the degree of the solution homogeneity. In the case of initially homogeneous solution, a complicated character of mass transfer between the drop and the surrounding medium is evidently due to the small values of surfactant diffusion coefficients in basic fluids. Penetration of the surfactant into the drop leads to the formation of local inhomigeneities of the solution density at both sides of the interface and to the development of a slow three-dimensional flow of gravitational nature. An increase in the concentration gives rise to a reverse process –diffusion of chlorobenzene from the drop, which occurs simultaneously with surfactant dissolution. This leads to the inhomogeneous distribution of the surfactant over the interface and, as a result, to the development of solutocapillary motion around the drop (Fig.1). At surfactant concentration exceeding 30% the capillary flow displaces the gravitational one and becomes a dominant flow pattern both in the drop and in the layer. At $C_0 = 50\%$ the phase boundary between the drop and the solution disappears leading to formation of a three-component liquid mixture (Fig.2). Phase separation of the fluid system vanishes within first two minutes after placing the drop into the surfactant solution.

In the case of initially inhomogeneous distribution of the surfactant, generation of the surface tension gradient on the surface of the absorbing drop occurs instantaneously initiating a capillary motion. A competition between gravitational and capillary mechanisms of convection gives rise to a large- scale oscillatory flow (Fig. 3). The results of the experimental studies have been analyzed to define the dependence of the flow oscillation frequency (Fig. 4) and the lifetime of this particular mass transfer mode on the initial concentration difference in the layer.

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Fig.1. Interferograms of absorbing drop (d = 8.0 mm) surrounded by alcohol solution ($C_0 = 45\%$) (a, t = 40 minutes) and of general view of the thin horizontal layer with drop (b, t = 120 minutes)



Fig.2. Dissipation of drop interface due to the absorption of alcohol from its solution $(C_0 = 50\%)$ (*a*, *t* = 2 minutes) and fingering instability of transition zone between former drop and surrounding liquid (*b*, *t* = 11 minutes).



Fig. 3. Interferogram of absorbing drop (d = 6.0 mm) surrounded by inhomogeneous alcohol solution $(\Delta C = 30\%).$



Fig. 4. Flow oscillation *versus* time for drops with different initial concentration of isopropyl alcohol.

The mechanics of particle-accumulation structures in thermocapillary flows

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The motion of small particles suspended in a cylindrical high-Prandtl-number thermocapillary liquid bridge is studied numerically. We consider the supercritical case when the flow arises in form of an azimuthally traveling hydrothermal wave [2, 4]. We have accurately computed numerous particle trajectories and found that small, neutrally buoyant particles of finite size segregate to form particle accumulation structures (PAS). Typically, a PAS consists of a thread of particles that is wrapped around a virtual torus. The thread rotates with the same angular velocity as the hydrothermal wave. While the PAS rotates, its shape remains unchanged.

The PAS obtained numerically (fig. 1a) are qualitatively similar to those observed experimentally by [3] and [1] (fig. 1b). We suggest a physical mechanism for the formation of PAS based on a fully passive advection of the particles in the bulk of the liquid and a particle–free-surface collision process that transfers particles from one streamline to another.

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FIG. 1. (a) Axial view of a PAS with period m = 3 obtained numerically for a liquid with Pr = 4. (b) PAS in a liquid bridge of NaNO₃ [1]. The arrows indicate the direction of rotation of the structure.

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A coalescence of two close liquid drops suspended in a another liquid of the same density

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In this work, the interaction of drops in surrounding liquid has been simulated. It was shown in experiments of Stebnovskii [1] that drops suspended in a different liquid of the same density initially at rest and at small distance between them (of order of radius) approach each other slowly until they contact and merge into one larger drop.

It is well known that a coalescence of small drops to a larger one is thermodynamically preferable. Nevertheless, for drops suspended in other liquid initially at rest, the physical mechanism of attraction of the drops before following coalescence should exist. Several possible mechanisms are Brownian motion, elastic forces in the case of viscous-elastic liquids, an electrostatic interaction, and also a mechanism of concentration distribution. The last mechanism arises due to a concentration gradient of the drop substance in the surrounding liquid because of mutual solubility of liquids that is usually very small, but not zero [2]. The concentration of molecules of the drop substance in another liquid in the region between drops is greater than aside. Hence, the resulting attractive forces arise between drops. Moreover, another mechanism of material transport from a small drop to a larger one is known, namely the diffusion of molecules of one drop to another through the surrounding liquid without direct contact between drops. In the case of macroscopic drops, the Brownian motion is negligible. The elastic tensions are absent if the drops were initially suspended in the viscous-elastic liquid accurately. The electrostatic fields and temperature gradients can be eliminated by screening of the system as was done in experiments carried out by Stebnovskii [1]. In this work, the possibility of coalescence mechanism caused by the concentration distribution was demonstrated. For computer simulation of this process, the variant of lattice Boltzmann equation (LBE) method for multi-component media was used [3]. In the variant of LBE method used in this work, the possibility of phase transition has been taken into account [4]. Moreover, for simulating of mutual solubility of liquids, we introduced special repulsive forces between pseudoparticles of the different components. Figure 1 shows the results of two-dimensional computer simulations of interaction of two liquid drops of different size ($R_1 = 31.5$, $R_2 = 22.5$ lattice units, respectively) suspended in other liquid of the same density.

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Fig.1 The material transport from a small drop to a larger one with concurrent coalescence between them

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Marangoni-Driven Macroscopic Core-Shell Structure

in Cu-Sn-Bi Immiscible Alloy

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During cooling through the miscibility gap, Marangoni motion driven by the temperature and/or concentration gradient between minor phase droplets and the liquid matrix plays an important role in the immiscible alloy microstructure evolution ^[1]. The previous reports have shown that the rapid cooling of melt can produce diametrical phase separation in final structure of immiscible alloys even under earth gravitational condition. Wang et al ^[2] reported the egg-type bimetallic balls of 24Cu-16Sn-60Bi (wt %) with ~80 μ m in size, a candidate of lead-free solder ball for chip-scale package, possessing high electronic conductivity in the core and low melting point in the periphery. However, state-of-the-art electronic packaging technology is being dominated by ball grid array which adopts solder balls with about 100-760 μ m in diameter. In view of massively reported results ^[3] showing that rare earth metal could modify significantly surface properties of liquid alloys, rare earth metals are expected to enhance Marangoni motion as dopants in 24Cu-16Sn-60Bi alloy and obtain larger coreshell balls. This paper intends to reveal the effect of Marangoni motion on core-shell type structure of liquid immiscible alloy by different Ce additions, as well as by different cooling rates



Figure 1 The morphology of slow-cooled 24Cu-16Sn-60Bi sample



Figure 2 The cross-sectional morphologies of rapid-cooled 24Cu-16Sn-60Bi columnar sample with 5 mm in diameter

The homogeneous 24Cu-16Sn-60Bi alloy melt was obtained with a high-frequency induction furnace under an argon atmosphere, and then *in-situ* cooled down in quartz crucible. Severe phase separation occurred in the as-solidified sample which is the most feature of liquid immiscible alloy, as shown in Fig 1. The rapid-cooled samples of 24Cu-16Sn-60Bi alloy were prepared by casting the melt into copper mold with different diameters. Asprepared samples demonstrate a typical core-shell type structure, as shown in Fig. 2. It should be noted that as-prepared sample size is large up to \emptyset 10×50 mm, much higher than those by Wang et al ^[2]. The macroscopic structure makes feasible for investigating experimentally the evolution of core-shell type structure formed in immiscible alloys. The phase analysis by X-ray diffraction shows that the coreshell type structure consists of (Cu₃Sn, Cu₁₀Sn₃) central core and Bi periphery.

0.05 wt% Ce-doped alloy columns demonstrate the similar core-shell structure in macroscopic scale but larger central core, as shown in Fig. 3. The area fractions of Cu-Sn phase were counted by using home-made image analyzing software and the statistic shows that the area of Cu-Sn central core increased by at least 20% after Ce addition while the whole area of Cu-Sn phase including droplets does not change significantly. A magnified micrograph at the edge of the black central core was given in the inset of Fig. 3. It can be seen that the large droplets are located in the region close to the central core,

The velocity of the droplet in steady state is given by:

$$V_m \approx \frac{-2r}{3(3\mu_d + 2\mu_m)} \cdot \frac{\partial \sigma}{\partial T} \cdot \frac{\partial T}{\partial x}$$
(1)

Where *r* is the radius of the droplet, σ is interfacial tension, μ_d and μ_m are the viscosities of the droplet and matrix liquid phases, respectively; *T* is absolute temperature and *x* is the distance. It is evident that the interfacial tension between the droplet and matrix liquid plays a key role in the formation of the core-shell type structure. The interfacial tension between two immiscible liquid phases was calculated based on Kaptay model^[3] as below.

$$\sigma = \frac{6.05T_{cr} + 7T}{1.06(N_{Av})^{1/3} (\sum_{i} X_{i} V_{i})^{2/3}} \cdot \left(1 - \frac{T}{T_{cr}}\right)^{1.26}$$
(2)

Where T_{cr} is the critical temperature of the alloys, N_{AV} is the Avogadro's number, X_i and V_i are the mole fraction and the molar volume of the component i, respectively. Obviously, the interfacial tension of immiscible liquid phases is determined mainly by T_{cr} , X_i and V_i . Our calculation shows that the temperature coefficient of the interfacial tension was remarkably increased by the addition of 0.05% Ce, as depicted by Fig 4(a). Both experimental and theoretical results show that Ce addition can enhance effectively Marangoni motion of minor phase droplets in liquid matrix to form larger-sized core-shell type structure.

The systematic experiments were carried out to investigate 24Cu-16Sn-60Bi alloys doped with varied Ce amounts from 0.05 to 0.1 wt%, and cooled with different cooling rates through adjusting copper moulds diameters. All of the as-preapred samples present the core-shell type structure. The velocities of Marangoni and Stokes motions were calculated and the comparison shows that gravity effect is limited during the formation of core-shell structure, as seen in Fig. 4 (b).



Figure 3 The cross-sectional morphologies of rapid-cooled 0.05 wt% doped 24Cu-16Sn-60Bi alloy with 5 mm in diameter



Figure 4 (a) Temperature dependence of interfacial tension of the 24Cu-16Sn-60Bi alloy with 0.05 wt% Ce addition and (b) Calculated V_m and V_s versus the droplet radius for the 24Cu-16Sn-60Bi alloy at given temperature 1020K

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Thermocapillary-Buoyancy Convection in Annular Two-layer System with Radial Temperature Gradient

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Czochralski method is widely used in producing crystals. Due to the significant effects in reducing the evaporation and avoiding crystallographic defects, the liquid-encapsulated crystal (LEC) growth has attracted a great attention. In order to understand the characteristics of thermocapillary-buoyancy convection in the annular two-layer system, a set of two-dimensional numerical simulations is carried out using the finite-volume method. The annular two-layer system is differentially heated with the hot outer wall and the cold inner wall, which is formed with 5cSt silicone oil and HT-70. The fluid-fluid interface and the free surface are assumed to be flat and non-deformable, the fluids immiscible, the bottom surface heat-insulated. Combined thermocapillary and buoyancy forces induce a basic flow when a temperature difference is applied.

The driven forces at the free surface and the fluid-fluid interface are in the same direction. Therefore, the flow pattern consists of a pair of counter-rotating convective cells in the upper layer, and a counter-clockwise rotating convective cell in the lower layer. Marangoni (*Ma*) number plays a dominant role in controlling thermocapillary-buoyant convection under gravity conditions. For sufficient small values of *Ma* number, the flow is steady. As shown in Fig.1, with the increase of *Ma* number, the flow is enhanced and the maximum value of the non-dimensional stream function increases. When *Ma* number exceeds a critical value, the flow transits into unsteady flow (see Fig. 2). Since the thermocapillary effect plays the dominant role, the additional cells appear mainly near the interface at the cold lateral wall where the temperature gradient is the biggest. During the oscillatory process, the thermal wave travels from left to right side (see Fig. 3). This thermal wave seems to be generated near the interface at the cold lateral wall, and most of the region in the right-hand part of the pool, remains pretty calm. We determined the critical *Ma* numbers using the linear extrapolation method. For the case of Γ =0.2 and ε =0.1, the critical *Ma* number is 2.888×10⁶. And the critical *Ma* numbers decrease with the increase of the aspect ratio. It means that it is possible to control the convection in the two-layer system by choosing a suitable dimension.



Fig. 1 Streamlines with different *Ma* numbers at Γ =0.2, ε =0.1.(a) *Ma*=8.0×10⁵, ψ (+)=0.107, ψ (-)=-0.552, $\delta \psi$ =0.037 5, (b) *Ma*=2.8×10⁶, ψ (+)=2.006, ψ (-)=-2.423, $\delta \psi$ =0.3



Fig. 2 Snapshots of streamlines for the thermocapillarybuoyant oscillatory flow at Γ =0.2, ε =0.1 and Ma=4.0×10⁶, $\delta\psi$ =0.225



Fig. 3 Snapshots of temperature fluctuations at $Ma = 4.0 \times 10^6$ ($\Gamma = 0.2$, $\varepsilon = 0.1$) within one oscillation period.
Interaction of Evaporation and Marangoni Effect at a vapor-liquid interface

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Evaporation and convection are the naturel process in nature and engineering applications, such as in the water cycle and many practical applications. In microgravity environment and small-scale thermal process where the Marangoni effect become dominant over buoyangcy effects, the evaporation and thermocapillary convection at the phase-change interface generally interact each other, for example in the heat pipe in space and the evaporating liquid thin film. While large amount of work has been performed to investigate evaporation phenomenon, only a few papers address Marangoni flow in the evaporating liquid. In this paper, we will focus on the combined effects of the Marangoni convection and evaporation in a pure liquid layer with a top free surface in contact with its own vapor, especially in microgravity condition.

We consider a physical model consisting of a rectangular cavity containing an evaporating liquid layer in contact with its own vapor. The left and right sides of the cavity are vertical rigid, isothermal walls. The deformation of the free surface is neglected and the surface tension at the interface is considered to be a linear function of temperature. The linearized Hertz-Knudsen equation is introduced to portray the relation between interfacial temperature and the evaporating mass flux under interfacial non-equilibrium conditions and the dimensionless Hertz-Knudsen equation has the form of:

$$j = Bi_{ev} \theta \tag{1}$$

The parameter Bi_{ev} called the evaporation Biot number is defined as:

$$Bi_{ev} = \frac{\alpha D\rho_v L^2}{\lambda} \sqrt{\frac{M}{2\pi R T_s^3}}$$
(2)

which measures the degree of non-equilibrium at the evaporating interface. $Bi_{ev} = \infty$ corresponds to the quasi-equilibrium limit, where the interfacial temperature is constant and equal to the saturation value, $\theta = 0$. $Bi_{ev} = 0$ corresponds to the non-volatile case in which evaporative mass flux is 0. We introduce local mass flux j(x, A) and integral mass flux $J = \int_0^1 j(x, A) dx$ to characterize the mass transfer at the vapor-liquid interface. The computing program developed for simulating the two-dimensional, time-dependent Navier-Stokes equations and energy equation by a seconder order accurate projection method. The properties of water with its own vapor are used in this numerical investigation.

The influence of evaporation Biot number Bi_{ev} and Marangoni number Ma on the interfacial mass and heat transfer represents the coupling of evaporation and thermocapillary convection (see in Fig.1), and three different regimes of the coupling mechanisms are found in this model.



FIG. 1: Interfacial local evaporating flux (j(x)) of the evaporating liquid layer for different Marangoni numbers (Ma) and evaporation Biot number (Bi_{ev}) .

Figure 2 reflects the contribution of thermocapillary convection to mass transfer (mass flux J) responding from steady flows at various evaporation Biot numbers. It demonstrates that greater evaporation mass flux is acquired with higher Marangoni numbers at all levels of evaporation Biot numbers.



FIG. 2: Integral mass flux at the evaporation interface in the cavity (J) versus evaporation Biot number (Bi_{ev}) .

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Rayleigh-Marangoni Instabilities in a Two-Fluid System under High-Frequency Vibrations

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Oscillatory motion in liquid layers has been known as an effective way to enhancing the performance of many applications. The related problems of oscillatory convection in a two-layer system has received significant attention because of its technological importance. In contrast to Rayleigh-Bénard problem in a one-layer system, the stability problem in a two-layer system is not self-adjoint, so that oscillatory instability is possible in general. Oscillatory instability also can be achieved by either vibration of the surface or pulsation in the fluid.

In absence of vibration, there are two different cases where the oscillatory instability has been predicted [1]. The first case is the densities of fluids are close to each other, so that the deviations of the densities with respect to temperature are comparable with the density difference of the reference state, the oscillation may be self-sustained as interfacial waves. This kind of oscillation was studied in [2– 4]. The second case is that the oscillations is driven by hydrodynamic and thermal interaction between the two layers at the interface. Rasenat et al. [5] showed that an oscillatory Rayleigh convection could develop involving no interfacial deformation.

In many situation, both surface tension and buoyancy play important roles in driving convection in liquid layers. Both instability analysis and experimental observation found two possible convective modes: thermal coupling mode and mechanical coupling mode. Thermal coupling mode is characterized by the rolls in each layers rotating in opposite directions, and mechanical coupling mode is characterized by rolls rotating in the same direction. Liu et al. [6, 7] investigated the oscillatory Rayleigh-Marangoni-Bénard convection in a real two-layer system consisting of silicon oil 10cS and fluorinert FC70 liquids. It is reported that oscillatory Rayleigh-Marangoni-Bénard convection may occur in a narrow gap $1.461 \le h \le 1.564$ in the system with the total depth of d = 12mm.

Vibrations as well as gravitation and surface tension, are known to be an effective way of affecting the convective behaviors of fluid systems. The problem of the effect of vibrations on the onset of convection in a two-layer system was investigated extensively in previous works. Most of existing works is devoted to the case of pure Rayleigh convection or Marangoni convection under high-frequency vibration. However, little research has been devoted to the influence of vibration on the narrow gap between thermal coupling mode and mechanical coupling mode. Motivated by previous works, we develop a tentative study on the mechanism of oscillatory Rayleigh-Marangoni convection under high-frequency vibrations.

We consider two dimensional thermal convection under high frequent vibration in a two layer system. The physical model consists of a fluid of depth d_1 underlying another fluid of depth d_2 . The combined layers are infinite in the horizontal directions. The upper and lower boundaries are rigid walls maintained at constant temperatures. The interface between the lower and upper fluid is assumed to be a non-deformable free surface, where the surface tension is assumed to be a linear function of temperature. The fluid is considered to satisfy the Boussinesq approximation, i.e. the physical properties are independent of temperature, except density which decreases with local temperature.

In figure 1(a), the critical Rayleigh number versus the depth ratios are presented. It is shown that with the increase of the vibration Rayleigh number Ra_v the critical Rayleigh number increases. We also find it is interesting that the the oscillatory gap significantly changes with Ra_v .

In figure 1(b-d) we present the marginal curves of the Rayleigh number versus the wavenumber at

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FIG. 1: (a): The critical Rayleigh number versus different depth ratio for selected vibration Rayleigh numbers. The parameters for the system d = 6mm, Bo = 15.35, vibration angel $\alpha = \pi/2$. The marginal curve of Rayleigh number versus the wavenumber for different vibration Rayleigh numbers. (b), (c) and (d): The other parameters for the system (b) d = 6mm, $Ra_v = 0$, Bo = 15.35, the depth ratio h = 1.0, (c) d = 6mm, $Ra_v = 0$, Bo = 15.35, the depth ratio h = 3.

several typical depth ratios. In these figures, the marginal curves display multi-modal structures. The influences of the vibration on different mode is complicated. In the present study, we will study the effects of vibrations on the coupling mechanism of different modes in detail.

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Experimental and numerical investigation on Marangoni effect induced by mass transfer during drop formation^{*}

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The liquid-liquid extraction is a basic and important unit operation for process industry. Previous studies indicate that drop formation plays an important role in solvent extraction because 10-50% of total mass transfer occurs in this stage^[1]. However, no sufficient attention was concentrated at the interfacial instability (Marangoni effect) induced by mass transfer during drop formation.

In this work, experiments were carried out to investigate the effects of drop formation time, solute concentration and Marangoni convection on the mass transfer coefficient during drop formation. The experimental results without Marangoni effect were found to agree well with the empirical predictions of drop surface area and mass transfer rate. When the concentration driving force reached the critical value, the Marangoni effect would be induced and enhanced the mass transfer during drop formation.

Mass transfer from a drop with constant interfacial tension was formulated mathematically and simulated numerically by solving the motion and mass transfer equations of a growing drop coupled with a level set equation for capturing the interface. The numerical predictions of drop formation time, drop shape and extraction fraction of mass transfer are in good accord with the experimental measurements. The simulation results illustrate the mechanism of the complicated phenomenon, which is difficult to be observed in experimental measurements.

Keywords: Marangoni convection, drop formation, mass transfer, level set method

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Fig.1 Comparison of experimental and predicted drop formation time and shapes during drop formation (*Q*=420 µL/min)



Fig.2 Simulation of Marangoni effect and concentration distribution during drop formation $(Q=420 \ \mu L/min, t=0.8807\sim 1.009 \ s)$

Stability of thermal boundary layer in the presence of vibrations

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We investigate the stability of thermal waves propagating in a fluid during a sudden change of temperature performed in the presence of harmonic tangential vibrations. Two different configurations are considered: 1) semi-infinite space filled with a fluid and bounded by a flat wall oscillating in its plane; 2) long cylinder of square cross-section subjected to vibrations perpendicular to the cylinder axis.

In the first case, a state with the fluid velocity parallel to the wall is possible. In this situation, the fluid motion does not influence the heat transfer and the propagation of thermal waves is described by classical relations. The stability of this state is studied by the Floquet method, under the assumption of a "frozen" temperature profile. Perturbations periodical in the direction parallel to the wall are considered. The Floquet indices for these perturbations, which depend on the wave number and the specific parameters of the problem, are calculated numerically by a finite difference method. For the numerical modeling, the condition of perturbation vanishing at infinity is imposed at a finite distance that is large in comparison with the thermal boundary layer thickness. The calculations show that the instability develops when the thickness of the thermal boundary layer attains a critical value depending on the Prandtl number and the vibration parameters. The wavelength of the most dangerous perturbations is several times larger than the thermal boundary layer thickness. For high vibration frequencies, that is, high enough such that the viscous boundary layer thickness is small in comparison with the thermal boundary layer thickness, the averaging method is applied. It permits to reduce the problem for small perturbations to a boundary-value problem for the system of ordinary differential equations. It is shown that for a common parameter range both methods give consistent results. For the opposite limit case of low frequencies, a quasi-static approach is used where time-dependent inertia forces are replaced by constant gravity force parallel to the wall.

In the case of a closed cavity, the thermal front interacts with the flow at any values of the parameters and, strictly speaking, there is no instability threshold. However, at a time-scale when the thermal boundary layer thickness is still small as compared to the cavity size, the central part of the front is nearly flat. One thus expects sharp changes of the front shape for parameter values close to those at which the instability of an infinite flat front occurs. The behavior of the system for this configuration is studied by the finite difference method with a non-uniform mesh, finer in the boundary layer areas. The calculations show that, when the thermal boundary layer thickness attains the critical value for the infinite flat front, strong deflection of the thermal domain boundaries parallel to the vibrations occurs. Fingers directed from the wall to the bulk form, in agreement with the results of direct numerical simulation for near-critical fluid [1].

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Horizontal vibration effect on Marangoni instability in a twolayer system of immiscible fluids with deformable interface

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The paper deals with the vibration effect on Marangoni instability in a system of two superposed horizontal layers of immiscible incompressible fluids of different densities (more dense fluid is located below). External boundaries are rigid and perfectly conductive. The system is subjected to the gravity force, horizontal vibrations of finite frequency and amplitude and vertical temperature gradient. The study is performed taking into account the interface deformations. The thermal expansion of fluids is neglected.

The results of calculations for two different two-layer systems in the absence of vibrations are presented in Figs. 1a, b. For the first system, considered earlier in [1], the dynamic viscosity of lower fluid is twice larger than that of the upper one. In this case for the heating from above the longwave monotonic perturbations are most dangerous (curve 1). This instability is substantially related to the interface deformations: with the increase of gravity force characterized by the Galilei number, the critical Marangoni number grows according to the linear law. There is also oscillatory instability mode with finite wave length (curve 2), however it is less dangerous than the long wave monotonic mode. For the heating from below there are monotonic growing perturbations with finite wave length (curve 3), for this instability mode the critical Marangoni number just weakly depends on the Galilei number, i.e. the usual Pearson instability mechanism, do not related to the interface deformations, is realized. The second two-layer system is Galden fluid - silicone oil with the dynamic viscosity of lower fluid being ten times smaller than that of the upper one. In this case for the heating from below the oscillatory finite-wave-length perturbations (curve 1) are most dangerous. There are also long wave monotonic growing perturbations related to the interface deformations (curve 2) however they are less dangerous. For the heating from above the Pearson monotonic instability do not related to the interface deformations is realized (curve 3); the wave numbers of critical perturbations are finite.



Fig.1. Critical Marangoni number Ma_c versus wave number k for the first (a) and second (b) twolayer systems in the absence of vibrations

In the presence of vibrations the problem allows the solution with flat horizontal interface where only horizontal components of fluid velocities are non-zero; these velocity components depend on vertical coordinate and time. The linear stability of this state to plane perturbations periodical in the direction of vibrations is studied. The base state is found analytically and the stability problem is solved numerically. The results of calculations for the first system in the presence of vibrations are shown in Figs. 2a, b. For the case of heating from above significant destabilization of the long wave deformational instability mode is found (Fig. 2a); the oscillatory instability mode is also destabilized but this effect is weaker, so, as in the absence of vibrations, this mode is less dangerous. For the heating from below the vibrations stabilize the Pearson instability mode (Fig. 2b).



Fig.2. Critical Marangoni number Ma_c versus wave number k for the first system in the cases of heating from above (a) and from below (b): 1 – in the absence of vibrations, 2 – in the presence of vibrations with the dimensionless amplitude A = 0.5 and the dimensionless frequency $\Omega = 100$.

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Thermo- and soluto-capillary-induced flows in FZ crystal growth

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The Floating Zone is a promising method for growing of high-quality semiconductor monocrystals. A great amount of works is dedicated to this problem (see for example [1-4]). This paper deals with the numerical investigation of the convective flows in a liquid bridge, maintained between the melting end of a feed rod and the solidifying end of a crystal, pulled through the ring heater, in the floating zone process. The surface tension is assumed to be dependent both on the temperature and on the solute concentration. The study is carried out for zero gravity conditions. The free surface deformations and the curvature of the phase change surfaces are neglected.

The first part of the paper concerns axisymmetric steady flows and their linear stability to threedimensional perturbations, periodical in azimuthal direction. Numerical modeling is performed by finite difference method for the parameters, which correspond to the floating zone growth of Si:P alloyed crystal. The calculations show, that the evolution of convective flow with the variation of thermal Marangoni number Ma_T at fixed value of the solutal Marangoni number Ma_c is accompanied by the hysteresis phenomena, which is related to the existence of two stable steady regimes in certain parameter range. One of these regimes is thermocapillary dominating, it corresponds to the two-vortex flow, and the other is solutocapillary dominating, it corresponds to the single-vortex flow. This phenomena are illustrated in Fig. 1a.



(a) Maximal values of stream function versus Ma_T for $Ma_C = 2000$, pulling velocity $V_g = 0.1: 1$ - solutocapillary mode, 2 - thermocapillary mode

(b) Stability map for $V_g = 0.1$: bs - bistability domain, 1 - instability domain to perturbations with k = 1, 2 - instability domain to perturbations with k = 2

Fig. 1 Axisymmetric steady solutions and their stability

For the analysis of the linear stability of axisymmetric steady regimes the exponential dependence of the perturbations on time is assumed. The discretization of the equations for perturbations by finite difference method leads to the generalized eigen-value problem. Numerical solution of this problem allows to determine the boundaries of the stability of axisymmetric steady regimes to threedimensional perturbations with different azimuthal numbers. Stability maps in the parameter plane thermal Marangoni number - solutal Marangoni number are obtained for different values of crystallization velocity and aspect ratio. An example is shown in Fig. 1b.

In the second part of the paper the numerical modeling of non-stationary three-dimensional flows is presented. The governing equations are discretized by finite differences method on the uniform in the azimuthal direction and non-uniform in the axial and radial directions grid. All spatial derivatives are approximated by second-order differencies. Fraction step method is used for the study of temporal evolution of the system. On the symmetry axis rectangular coordinate system is used instead of cylindrical in order to avoid the singularity. The discretized Fourier transformation is utilized in azimuthal direction for solving the Poisson equation for the pressure. The resulting systems of linear equations with banded matrices are solved using standard subroutines of the LINPACK package. Both the evolution to stationary axisymmetric regimes and the development of non-stationary three-dimensional flow modes are observed. An example of non-symmetrical flow is presented on the Fig. 2.



Fig. 2 Flow and concentration fields for $V_g = 0.00412$, $Ma_T = 16$, $Ma_C = 5000$ in the plane $\varphi = 0.180^\circ$ (a, b, c) and in the plane z = 1 (d, e, f)

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Bénard instabilities of a binary liquid layer evaporating into an inert gas: stability of quasi-stationary and time-dependent reference profiles

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This study treats an evaporating horizontal binary-liquid layer in contact with air. In the present work, the liquid is an aqueous solution of ethanol. Due to evaporation, the ethanol mass fraction changes and, besides, a cooling occurs at the liquid-gas interface. This can trigger Rayleigh-Marangoni instabilities (both solutal and thermal) in the system, which are the subject of our work. Several steps were taken in order to analyze the stability of evaporation-induced horizontally uniform reference profiles. First we have analyzed the stability of a quasi-stationary reference profile for which the mass fraction of ethanol is assumed to be fixed at the bottom of the liquid. This system appeared to be highly unstable, with critical liquid thicknesses between about 10 nm - 1 μ m. Comparing the Marangoni, Rayleigh, thermal and solutal effects with one another, we have shown that the Marangoni effect is much more important than the Rayleigh effect (which is not surprising in view of such small critical liquid thicknesses) and that the solutal effect is also by far more important than the thermal effect. It is thus concluded that the instability mechanism is primarily of the solutal Marangoni type. Given the extremely small critical liquid layer thicknesses obtained in our analysis, we deduce that an initially well mixed binary liquid, with a realistic thickness of, say, 1 mm, should become unstable very shortly after its exposure to air, much before the transient diffusional boundary layers developing from the free surface have reached the bottom of the liquid. Thus, it is of high interest to study the instability with such a time dependent reference diffusive solution. More precisely, we have considered a situation when the temperature and ethanol mass fraction are initially uniform throughout the liquid and the upper gas phase. Then, due to evaporation, temperature and mass fraction gradients progressively appear close to the interface and can induce instabilities after some time. Moreover, this transient character implies a separation of spatial scales which is not accounted for in the framework of the quasi-stationary model discussed above, and thus further motivates such a study: first of all this concerns the thickness of the developing diffusive layers versus the thickness of the entire liquid layer. Furthermore, due to a small Lewis number, the transient thermal layer will penetrate much faster into the liquid than the mass fraction boundary layer, thus increasing the chances for the thermal Marangoni and Rayleigh effects to get more noticeable versus the solutal ones. The competition between the Rayleigh and Marangoni effects could also be different given such a separation of scales. As a first step in a detailed study of the stability of the time dependent diffusive layers just introduced, an intermediate qualitative model was considered, based on a "broken" reference diffusive profile. Such an approximate profile for the boundary layer consists of a linear mass fraction profile in the diffusive layer, having a thickness d_l that evolves with time, under which a semi-infinite layer with fixed concentration is assumed. From a mathematical point of view, this model resembles the stationary model discussed above, except for the boundary conditions imposed at the bottom of the diffusive boundary layer. Fig. 1 shows an example of the solutal Marangoni number Ms^* as a function of the wavenumber k for $H = 1 + d_g/d_l = 2$, 11 and 101 (d_g is the gas layer thickness above the binary liquid). It can be seen that the form of the curves is not of the standard "Pearson" type, the former having their minimum close to $k \to 0$ instead of around $k \approx 2$ (k is non-dimensionalized with respect to d_l). Note that Ms^* is defined as $Ms^* = \gamma_C \Delta c_{ref} d_l / D_l \mu_l$, where $\gamma_C = -\partial \sigma / \partial c$ (σ is the surface tension, c is the ethanol mass fraction), Δc_{ref} is the ethanol mass fraction difference between the bulk and the interface in the

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reference profile, D_l and μ_l are the diffusion coefficient and the dynamic viscosity in the liquid.



FIG. 1: The solutal Marangoni number Ms^* as a function of the wavenumber k for H = 2, H = 11 and H = 101 within the "broken-profile" approach

In a more realistic model, we consider an actual time-evolution of the concentration boundary layer in the liquid. Yet the quasi-stationary assumption is retained for the temperature and the gas phase. Indeed, the time scale for ethanol diffusion in the liquid is much slower than the thermal time scale in the liquid and in the gas $(D_l << \kappa_l, \kappa_g)$ and the diffusion time scales in the gas $(D_l << D_g)$. Local equilibrium is assumed at the free flat surface. Air absorption in the liquid is neglected. The temperature of the environment is fixed at $T_b = 300K$. At a distance d_g of the surface, it is assumed that no ethanol vapor is present. The preliminary results of this model are presented in Fig. 2.

The right part of Fig. 2 shows an example for H = 11 of the mass fraction profiles of ethanol in the liquid for the dimensionless times t (from right to left): 0.00079, 0.0016, 0.0024, 0.0039, 0.0071, 0.016, 0.028, 0.039, 0.079, 0.16, 0.39, 0.79, 1.18 and 1.57. The time is non-dimensionalized using the diffusive characteristic time d_l^2/D_l . The left part of Fig. 2 shows an example of one of the results of the stability analysis, presenting the time threshold of instability as a function of the liquid layer thickness for H = 11. It can be seen that for larger thicknesses, less time is needed for the system to become unstable. It can also be seen that the curve is divided in two parts: a solid line and a dotted line. The solid line stops at a liquid thickness of about 150 nm and a time of 0.39. This means that at liquid thicknesses below 150 nm the system is always stable for this configuration. This point corresponds approximately with the mass fraction profile (in the right part of Fig. 2) reaching the bottom of the system. The dotted line represents the time after which the instability would theoretically stop if the reference profiles were not disturbed by the instability.



FIG. 2: Left: the time t threshold of instability as a function of the liquid thickness d_l for H = 11. Right: mass fraction profiles in the liquid as a function of the z-coordinate for various instants of time t

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Convective instabilities in films of binary mixtures

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Thin polymer films are often used in advanced technological applications either as homogeneous coatings or as structured functional layers. Their stability and potential use is mostly determined by the wettability properties of the substrate and is well understood for single component liquids. However, in many relevant applications the film consists of a binary mixture such as a polymer blend. For such systems the dynamics of the decomposition within the film and of the dewetting of the film itself may couple. This allows for new pathways of structuring like decomposition induced dewetting [1].

We present a model for thin films of binary mixtures, such as polymer blends, with free surfaces that allows the study of the coupling between profile evolution and decomposition. The model is based on model-H [2] describing the coupled transport of concentration (convective Cahn-Hilliard equation) and momentum (Navier-Stokes-Korteweg equations) fields supplemented by boundary conditions at the substrate and the free surface.

After determining homogeneous and vertically stratified base states of free surface films of polymer mixtures we analyse their linear stability with respect to lateral perturbations [3]. For purely diffusive transport, an increase in film thickness either exponentially decreases the lateral instability or entirely stabilizes the film. The inclusion of convective transport leads to a further destabilization as compared to the purely diffusive case. In some cases the inclusion of convective transport and the related widening of the range of available film configurations (it is then able to change its surface profile) change the stability behavior qualitatively [4]. We show two dominant driving mechanisms for the convective motion in binary mixtures with diffusue interface: Marangoni driving for energetic biased surfaces, and Korteweg driving for neutral surfaces.

We furthermore present results regarding the dependence of the instability on several other parameters, namely, the Reynolds number, the Surface tension number and the ratio of the typical velocities of convective and diffusive transport.

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Effect of Ambient Gas Flow on the Onset of Oscillatory Marangoni Convection in Liquid Bridge

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Marangoni convection in a liquid bridge suspended between coaxial disks heated differentially exhibits an onset of oscillation at a critical temperature difference between the disks. This critical value, ΔT_{cr} , is dependent on the Prandtl number of the liquid, the aspect ratio of the liquid bridge (length-to-diameter ratio, *AR*) and the volume ratio of the liquid bridge (liquid volume-to-gap volume ratio, *VR*). Some recent experiments [1][2] have revealed a striking sensitivity of ΔT_{cr} on the ambient gas temperature and on the ambient gas flow. The present study aims at clarifying this sensitivity in a liquid bridge of silicone oil. A particular attention is paid to the dependency of the sensitivity on *VR* because this seemingly complex phenomenon can be explained by a simple picture if it takes the effect of *VR* into account.

Figure 1 shows the present experimental setup, where a liquid bridge is suspended between coaxial rods 5mm in diameter. The upper heating rod is made of sapphire that is transparent for flow observation through the rod, while the lower cooling rod is made of aluminum. They are placed in the space inside an acrylic pipe. It plays as an external shield (ES) with providing an annular gap between the inner surface of the pipe, 25mm in diameter, and the outer surface of the rods. The width of the annular gap is therefore 10mm. The flow is given into this annular gap in the direction either vertically upward or downward with a range of velocity. This ambient gas flow is driven with a vacuum pump and the flow rate is measured with a mass flow meter. The temperature of the ambient gas flow is measured with a fine thermocouple inserted into the annular gap. The silicone oil (5cSt, Shin-Etsu Chemical Co., Ltd) is used as working fluid and *AR* of the present liquid bridge is fixed to 0.5.

The effect of the ambient gas flow is studied for vertically upward flow ($U_a>0$) and downward flow ($U_a<0$) in the annular gap in ES. The mean flow velocities given are -20, -10, 0, +10 and +20mm/s. These values are chosen because they have negligible mechanical effect on the liquid bridge while they affect ΔT_{cr} remarkably. Note that the surface velocity due to Marangoni effect is about 10mm/s for the present conditions. ΔT_{cr} is detected by means of flow visualization using tracer particles in the liquid. The azimuthal mode of oscillation is also determined from the flow visualization.

Figure 2 shows the critical Marangoni number, Ma_{cr} , plotted as a function of VR. Here, $Ma_{cr} = |\sigma_T| \Delta T_c \cdot H/(\mu \cdot \alpha)$, where σ_T , μ and α are the temperature coefficient of surface tension, the dynamic viscosity and the thermal diffusivity of the fluid used, respectively. The figure includes the literature data reported by Chen et al. [3] and Wang et al. [1]. The present data shown here are those taken in ES in the absence of the ambient gas flow (i.e., $U_a=0$). All results show that Ma_{cr} becomes maximum at VR=0.75-0.95, indicating that the Marangoni convection is most stabilized in such a slightly slender liquid bridge.

Figure 3 presents the measured dependency of Ma_{cr} -VR relation on the ambient gas flow for U_a =-20, 0 and +20mm/s. The results are similar to each other in that they show a maximum Ma_{cr} at a certain VR, i.e., VR=1.0, 0.95 and 0.85 for U_a =-20, 0 and +20mm/s, respectively. It may be said that the effect of ambient gas flow appears in the peak position (VR^* , hereafter) of the Ma_{cr} -VR relation. As indicated in the figure, the azimuthal mode of oscillation is one (m=1) for VR< VR^* while it is two (m=2) for VR> VR^* . The flow visualization reveals that the m=1 mode corresponds to a pulsating oscillation of the flow field in a certain radial direction as depicted in the figure while the m=2 mode corresponds to a pulsating oscillation of the flow field in two orthogonal radial directions. These features of the oscillation remain unchanged by the change of the magnitude of the ambient gas flow, at least within the range of the present experiments. Such a simple picture might not be revealed if the experiments are conducted only for some constant VRs, as usually done.





Fig. 3 The dependency of Ma_{cr} -VR relation on the ambient gas flow (U_a =-20, 0, and +20mm/s)

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Flow visualization of Marangoni Convection in Low Pr Liquid Bridge

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One of still open problems concerning the thermocapillary flow is to reveal the transition of flow pattern and the mechanism of onset of oscillatory flow. In low Prandtl number fluid, few experimental works performed because of its difficulties which were relatively high temperature, necessity of anti-oxidation, measurement of very small temperature change, and so on. By overcoming those difficulties, we detected the transition behavior from axisymmetry to asymmetry steady flow and to oscillatory flow by the very fine temperature measurement [1, 2]. However, the flow field has never been observed directly since almost of low Prandtl number fluid (e.g. molten metals and semiconductors) were opaque. So, visualization techniques using a visible light could not used. Therefore, in order to obtain the internal flow pattern directly, the visualization technique using an ultrasonic wave has been developed. We introduced the concept of visualization technique, the constitution of system, and the result of the verification test to check the capabilities. The target for flow visualization is a thermocapillary convection of molten tin in liquid column.

The concept of visualization is quite conventional idea which is particle tracking technique. The three-dimensional positions of particles mixed into the working fluid are detected every moment. Therefore, the flow pattern can obtain by tracking the tracer positions. Nevertheless, the important issue of the system is how the particle positions can be searched in an opaque liquid. We employed the ultrasonic wave and spherical reflector as a tracer particle. The pulse-like ultrasonic wave was radiated to the working fluid and the echo from the tracers was sensed by eight transducers. As a result, the particle positions are made clear by a three-dimensional synthetic aperture method. Accordingly, the internal flow pattern can be visualized from dynamic position change of the tracers. The tracer particle with multi-layer structure, which diameter is from 100 to 500 μ m, was adopted. Figure 1 shows the typical results of visualization in the actual configuration, which is voxel expression after the aperture synthesis photographic processing. At the upper left cell is X-Y plane viewing angle of molten tin liquid column. At the upper right cell is Y-Z plane viewing angle, and at the lower left cell is X-Z plane viewing angle. This result demonstrates us to be able to visualize the internal flow filed in low Prandtl number liquid column.



Fig. 1 Detection of tracer position in actual liquid bridge Configuration

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Onset of Oscillation of Marangoni Convection in Large Aspect-Ratio Liquid Bridges

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Marangoni Experiment In Space (MEIS) has been conducted in the Fluid Physics Experiment Facility (FPEF) on the International Space Station in 2008 and 2009. The critical temperature difference (ΔT_c) is measured for Marangoni flow in a liquid bridge of silicone oil (5cSt) suspended between differentially heated disks. The disk diameter, *D*, is 30mm and the aspect ratio, Ar=H/D, where *H* is the length of the liquid bridge, is varied from 0.1 to 2.0. The critical Marangoni number, Ma_c , is defined as $Ma_c = |\sigma_T| \Delta T_c \cdot H/(\mu \cdot \alpha)$, where σ_T , μ and α are the temperature coefficient of surface tension, the dynamic viscosity and the thermal diffusivity, respectively. For comparison, a terrestrial experiment has been conducted with a small liquid bridge (2cSt) formed between 2mm-diameter disks. The aspect ratio is varied from 0.2 to 1.1. This small diameter can suppress the natural convection inside the liquid bridge and the gravitational deformation of the gas-liquid interface.

In MEIS, ΔT_c is determined from the stepwise increase of the hot disk temperature, where a typical temperature increase is 0.1K for Ar=2.0. More details of MEIS will be presented elsewhere. In terrestrial experiment, the hot disk temperature is raised slowly, where the heating rate is 1K/min. The onset of oscillation is detected from the dynamic deformation of the gas-liquid interface and also from the surface temperature visualized with an IR imager. This visualization of the surface temperature enables the mode numbers and the oscillatory structures to be determined.

Figure 1 shows Ma_c plotted as a function of Ar, where the results of the sounding rocket experiment (Schwabe 2005 [1]), the terrestrial experiment (Kawamura 2008 [2]) and the linear stability analysis (Ermakov 2009 [3]). The MEIS results indicate the presence of two local maxima at Ar=0.87 and 1.25. The first maximum is seen in the present and Kawamura's terrestrial results. Qualitative agreement is seen in the MEIS results and Ermakov's result. Figure 2 shows the dimensionless oscillation frequency plotted as a function of Ar as proposed by Preisser et al. [4]. An abrupt decrease of the oscillation frequency is seen at around Ar=1.0. The results presented here suggest the presence of bifurcation of instability at around Ar=1.0. More details of this bifurcation will be presented in the presentation.



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Accumulation of rigid particles in non-isothermal liquid bridge

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Effect of accumulation of small rigid tracers in thermocapillary flow in liquid bridges (called particles accumulation structures, or PAS [1,2]) was experimentally discovered but still needs a good theoretical model. This very interesting phenomenon occurs in far supercritical oscillatory flow regimes both under gravity and weightlessness. Usually the imposed temperature difference ΔT should be high enough, about twice larger than the critical value.

We report on results of successful **3D** numerical modelling of real experiment in similar geometry and with exact physical properties of liquids where the PAS was found. Two liquids are considered, sodium nitrate (NaNO3) [1] and n-decane [2]. Assuming that the tracers are uniform rigid spherical particles, we employ a simplified form of the Maxey-Riley equation [3] as the governing equation for describing motion of the particles in a fluid. The force acting on a particle due to counteraction with the fluid is a sum of the pressure-viscous force, the added mass force, the buoyancy force, and the Stokes drag force. Since the problem is non-stationary, a system of full three-dimensional Navier-Stokes and energy equations was solved together with the three-dimensional Maxey-Riley equation.

The steady flow at small temperature difference consists of a single toroidal vortex and the motion at the interface is directed downward from the hot upper disk to the cold lower one. Increase of Δ T above some critical value leads to instability in the form of standing or azimuthally travelling hydrothermal wave. First of all we successfully reproduce in computations the same flow organization as in the experiment, i.e. azimuthally travelling wave m=3 in case of sodium nitrate (three hot and 3 cold temperature patterns) and m=2 for n-decane. Disturbances of temperature and velocity fields are shown in Fig.1(a). We are able to reproduce PAS exactly as it was found in the experiments (Fig. 1(b)). Figure 1(c) demonstrates that PAS is not formed for particles e.g. smaller than those in Fig.1(b) although the liquid flow is the same. Moreover, varying both ΔT and diameter *a* of the tracers, a PAS map was plotted, showing regions of PAS formation on $a - \Delta T$ plane.



(a) Flow and temperature disturbances at $\Delta T=30$

(b) Tracers of a=100 microns in diameter in flow at $\Delta T=30$ Fig.1 Flow and particles dynamics in NaNO3 liquid bridge under weightlessness

(c) Tracers of a=60 microns in diameter in flow at $\Delta T=30$

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Delayed Droplet-Coalescence for Miscible Liquids

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A Computer-Simulation of two miscible droplets flowing together was derived using a modified lubrication approximation for thin films. The motivation arised due to recent experimental observations showing that sessile droplets not only fuse instantaneously after peripheral contact but also coalesce delayed for many seconds [1]. This rather unusual behavior occurred for droplets with small contact angles consisting of different liquids. After initial contact a thin gap between the droplets was observed through which liquid is exchanged from the low to the high surface tension liquid (see Fig. 1). The lack of measured data suggested the use of numerical simulation that could give further insights in the necessary initial conditions and the basic process. As a first approach 2D-simulations with typical thinfilm-approximations were performed taking into account the convection terms for velocity and concentration fields. As a result the simulation qualitatively reproduces these two distinct dropletfusions as observed in experiments and gives basic explanations for the driving mechanism (see Fig. 2). Nevertheless, further studies and less approximated simulations should be conducted for a more profound understanding.



FIG. 1: stable thin film connecting the two liquids



FIG. 2: Numerical Simulation

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Stationary Patterns in thin liquid films

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In this presentation we focus on a finite wavenumber pattern formation process in thin liquid films. Generally a thin-film equation with destabilizing forces shows a typical nonlinear long-time behaviour in the strongly nonlinear regime, well-known as coarsening. Here we apply additionally an outer temperature gradient and allow evaporation/condensation of the liquid. Evaporation can suppress the typical coarsening and gives rise for formation of stationary patterns. The extended dimensionless nonlinear evolution equation for the free surface h = h(x, y, t), derived from the Navier-Stokes-equations in lubrication approximation, reads then

$$\partial_t h = \nabla \left(\frac{1}{3}h^3 \nabla \mathcal{N}\right) + J(h),\tag{1}$$

where J is the evaporation rate (mass flux) and \mathcal{N} are typical normal forces acting on the free surface [1]. The investigation of (1) reveals unstable wave numbers for a finite k range. We perform a weakly nonlinear analysis in terms of amplitude equations. This allows to calculate the stability of patterns like hexagons, stripes ...

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Forced impalement of liquids by drop impacts on non wetting surfaces

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We report experiments of drop impacts on different micro or nanofactured substrates in order to study the impalement of liquids. The first kind of surface consists of a silicon substrate covered by a carpet of nanowires chemically coated with a low surface energy fluoropolymer. The micro-structure of the surface is similar to some biological superhydrophobic surface (Lotus leaf for example) ([1],[2]). The contact angle is close to 160 degrees, with very small hysteresis. The situation is very similar to that of a drop impacting a hot smooth plate - the so-called Leidenfrost effect. Nevertheless, wheras in Leidenfrost situation the impact velocity could not exceed a certain value in order to avoid the break of the vapour film, here the impact velocity can be increased until a limit beyond which the drop is impaled into the nanowire carpet. In absence of impalement, after the drop hits the surface, it experiences a flattening phase due to its initial inertia, taking the shape of a pancake. Once it reaches its maximal lateral expansion, the drop begins to retract and bounces back. We have extracted the lateral extension of the drop, and we propose a model that explains the trend [3]. We also measured the limit initial velocity beyond which the drop is impaled into the nanowire carpet, and it turns out that the nano-scale and high aspect ratio of the wires provide a very high resistance to impalement. The second kind of surface is a hydrophobic micro-grid, of typical spacing 50 μm used for studiying the impalement in a more fundamental point of view. Different impact configurations were tested. We found a threshold in impact speed above which a small amount of liquid protrudes the grid and emerges to the other side. For small grid spacings, the emerging liquid takes the form of micro-droplets which size is about the size of the grid step leading to method to produce either a mono-disperse spray or a single tiny droplet of sub-nanoliter volume. We propose some attempts of theoretical explanations for various aspects highlighted by the experiments [4].



FIGURE 1: Left : Nanowires carpet. Centre : Drop impact sequence on the nanowires carpet substrate Right : Impact on a microgrid

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Nonlinear dynamics of long-wave Marangoni convection in a liquid layer with insoluble surfactant

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We study the nonlinear Marangoni convection in a liquid layer with free upper surface, located on the horizontal rigid plane. An insoluble surfactant absorbed at the free surface that is convected by the interfacial velocity field and diffuses over the interface but not into the bulk of the fluids. The layer is subjected to a transverse temperature gradient. It is assumed that the film is sufficiently thin, so that the effect of buoyancy is negligible compared to the impact of the Marangoni effect. The surface tension on the interface is assumed to depend upon both temperature and surfactant concentration. Both cases of flat nondeformable and deformable upper surface are considered.

Our study of the linear stability problem shows the stabilizing influence of insoluble surfactant on monotonic and oscillatory instability modes. The stabilizing effect on the monotonic instability is stipulated by the fact that for monotonic disturbances the tangential forces, caused by temperature and surfactant distribution heterogeneity are of opposite directions. The oscillatory instability is characterized by the property that temperature and surfactant disturbances oscillate with a phase shift. Analysis shows that under definite conditions the long-wave oscillations are most dangerous. Deformability of the surface complicates the situation.

Also, we derive the set of long-wave nonlinear evolution equations that govern the spatiotemporal dynamics of a thin liquid layer with surfactant on its interface. Based on these evolution equation we then carry out a weakly nonlinear analysis of the system.

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Front dynamics and pattern formation in a rectangular fluid layer under nonhomogeneous heating

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We present results on the study of a experimental cell which consists of a rectangular fluid layer (450 mm × 60 mm) opened to the atmosphere and heated along a central line. The fluid is silicone oil 5 cSt ($Pr \approx 80$) and we have worked with layers of depths d = [2-5 mm]. As we change quasi-statically the vertical temperature difference ΔT , the Bénard-Marangoni mechanism is responsible for the different convective patterns. The 1D-dynamics along the heating line is recorded implementing the shadowg-raphy technique (see Fig. 1(a)). Our aim is to understand the dynamics of a spatially extended and dissipative system undergoing a cascade of symmetry-breaking bifurcations towards weak turbulence.

For intermediate $(4 \le d \le 4.5 \text{ mm})$ and shallow $(d \le 3 \text{ mm})$ layers, we study secondary bifurcations from an initial stationary cellular pattern (ST) with mode M_s . Depending on d, the new emerging modes can be: one oscillatory mode $(M_{v+} \text{ or } M_{v-})$ for a traveling wave pattern (TW), or two counter-oscillatory modes $(M_{v+} \text{ and } M_{v-})$ for an alternating pattern (ALT) (see Fig. 1(a)).



FIG. 1: (a) Shadowgraphy images of the TW and ALT patterns with a zoom view below; (b) Spatiotemporal diagram of an asymptotic regime with a stationary 1D-front connecting the TW and ALT patterns.

At the asymptotic regimes and close to the threshold, these patterns are bistable. We show that this bistability arises from subcritical bifurcations [1, 2]. Bistable patterns are connected by well-definite 1D-fronts with null front velocities v_p (see Fig. 1(b)) or by fluctuating 1D-fronts that collapse in the form of irregular islands stochastically spread over the stationary cellular pattern (mixed ST/ALT pattern).

The front dynamics in the mixed ST/ALT pattern obeys to the presence of a resonant triad (M_s, M_{v+}) and M_{v-} which is locally triggered over the stationary cellular pattern in the form of islands in the ALT pattern (see Fig. 2(a)). This spatiotemporal chaotic regime corresponds to a subcritical bifurcation "taken to the infinity". As we further increase ΔT , an inhomogeneous growth of these island is produced monotonically until a large coherent structure appears with a zig-zag characteristic pattern (ZZ). This

ST/ZZ pattern corresponds to a spatiotemporal beating regime [2, 3] (see Fig. 2(b)) that takes place through a supercritical bifurcation.



FIG. 2: Surface images of demodulated amplitudes of the critical modes where warm colors represent: (a) irregular clusters in the mixed ST/ALT pattern with fluctuating fronts; and (b) stationary clusters in the spatiotemporal beating regime ST/ZZ.

Stationary 1D-front are characterized by measuring an attenuation length of the new mode inside the coexistent pattern. We find out a constant attenuation length $\xi \approx 30$ mm which corresponds to the number of convective cells making up the front (from 3 to 5 wavelengths depending on *d*).

We show that the characteristic front dynamics of each regime remains unaffected in transients which are performed by perturbing locally the fluid surface. Except for the front dynamics of a convectively unstable pattern when we place the system close and below the TW threshold. We find out that the front dynamics in transients follows the nonzero group velocity ($v_p \approx v_g$) which is $v_g \approx -3.5 v_{\phi}$ (where v_{ϕ} is the phase velocity).

For intermediate layers, when the system is forced to follow the fastest quenches from the cellular pattern (close and below the secondary bifurcation) fluctuating 1D-fronts emerge (ST/ALT) towards the ALT pattern, meanwhile the TW domains do no longer appear. These results [4] show that the fluctuating 1D-fronts are robust coherent structures of the system. Under these conditions, we infer that the response dynamics of the system cannot follow adiabatically the change of the control parameter. Except for the mixed ST/ALT pattern, we conclude that these experimental results on 1D-fronts fit well with the theory of reaction-diffusion systems with two extremely different viscosities [5, 6]. Accordingly, if we take the thermal diffusivity (κ) in front of the vorticity diffusivity (ν) then $v_p \sim \sqrt{\kappa} \approx 0$ mm/s.

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Interaction Between the Buoyant and Solutocapillary Convections Induced by Surface-Active Source Placed Under a Free Surface

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We present the results of experimental and theoretical study of onset and stability of solutal (buoyant and capillary) convection initiated by source of pure soluble surface-active substance (SAS) placed under a free liquid surface. In this situation the solution of a lighter SAS leads to formation of a buoyant convective jet delivering a surfactant towards the free liquid surface, where the latter initiates a solutal Marangoni convection. In the present study it is shown that interaction between these two convective mechanisms can lead to different scenarios of further development of convective flow.

The experiments have been carried out in Hele-Shaw cell with 90*45*3.8 mm size. The glass walls of the latter form the Fizeau interferometer cell to visualize a concentration distribution. The standard trace particles technique is used to study the structure of a convective flow. In addition we can measure a flow velocity in a certain point of the liquid with the help of small anemometer. A small dissolving drop held on a tip of a thin needle and placed at a certain depth under a free surface was used as a source of SAS. Triply deionized water was used as surrounding liquid. The series of aliphatic alcohols from n-butanol to n-heptanol was used to create the SAS source. We found out that depending on the ratio of the buoyant and Marangoni forces different structure of the resulting convective flow can be obtained. Under relatively small ratio an oscillatory convective flow is observed. The period of oscillations depends on size of the drop and its position relative to the free water surface. With increase of the ratio the period decreases and under a certain value of the ratio the oscillatory regime changes to the stationary one. It is shown that one can introduce concentration dynamic Bond number on the analogy of thermal one. It turned out that oscillatory convection changes to stationary one when dynamic Bond number becomes close to that typical for the thermal case.

On the basis of experimental results the theoretical model is proposed. Here we present a simple mathematical model, which shows that a competition between the solutal gravitational convection and the Marangoni convection at large values of the Schmidt number can lead to the onset of the oscillatory mode of convection. The finite difference method has been used to simulate numerically the development of the free convection over a localized source of light surfactant in a rectangular horizontally elongated cavity. The fluid density and the coefficient of surface tension decrease with the growth of the surfactant concentration. The typical values of the Grashof and Marangoni numbers, at which the oscillatory modes of convection arise, are 500 and 700000, respectively. Even though the size of the source is rather small (1/8 of the submergence depth), the oscillation period is about three viscous time units, in which the Marangoni convection proceeds for less than one unit. Time dependence of the convection intensity has been derived and a variation of the convective flow structure has been analyzed. It has been found that the oscillation period is strongly affected by the Grashof number, with the increase of which the oscillation period decreases. The oscillation period also depends on the depth of the source submergence. At low submergence depth one can observe a transition to double-period oscillations. The calculated distributions of surfactant concentration at different phases of oscillations qualitatively agree with the results of the experiment.

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Fig. 1 Flow velocity near the water surface over the hexanol drop vs time. Size and depth of immersion are 1.5 mm and 2.1 mm consequently.



Fig. 2 Intensity of convective motion vs time. Ma= $7 \cdot 10^5$ and Gr=500



Fig. 3 Period of oscillations vs inverse dimensionless immersion depth of SAS source. Drop size: 1 - 1.9 mm, 2 - 1.2 mm.



Fig. 4 Dimensionless period of oscillations *vs* inverse dimensionless immersion depth of SAS source. Ma= $7 \cdot 10^5$, Gr=500 (1) and Gr=1000 (2)

Onset Conditions and Structure of Standing Hydrothermal Waves In a Thermocapillary Liquid Bridge

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It is well known that in a non-uniformly heated liquid bridge a stationary flow is replaced by an oscillatory flow as soon as the Marangoni number approaches some critical value. The resulting hydrothermal wave propagates in the azimuthal direction and takes the value of the azimuthal wave number depending on the Marangoni number and aspect ratio. As it is known from experimental and theoretical investigations [1,2], generation of the hydrothermal wave is preceded by a flow in the form of a standing wave as a result of superposition of two traveling waves. Then, accidentally, only one wave of any direction is left. If the boundary conditions of the liquid bridge are completely symmetric, there is an equal probability of occurrence of waves traveling in both directions. The time of flow generation in the form of a traveling wave strongly depends on the experimental conditions. Long-lived standing waves are generally observed at the boundaries of two regions where the traveling waves with different modes m exist.

However, the results of our experiments suggest that under certain conditions the standing wave rather than the traveling wave may become the preferential and even the only possible form of a flow. One of the ways to change the situation is the asymmetry of the boundary conditions. The other, yet stronger effect is gained by adding the surface-active impurities. The experiments show that even a small amount of such substance precludes the existence of a traveling hydrothermal wave. At any values of the Marangoni number and aspect ratio the flow in the liquid bridge can develop only in the form of standing waves with different azimuthal numbers. During experiments we investigated the flow structure and the temperature distribution over the lateral surface of the liquid bridge. As in the case of a traveling wave, the visualizing particles are accumulated in the ordered structures. The particles gather in the 2m sectors, whereas in the narrow regions separating these sectors they are entirely absent.

We suppose that the effect of the surfactants on the structure of the flow in a liquid bridge discovered in our experiments could be also detected in experiments by other researchers. We have found out that Fluorad coating of a lower rod frequently used in experiments causes partial dissolution of coating, which eventually leads to formation of a surface-active layer. Thus, for example, in [3] the application of FC-725 seems to be the main reason for appearance of an extensive region of standing waves.

It can be reasonably expected that in real technological processes of crystal growth in the melting zone the flow in the form of standing waves is most dangerous. The free surface of the melt often contains additives (carried from the melt volume or generated on the surface by oxidation), some of which might be surface-active. We propose a technique for identification of the wave types encountered in real technological processes, where the use of traditional methods of visualization is not possible. It is essential that these peculiarities of the flow should be taken into account during development of the methods of control and suppression of the oscillatory modes in a liquid zone (by magnetic field, vibrations, local periodic heating, etc.).

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Threshold Onset of Marangoni Convection in Narrow Channels

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There is a number of recent experimental studies [1,2], where the Marangoni convection in Newtonian fluids begins only at certain difference in the surface tension, i.e. in a threshold way, that is quite contrary to the existing theoretical models. To explore the observed phenomenon in more details we investigated the development of the Marangoni convection at the free surface of water placed in the Hele-Shaw cell of different thickness. A capillary motion was initiated by injection of droplet of a surface-active substance. We used an aqueous solution of several aliphatic alcohols as a surfactant that allowed us to vary solubility and surface activity. The resulting distribution of the alcohol in the near-surface layer was visualized by the interferometric method.

The experiments have shown that development of the capillary flow at the horizontal free surface is also of a threshold nature (Fig. 1). It has been found that the value of the surfactant concentration threshold decreases essentially with an increase in the initial concentration of the alcohol in water (in the event of using alcohol solution instead of water). The threshold for the onset of the solutal Marangoni convection decreases with increasing of the cell thickness (Fig. 2), i.e. the considered phenomenon is most essential in narrow channels. The obtained results allow us to suppose that the threshold development of the fluid. This conclusion is also supported by the fact that the value of the threshold decreases with increasing of water purification degree. With the help of chromatography very low content of high-molecular organic compounds, which could play a role of the uncontrolled surfactants, has been found (Fig. 3). These substances have as a rule rather high boiling point. They cannot be removed from water completely even during distillation and deionization processes. The listed above effects observed in water were as well detected in experiments with some other liquids of ordinary purity used in industry and science.

The results of the presented experiments show that an accurate definition of the boundary conditions is of primary importance during development of technological processes especially in narrow surfaces. On the other hand, the experiments show that adding of low molecular surfactants to liquid systems, in which they play the role of a surfactant, is an effective way to control the value of the threshold for the onset of the capillary motion.

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Fig.1. Evolution of the concentration field after injection of a drop formed by aqueous solution of isopropanol with concentration C_0 at the free water surface: (a) - $C_0 = 5$ % (absence of the Marangoni convection), (b) - $C_0 = 6$ % (arising of the convection motion). The cell thickness d = 1.2 mm.





Fig. 2. Threshold concentration of alcohol in the drop *versus* cuvette thickness for different degrees of water purification: *1*- distillate, *2* – deionized water.

Fig. 3. Chromatograms of pure dichloromethane (1) and that comprising the impurities extracted from deionized water (2).

Experimental observations of the effect of ultrasound forcing on liquid structures

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Experimental investigation of the dynamics of liquid structures subjected to external forcing was carried out. Harmonic vibration with the frequency of 20 kHz was applied to flat and cylindrical solid surfaces being in contact with a liquid structure. Various liquids used in the investigation were water, glycerol and mixtures of water and glycerol. In this poster, we present two different experimentally observed effects arising from the presence of forcing which may be of a probably similar nature.

The first experiment is with a thin liquid coating of a stationary horizontal cylindrical needle. In this case, small droplets develop on the underside of the surface a needle due to the action of gravity. Once a droplet becomes sufficiently large it falls down because surface tension cannot stabilize gravity (Rayleigh-Taylor instability). When the needle is subjected to ultrasound axial vibration the droplets emerging earlier on the needle surface become more symmetric with respect to the needle axis and wrap around the surface, thereby generating a structure resembling a series of beads (Figure 1a).

The second experiment re3ported here is that of a flat solid surface touching a liquid surface from above. Upon a contact, the solid surface is slowly pulled up with a liquid bridge of a cylindrical shape attached to it (Figure 1b). The height of the bridge increases significantly if ultrasound forcing is applied to the solid surface in the direction normal to the liquid surface. The effect intensifies when more viscous liquids are used and leads to an increase in the height of liquid bridge before it snaps. The latter is measured for various amplitudes of forcing and liquid viscosities.

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Fig. 1 – The effect of ultrasonic forcing on liquid structures: (a) beads on a cylindrical needle (1 mm in diameter); (b) a liquid bridge pulled by a flat cylindrical surface (4 cm in diameter).

Long-wave Marangoni instability in a binary-liquid layer with deformable free surface in the presence of Soret effect and surfactant adsorption

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The Marangoni instability in a heated binary-liquid layer with a deformable free surface in the presence of the Soret effect has been studied formerly in the case of a prescribed temperature of the solid substrate [1], [2], a prescribed heat flux at the substrate [3], [4], and for arbitrary heat conductivity of the substrate [5]. In the above-mentioned papers, the surface tension was assumed to be a function of the temperature and the bulk concentration of the solution near the free surface.

In reality, the surface tension is determined by the composition of the surface. If the characteristic time of adsorption/desorption processes is small (that is typical for relative small surfactant molecules), the concentration of a surfactant adsorbed at the surface is close to its equilibrium value, hence, it is nearly proportional to its bulk concentration, which leads to the model used in the above-mentioned papers. Otherwise, the adsorption/desorption kinetics and the surfactant advection along the interface have to be taken into account for finding the surface concentration of the surfactant and the local value of the surface tension.

Here we consider the Marangoni instability in a binary-liquid layer with a deformable interface in the presence of the Soret effect and the surfactant adsorption/desorption. The heat flux at the substrate is prescribed; at the free surface, the heat flux is governed by the Newton law. A number of new nondimensional parameters characterizing the problem have been introduced. A special attention is paid to the case of a small Galileo number, where the instability mechanism connected with the surface deformation prevails. Also, the Lewis number is assumed to be small.

The linear stability theory reveals a competition between a monotonic mode and an oscillatory mode of instability, as well as between long-wave and short-wave types of instability. A significant influence of the surfactant adsorption on the stability criteria has been found. By means of long-wave asymptotic expansions, parameter domains for different kinds of instability have been obtained, and transitions between them, which take place with the change of the mean surfactant concentration, are investigated.

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Three dimensional unsteady simulations of a liquid bridge with external gas flow

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The effect of external gas flow on a liquid bridge is examined with the aid of numerical three dimensional unsteady solutions of the Navier-Stokes and energy equations in both the gas and liquid phases. The mathematical model of the flow takes the asymptotic limit of large surface tension, therefore precluding temporal deformations of the gas-liquid interface. In addition, zero gravity conditions are assumed. The gas flow affects the liquid flow by changing the heat transfer and shear stress seen by the liquid at the interface. This effect has the capability of significantly changing the critical Marangoni number in the liquid bridge and therefore attention is focused on this effect of the gas flow. In particular, different velocities (including the direction) of the gas flow are examined in this context. The changes in the features of the flow in the liquid bridge resulting from the gas flow are analyzed with the goal of understanding how the gas flow affects the liquid bridge. In addition, the numerical issues involved with modeling this problem are discussed. Support from BMVIT through the Austrian Space Application Programme under grant number 819714 is gratefully acknowledged.

Control of hydro-thermal waves in a Marangoni and buoyancy driven flow using a gradient-based control strategy

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The possibility of stabilizing the surface tension and buoyancy driven flow in an "open boat" (Fig. 1) geometry through free surface heating is addressed using a gradient based control strategy. The state equations are the two-dimensional unsteady incompressible Navier-Stokes and energy equations which are coupled using the Boussinesq approximation. The control is a spatially and temporally varying heat flux at the free surface. The mathematical model of the flow takes the asymptotic limit of large surface tension, therefore precluding temporal deformations of the gas-liquid interface. The flow is driven by a temperature difference between the vertical walls. At the aspect ratio considered (24.67), two-dimensional unsteady hydro-thermal waves have been previously reported. The aim of the present work is to suppress these waves. The control which does so is found using a conjugate gradient method, where the gradient of the objective function with respect to the control variables is obtained from solving a set of adjoint equations. The effectiveness of choices for the objective function are examined along with the effect of the event horizon length over which the control is obtained by the optimizer. The numerical issues involved with finding the control are discussed and the features of the resulting control are analyzed with the goal of understanding how it affects the flow. Support from BMVIT through the Austrian Space Application Programme under grant number 819714 is gratefully acknowledged.



FIG. 1. open boat geometry

Induced flow in coalescing droplets in a circular tube

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The authors focus on coalescence phenomena of liquid droplets in a circular tube. Especially, induced flow is investigated using fine particles when two liquid droplets coalesces. The particles are suspended in each or both droplets of O(mm) in diameter.

Creeping motion of liquid droplets through a capillary tube is generally employed as a fundamental problem. As example applications, there are fluid handling technique, controlling chemical reaction and so on. The problem is also underlying basis on analyzing the flow of multiphase fluids through porous media[1]. Such phenomena can be seen, for instance, in enhanced oil recovery, breaking of emulsions in porous coalescers and so on[2]. Examples of studies of coalescence phenomena of droplets are flow-induced coalescence of viscous drops in a viscous fluid by Leal, et al.[3] and the interaction and coalescence of liquid droplets in flow through a capillary tube by Olbricht, et al.[2]. However, as far as the authors know, there have been no previous cases in which the induced flow is investigated when droplets coalescence.

In this experiment, a vertical glass tube of 3.5 mm in inner diameter, 8 mm in outer diameter, and 1500 mm in length is used as a test tube. The test tube is filled with a quiescent fluid which is a mixture fluid of glycerol and pure water. Silicone oil droplets with suspended particles are successively injected into the test tube using micro-syringes. Behaviors of droplets and suspended particles are simultaneously monitored by a digital video camera, CCD cameras and high speed cameras placed on a sliding stage. The motion of the stage is electrically controlled to follow the travelling droplets in the tube. Coalescence time is measured. The coalescence time indicates a period between instances when the relative velocity of two droplets becomes zero after their apparent contact and when the coalescence takes place. The spatio-temporal behavior of the particles before/after the coalescence of the droplets takes place is reconstructed. Induced flow (Figure 1) is investigated from the spatio-temporal images. Effect of suspended particles on the droplet coalescence is also discussed.



Fig. 1 Induced flow in coalescing droplets

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Marangoni instability of a heated layer in the presence of a soluble surfactant

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We consider a heated layer of a liquid, which contains a soluble surfactant. A constant heat flux is prescribed at the substrate. Due to the Soret effect the gradient of the volume concentration of the surfactant arises. The surface tension on the free surface depends on both the temperature and surface concentration of the surfactant. Both surface deformation and gravity effects are disregarded. Adsorption kinetics is taken into account.

In the absence of the adsorbed phase the problem was studied in [1] within longwave approximation, both monotonic and oscillatory modes were found. Also, shortwave oscillatory perturbations become critical within a certain interval of the Soret number, $\chi > \chi_c$, where $\chi_c \approx 0.1$ [2].

We have shown that the presence of adsorption leads to stabilization of the layer for both heating from below and from above. The main effect, which leads to the stabilization, is the advection of adsorbed surfactant along the free surface. This advection results in decrease of the surface-tension gradients and, hence, in suppression of the convection. It is characterized by dimensionless equilibrium value of the surface concentration $\Gamma_0 = C_a + \chi \operatorname{sign} m$, where C_a is the volume concentration of the surfactant on the bottom and m is the Marangoni number.

Another stabilizing effect is the adsorption kinetics, which slows down the transport of the surfactant to the free surface. This effect is governed by dimensionless rate of adsorption K_a . It is worth mentioned that $K_a = 0$ corresponds to the absence of the surface phase, see [1, 2]. In most of calculations we neglected the surface diffusion of the surfactant.



FIG. 1: Stability map $m(\chi)$ for the longwave perturbations, $\Gamma_0 = 1$, black, red, and blue lines correspond to $K_a = 0, 0.04, 0.1$, respectively. Solid lines are the stability boundaries for the monotonic mode, dashed – for the oscillatory mode. Domains of stability are marked by "S". Domain to the left (right) of the vertical dotted line, $\chi > 1, m > 0$ ($\chi < -1, m < 0$), marked by "unphys." are unphysical, since they correspond to negative value of C_a .

Owing to these effects, the interval of the Soret number, where the longwave monotonic mode emerges, considerably diminishes, see Fig. 1. The shortwave oscillatory mode does not play any role for the surfactant, since even for K_a as small as 0.001 the shortwave perturbations are completely damped
by advection of the surfactant. With increase in Γ_0 the critical value $\chi_c(\Gamma_0, K_a)$, corresponding to emergence of the shortwave oscillatory mode, grows fast and leaves the physical domain $\Gamma_0 > \chi$.

Both the above-mentioned tendencies, the shift of the monotonic mode to smaller χ and the strong damping of the shortwave oscillatory mode, yield the increase of the interval of the Soret number, where the longwave oscillatory mode is critical.

Thus, the impact of the surfactant is completely different from that reported earlier [3] for thermally conductive substrate. Recall that the surfactant is found to lead to destabilization of the layer in the cited paper. In spite of profound stabilization of the monotonic mode, an oscillatory one emerges and becomes critical (see also [4]).

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Marangoni Convection Experiments in 'KIBO' on ISS - The Second Series of Experiments -

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Marangoni convection experiments have been carried out in 'KIBO', the Japanese pressurized module, on ISS in the period from July 30 to August 25, 2009. This is the second series of Marangoni Experiment in Space (MEIS2), with the first one (MEIS1) being carried out in the period from August 22 to December 16, 2008. MEIS2 aims at (1) determining critical temperature difference, ΔT_{cr} , for the onset of oscillatory flow for AR up to 2.0, where AR is the aspect ratio of the liquid bridge, (2) revealing three-dimensional flow fields in oscillatory state and (3) clarifying the characteristics of surface velocity. In this presentation, some important results from MEIS2 are given.

A liquid bridge of 5cSt silicone oil is suspended between two circular disks heated differentially. The temperature difference between the disks is controlled for the determination of ΔT_{cr} at which the flow field in the liquid bridge starts to oscillate. The volume ratio of the liquid bridge is set to be 0.95. All the functions of the Fluid Physics Experiment Facility (FPEF) are exploited to determine ΔT_{cr} and to observe oscillatory flow. Main apparatuses used are an IR imager, a 3-D PTV system, a photochromic surface velocimetry system and an immersed fine thermocouple sensor. The procedures developed in MEIS1 for generating bubble-free liquid bridge are applied successfully. Figure 1 show a side-view picture of the liquid bridge, 60mm in length, suspended between 30mm-diameter disks, thus giving AR=2.0.

A sufficiently long waiting time to achieve full diffusion of temperature and velocity fields inside the liquid bridge is given to each run of experiments. It is, e.g., 30min for Ar=2.0 where the diffusion time estimated is 15.8min. The photochromic dye visualization is used to detect the onset of oscillation because this technique is found to be very sensitive measure for large Ar. Figure 2 show the relationship between ΔT_{cr} determined and AR, together with that obtained in MEIS1. In general, ΔT_{cr} decreases with AR, but not monotonically. Such is behavior is more clearly revealed in the plot of the critical Marangoni number, $Ma_{c,H} = |\sigma_T| \Delta T_{cr} \cdot H/(\mu \cdot \alpha)$, as function of AR. Some details of the characteristics of oscillatory flows are obtained from (1) surface temperature measurement, (2) 3-D velocity measurement, and (3) surface velocity measurement. Those results will be reported in the presentation.



Fig. 1 A long liquid bridge (AR=2.0)



Fig. 2 Critical temperature difference vs. AR

Coating of a fiber with a non-newtonian fluid

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During the coating process, a liquid film can become unstable developing structures which alterate the final film. We consider a one-dimensional problem in which a viscous film flows down a vertical fiber under the action of gravity (see Figure 1). In the case of newtonian liquids, three regimes were observed depending on the dominance of the physical mechanisms, i.e. the surface tension, the viscous forces, the gravity and inertia [1]. Since many industrial applications are based on the coating of fluids exhibiting non-newtonian properties, modeled experiments with polymer solutions are investigated. Also, for polymer solutions, the non-Newtonian effects can be either shear thinning (the effective viscosity decreases with increasing flow velocity gradient, i.e. shear rate, in the fluid), or elastic effects (Figure 1). In the case of a shear flow on a fiber, since the velocity gradient becomes larger near the thinner regions of the film, the viscosity and the elastic effects of the solution are modified there. Thus the structures of the film flowing down is strongly dependent on the polymer properties.



FIG. 1: (Left: macroscopic view) Coating of a thin film axisymmetrically along a vertical cylindrical fiber. Topographic variations are strongly dependent on the physico-chemical system. (Right: microscopic behavior) sketch of a polymer chain under equilibrium configuration (a), under shear flow (b) causing anisotropy in the tension field.

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Drying a colloidal dispersion in a confined geometry

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Desiccation of a film of complex fluids is often accompanied by hydrodynamic instabilities within the liquid, generating non-planar films (see Figure 1a). In the case of curved liquid-vapor interface, non-uniform evaporation induces a complex flow within the liquid. That can be quantified in a modelled experiment by sandwishing a drop between two parallel circular glass slides [1]. This thin cell allows the drop to dry through a radial evaporation flux. In the ideal case of ultra pure solvents the drop completely evaporates as gas diffusion proceeds from the edge of the drop towards the edge of the cell. However in the case of a solution, non-volatile species are advected to the drop edge. During this process the solute concentration near the drop edge increases; further accumulation of solute leads to a skin formation. Depending on the nature of the solute, it can result in a gelled or glassy skin [2]. Here the case of a suspension of hard colloidal particles is reported. Experimental investigations by particle tracking using fluorescent probes added to the system allow quantifying the concentration gradient and particle transport within the liquid near liquid-vapor interfaces. Particularly the case of concentration gradient within the liquid near concave or convex interfaces are considered. Figure 1b gives an example of experimental evidence of this process.



FIG. 1: (a) Sketch showing concentration gradient near a curved liquid-vapour interface. (b) Observation by fluorescence microscopy: white arrows illustrate the flow of particles within the liquid phase near a convex liquid-vapour interface.

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Numerical simulation of the Faraday instability

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In 1831, Faraday discovered that the interface of a fluid contained in a vibrating vessel could form standing crystalline patterns [1]. Since then, new kinds of patterns have been observed: quasipatterns, oscillons and superlattices. The richness of the patterns has made the Faraday instability very well-known, and therefore, it has been the subject of numerous theoretical and experimental investigations, but the numerical computation of this problem has never been completely performed. Here we report our work on nonlinear, fully 3D simulations of the Faraday instability in a horizontally periodic box [2]. We have first validated our numerical approach in the linear regime where the instability thresholds and the main eigenmodes have been compared with those predicted by the linear theory [3]. Then, validation of the nonlinear regime has been conducted by comparing the patterns at saturation (squares and hexagons) and their main spatial modes with the first experimental results that provided accurate measurements of the local interface height [4], [5]. However, we observe that the hexagonal state is only transient and vanishes in favor of patterns with other symmetries. The hexagons may constitute a fixed point of a homoclinic orbit. Simultaneously, we have undertaken the analysis of the mean flow instability in an annular container subjected to vertical vibration. A secondary threshold is found, above which a mean horizontal flux is generated. This bifurcation is explored numerically and the corresponding flow is characterized.



FIG. 1: Snapshots of hexagons taken at different times. Surface: interface height. Colormap: vertical component of the interface velocity. Vectors: velocity fields on horizontal planes

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Instabilities in clustering of colloids by alternate electric fields

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To prepare homogeneous colloidal crystals by vertical deposition method [1-4], it is necessary to maintain during the process: (a) a constant and homogeneous concentration of colloid and (b) a constant speed of the contact line. To explore further, it is interesting to expose these systems to external fields such as electric, thermal or magnetic to understand the response of colloids to these fields.

We focus on the effect of alternate electric fields on water-based colloidal suspensions where the particles are negatively charged. Instabilities observed are mainly related to electro-wetting phenomena where the particles follow the induced flows near the contact line. We measure *in situ* the evolution of the system. We also consider the effect of interaction between the particles focusing on the cluster formation and its evolution.

The experimental setup replicates a vertical deposition design with a free-surface, but with significantly minimized evaporation. The experiments are performed using a cell which is built in such a way that two substrates are placed vertically and the distance between them is of 1 mm. Electric fields are applied perpendicular to the substrates. The magnitude and frequencies of the applied fields are of the order of 1 V/mm and 1 Hz respectively.



FIG. 1: Response of the particles for the applied amplitude at constant frequency 1 Hz (left) and the applied frequency at constant amplitude 1.8 Vpp (right).

In this present work, we measured the frequency of particles which is then compared with the applied frequency. At constant frequency of 1 Hz (Fig.1 – left) and constant amplitude of 1.8 Vpp (Fig.1 – right), the frequency response of the particles is close to the applied frequency. This is the expected behavior at low frequencies where the time response of the contact line is fast enough to follow the change in the electric fields. This behavior might be valid at low values of applied frequency and amplitude. At higher values, studies from various research groups [5, 6] explain the generation of flows which are less pronounced in lower values of applied frequency and amplitude . Even though, by considering the long term evolution of the clusters at lower values, bulk flows play a significant role in the evolution of clusters by drifting the direction of particles to different positions.



FIG. 2: Snapshots of the region near the contact line at different times. (from top to bottom) Initial, After 90 seconds, Final. Bright zones are clusters or particles. Scale bars are 25 μ m. Dark regions in the images towards the edges are due to inhomogeneous illumination.



FIG. 3: Intensity profiles for the clusters shown in Final snapshot of FIG. 2.

A long term evolution of the system to applied frequency of 3 Hz and amplitude of 1.8 Vpp is shown (Fig.2). These snapshots explain the cluster formation near the contact line and the influence of bulk flows in the evolution of cluster. Also the variation of contact angle due to electro-wetting [7] depletes the depositing zone into two parts. The first region (near the contact line) contains particles which are the major components to form clusters. The second one is the bulk suspension. Intensity profiles (Fig.3) are calculated from the space-time diagram and the spatial frequencies for the forming clusters are calculated which will explain the mechanism of formation and interaction between clusters.

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Experimental studies on colloidal crystallization: Effect of electric fields on the dynamics of contact line

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Self assembled monolayers of colloidal particles have encountered potential applications starting from fabrication of photonic devices to materials processing. To fabricate colloidal crystals of desired order, vertical deposition is one of the simplest process which is derived from the Langmuir-Blodget technique [1–3].

We present the experimental results on the effect of DC fields on the deposition of negatively charged micron-sized polystyrene particles in a vertical deposition configuration. The electric field effects are explored by measuring the speed of contact line at micro and macroscopic length scales. The formed structures and patterns are correlated with the measured speeds.

The experiments are performed by using a deposition cell which is maintained in a chamber of controlled temperature and fixed humidity. The cell is designed in such a way that two substrates can be placed vertically and the distance between them is of 1 mm. Also, the design allows us to follow the receding contact line which is recorded by a video camera. The electric fields are of the order of 1 V/mm and are applied perpendicular to the ITO coated glass substrates. We measure the *in situ* evolution of the contact line on the substrate where the voltage is applied with respect to ground.

In this work, we measure the speed of contact line at different applied voltages (from -1.2 V to +1.2 V), at different concentrations (from 0.1% w/w to 0.7% w/w) and also at different length scales. At macroscopic length scale, for every experiment, we build a histogram of these speeds for every position and time. The characteristic speeds are calculated by fitting the cumulative histogram with normal distribution functions. The mean value of each distribution represents the corresponding characteristic speed. At microscopic length scale, for each type of structure we measure the average speed of contact line and they are plotted against initial concentrations as well as applied voltages.

The characteristic speeds mainly depend on local concentrations, forming morphologies, previously deposited structures and dynamical perturbations (unbalanced forces due to pin-depin in the contact line at the counter substrate). All these factors may occur in the same experimental realization and they are interrelated. The morphologies dominantly observed in these kind of systems are multilayer (ML), compact monolayer (CM), non-compact dense (NCD) structures and non-compact sparse (NC) deposit. In addition to these structures, vertical non-dense stripes (at lower concentrations) and vertical dense columns (at higher concentrations) are found due to an instability in the local concentration near the contact line [4].

Electric fields affects the conditions at which the deposition takes place (e.g. contact angle, local concentration of particles, ...). Increase in voltage brings more particles near the depositing zone which increase the probability of forming more compact structures (ML or Monolayer or Vertical Column Multilayer). Flows are partially generated due to the capillarity rise into the previously deposited structure. These flows drag further more particles towards the depositing region [5]. This results in a vertical column multilayer which depends on the strength of the flows and on the local concentration. At negative voltages, many multilayer or monolayer form in counter substrate which frequently executes pin-depin behavior. This leads to rapid advancement of the contact line in the observed substrate. These type of behavior completely distorts the local parameters which make the system more complex and non linear.



FIG. 1: Speed of contact line as a function of concentration at 1.2 V for Multilayers and Vertical column multilayers.



FIG. 2: Optical micro-graphs (Magnifications: A-10X, B-2X, C-20X, D-10X) of the colloidal crystals. A - *In-situ* deposition of Vertical Column Multilayer (VCM) at +1.2 V, 0.7%; B - Dried VCM at +1.2 V, 0.7%; C - Dried Monolayer at -0.9 V, 0.5%; D - Dried pair of ML at 0 V, 0.5%. Scale bars are 25μ m for A,C,D and 500μ m for B.

Also an increase in concentrations (local as well as global) initially produces non compact structures and they transform to other compact structures as the contact line recedes. At 1.2 V (Fig.1), the speed of contact line for multilayer (ML) increases as we increase the concentration which shows the formation of more compact structures[4]. Also at higher concentrations for the same voltage, the availability of particles transform the ML to VCM (Fig.2) as contact line evolves. The electric fields and increase in concentration control the amount of particles reaching the substrate resulting in different morphologies and thus the applied fields have an impact on the speed of contact line.

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Domain self-adaptation induced by Faraday instability

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When subjected to vertical oscillation, the free surface of a liquid is unstable to waves. This instability was discovered by Faraday in 1831 and widely investigated in confined geometry afterwards. It is well known that, at fixed forcing frequency, the threshold amplitude for the Faraday waves formation increases as the liquid viscosity increases. Starting from this fact we can create a system in which the Faraday instability can develop in a deformable domain. If we deposit a drop of a liquid on a bath of another liquid that is more viscous and denser, the two liquids being immiscible, we can induce Faraday waves in the drop and not in the bath. Such waves deforms the drop from its initial circular shape by exerting a pressure on its borders and an elongation of the drop is usually observed.

This behavior has been investigated for different couples of liquids differing in surface tension, density and viscosity. Two archetypes of the phenomenology show up. In the first type of behavior the drop deforms in a stable elongated shape (Fig. a), its length increasing with the forcing amplitude increase (b). Such an elongation has a limit due to the forcing, over which the shape stability is lost. In order to describe this stationary shape we developed a model based on the equilibrium of two quantities: the line tension, that tends to keep the drop circular, and the pressure exerted by the waves on the borders. In the second type of behavior, we also observe an elongation but the dynamics is different: the elongation does not saturate. The resulting filament curves (c) and ultimately breaks. Its remnants are observed to stabilize in four different regimes: small drops at rest (with no waves), short stable filaments, rings or propagating croissants (d).











(d)

Emergent spatiotemporal ordering of particles in non-stationary thermocapillary flows

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The effect of focusing of small rigid tracers in a one dimensional dynamical structure [1, 2] was discovered experimentally more than a decade ago. This remarkable effect, dubbed PAS, remains poorly understood theoretically despite being well-studied experimentally.

The goal of our current contribution is two-fold: first, we give a discussion of the essential features of the phenomenon leaning on results of our numerical simulation. Second, we propose that that PAS formation is an emergent phenomenon, which is brought about by *phase-locking* between the turnover motion of particles in the axial plane and the azimuthally traveling hydrothermal wave.

In the first part of our presentation we look at projections of particles' trajectories on the (r,z)-plane; we suggest that the observed formation of clearly defined attractors is a signature of the arising spatial ordering in the system. Next, we propose and verify numerically a synchronization condition, which links the angular frequencies of the particle motion and of the hydrothermal wave. This synchronization condition has a transparent physical meaning of 'angular Doppler shift'.

In the second part of our presentation we proceed to build a simple discrete model of the particle-wave interaction, which displays the *emergence* of the ordering for certain values of parameters. Our model is closely related to the celebrated circular map [3]. We show that the geometrical shape of the emergent particle accumulation loops is determined only by the traveling wave mode m, and the rational winding number of the circular map, W = p:q. The loop projections are closely related to the well-known *rhodonea* or *rose curves*, and the number of 'petals' n = m q.

Finally, we give a discussion of this effect in a broader context of particle segregation and focusing phenomena in complex flows.

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Deforming Interfaces with Magnetic Fields

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In the experimental exploration of interface instabilities and more generally of interface dynamics, a key issue is the initial shape of the interface. In a normal situation, the shape of a static fluid-fluid interface is governed by the hydrostatic balance between pressure and gravitational forces, sometimes corrected by local capillary effects. As a consequence, most of the time, the equilibrium static interface is flat. Many experiments, though, would benefit from the possibility of creating static interfaces of arbitrary shape, for example serving as initial conditions for dynamic processes. In this presentation, we will demonstrate how magnetic levitation can be put to that use.

One of the authors pioneered paramagnetic levitation of classical fluids, in which a magnetic compound is dissolved in a classical fluid in order to increase its magnetic susceptibility [1]. By applying a magnetic field gradient to such a fluid, a body force is created which can be controlled arbitrarily in time (time-dependent effective gravity) and in space (non-uniform effective gravity). Here, we apply this technique to a system made of a glass fluid cell in which two immiscible fluids of different densities are confined, one of which is magnetic. Without any magnetic force, the stable situation ensues when the denser fluid rests at the bottom of the cell with the less dense fluid lying on top, separated by a horizontal interface. By using an electro-magnet with carefully designed Faraday pole pieces, we create a region of almost homogeneous magnetic force, which is equivalent to applying an arbitrary effective gravity to the magnetic fluid. Between the magnet's pole pieces and the fluid cell, we then place deformed wires made of a magnetically permeable material (see figure 1). As a result of the presence of the wires, the local magnetic force is distorted. The resulting situation is that of a stable static interface in hydrostatic equilibrium under a non-uniform effective gravity field. An example of such a process is represented in figure 2, where permanent magnets were used to deform an interface between air and a magnetic liquid in hydrostatic equilibrium.

We developed a numerical procedure in order to solve the magnetostatic problem associated with the Faraday pole pieces and the permeable wires and then used that result to predict the shape of the fluid-fluid interface by energy minimization. We observe a very rich set of possible interface shapes, even for the simple geometry of sinusoidal wires. More complex shapes of wires were then explored, leading to interesting possibilities in terms of interface control. The application of this technique is illustrated in the example of a Rayleigh-Taylor instability [2], for which we explored the dispersion relation by forcing initial periodic perturbations of the interface of arbitrary wavelengths.



Figure 1: Experimental setup



Figure 2: Deformed hydrostatic interface between air and a magnetic liquid in the presence of permanent magnets

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Experimental Exploration of the Dispersion Relation of Rayleigh-Taylor Instability

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Although it has been known theoretically for more than a century, the Rayleigh-Taylor instability has eluded careful experimental exploration. The reason for this lies in the very nature of the instability, driven by the hydrostatic imbalance between a dense fluid resting on top of a less dense fluid in a gravity field. For example, it is almost impossible to control a Rayleigh-Taylor-unstable interface sufficiently long to run a careful experiment where the initial conditions are known with precision. To circumvent this difficulty, many strategies have been proposed, from the rapid removal of a physical partition between the two fluids to an abruptly accelerated fluid cell wherein the effective gravity changes sign during the acceleration.

In 2006, we demonstrated the use of magnetic levitation of classical fluids to run carefully controlled Rayleigh-Taylor experiments. In our protocol (based on the pioneering work of one of the authors on magnetic levitation of fluids[1]), a glass cell filled with two fluids of different densities was placed in an electro-magnet equipped with carefully designed pole pieces. One of the fluids had a large magnetic permeability, and the pole pieces were designed so as to create a strong magnetic field gradient. This resulted in a quasi-homogeneous body force applied to the magnetic fluid, enabling us to stabilize an otherwise unstable interface. A Rayleigh-Taylor instability could then be induced simply by turning off the magnet [2]. Beyond that first demonstration, a novel technique has been designed to impose an initial deformation on the interface. By placing magnetically permeable wires between the fluid cell and the pole pieces, we are able to distort the magnetic field locally and create a non-uniform effective gravity at the interface. Under such conditions, the initial interface shape is no longer horizontal, but can exhibit an almost arbitrary shape depending on the shape of the wires. With this experimental set-up, we are able to force an initial perturbation on the interface and then observe its growth by Rayleigh-Taylor instability (see figure) [3].

In this presentation, we will show recent the recent results of a systematic exploration of the dispersion relation of the instability. Beyond the verification of a known theory, this result paves the way for a more systematic exploration of non-linear growth and mode couplings in the Rayleigh-Taylor instability for arbitrary (and controlled) distributions of initial perturbations.

a) Rayleigh-Taylor growth for an initially flat interface



b) Rayleigh-Taylor growth for an initially sinusoidal interface



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Deformation of partially engulfed compound drop undergoing thermocapillary migration

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Two-phase hybrid drops, which are comprised of immiscible phases, occur in various natural and technological processes and environments, e.g. the atmosphere, liquid membranes and liquid bi-layers,

direct contact heat transfer and phase separation processes. One phase of such an aggregate is completely or partially engulfed by the other one. A compound drop with partial engulfment has three interfaces between the components of the aggregate and facing the ambient fluid (see Figure 1 for a schematic description). At equilibrium, all three interfaces are segments of spheres. The angles at the three-phase contact line are determined solely by the ratios of the interfacial tensions, and the resulting configuration of the aggregate depends on the relative volumes of the drop's components. Exact analytical solutions describing creeping motion of such a hybrid drop in an infinite viscous domain under the influence of Marangoni effect due to various temperature distributions are constructed in [1] and [2]. However, when the drop moves in a non-



Fig 1. Schematics of a compound drop

isothermal ambient medium, the interfaces are deformed due to viscous stresses and to the nonhomogeneous surface tension. We assume that the surfaces deform as the drop moves through the ambient fluid, and consider the deformation making use of perturbation method. It is assumed that the capillary numbers associated with all 3 interfaces are relatively small, $Ca_i = \varepsilon \delta_i, \varepsilon \ll 1, \delta_i = O(1), i = 1, 2, 3, \text{ and thus, corrections of the solutions obtained in the}$ undeformable case, $\varepsilon = 0$, can be constructed making use of a regular perturbation technique. The problem is reduced to a 6-th order system of ordinary differential equations with four boundary and two integral conditions, the latter reflecting conservation of mass for the two phases comprising the drop. Results concerning the axisymmetric deformation of drops undergoing Marangoni migration are presented for a variety of the physical parameters involved, such as viscosity ratios, initial configuration of the compound drop and temperature dependence of the surface tension of each interface. The cases of spontaneous thermocapillary migration and the motion in an externally imposed temperature gradient are considered. For the latter case the evolution of the interface of the drop propagating to hotter region is taken into account, while for spontaneous migration, the deformations are steady.

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Roles of Organic Layers on Anomalous Surface Tension Temperature Dependencies of Metallic Nanofluids with Dilute Long-Chain Alcohols

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Dilute aqueous solutions of higher carbon number (≥ 4) alkyl alcohols have particular properties such that the surface tension increases with temperature. Using these kinds of fluids, named "Self-rewetting fluids" as working fluid of thermal management devices has led to improvement of heat transfer performance because of the supply of cooling liquid at the dry patch on the heated surface, induced by the positive thermocapillary effect. We reported that 1-butanol containing metallic nanoparticle solutions (nanofluids) synthesized by an aqueous-phase reduction method have been shown several times larger positive surface-tension gradient with temperature [1], [2].

In this study, therefore, to reveal mechanism of the acceleration of surface tension anomaly induced by metallic nanoparticles, the details of surface tension temperature dependency of the higher alcohols containing citrate-capped gold nanofluids and four different molecular weight (MW) macromolecular polyvinylpyrrolidone (PVP) -capped silver nanofluids have been investigated experimentally. The enhancements of the positive surface tension gradients with temperature compared with the base aqueous alcoholic solutions were observed in alcohol containing citrate-capped gold nanofluids and some particular MW PVP-capped silver nanofluids. From the results of zeta potential measurements for the nanofluids, these particular surface tension tendencies seem to be related with the different two kinds of alchol-nanoparticle interactions, (a) the hydrogen bond with citrate (low MW) organic or lower MW PVP layers on nanoparticle and (b) absorption on active bare metal surface of rod-like silver nanoparticle covered with higher MW PVP.



Citrate-capped Au nanofluid with higher alcohols PVP-capped Ag nanofluid with 1-butanol

Fig. 1 Surface tension as a function of temperature for metallic nanofluids with alcohols.

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Particles of different density in thermocapillary liquid bridges under the action of hydrothermal waves and the role of gravity

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We investigate small cylindrical liquid bridges (LBs) from sodium nitrate melt (density ρ = 1.9 g/cm³) with radius a= 3.00 mm and length L of the order of a few mm at normal gravity (g₀), heated from above by ΔT . These LBs display time-dependent thermocapillary flow in the form of degenerated hydrothermal waves (HTWs), (traveling azimuthally) when a critical Marangoni number Ma^c= $\delta\sigma/\delta T \cdot \Delta T \cdot L^{-1} \cdot a^2 \cdot \eta^{-1} \cdot \chi^{-1}$ of the order of 8000 is exceeded (surface tension = σ , dynamic viscosity = η , thermal diffusivity= χ). For special aspect ratios A(m)=2.1·m, with m= 1,2,3,...,only one pure mode m and its harmonics of the instability are exited. The mode m is defined with the azimuthal wavelength λ of the instability as the number of wavelengths m on the circumference of the LB: m· λ = 2· π ·a.

If we bring small particles (typical diameter 20μ m) in dilute concentration for flow visualization into the LB with an A(m) and at aproximately 2 Ma^c, we observe an interaction of the HTW with the particles which brings them from the homogeneous 3-D distibution in the bulk fluid of the LB into the 1-D structure of a dynamical spiral closed particle-string, called dynamic particle accumulation structure (PAS) /1,2/. The spiral particle string rotates with the basic flow, it has m windings and reflects the mode of the HTW /1/.

To form (PAS) the particles must perform two steps.

(1) The particles must migrate from the LB interior to stream lines that pass near the free surface to come under the action of the HTW /3/.

(2) The HTW must interact with the particles in a resonant fashion. The period of the HTW and the orbiting time of the particles in the vortex must be multiples of each other /3/.

We are now interested in step (1) and in the role of gravity in case of particles which are considerably less dense than the fluid, $\Delta \rho = (\rho_p - \rho_f) < 0$. With particles with higher density than the fluid, $\Delta \rho > 0$, we could demonstrate the formation of PAS under microgravity, and we concluded that PAS is a pure Marangoni effect, not needing buoyancy for its formation /4/. This is not true for all particle densities, because buoyancy allows to form PAS under normal gravity g_0 with particles with $\Delta \rho < 0$, whereas these particles cannot form PAS under microgravity.

The observations and explanations are as follows: In a LB ($\rho_f = 1.9 \text{ g(cm}^3)$ at at $g = g_0$ and Ma<Ma^c, seeded with two kinds of particles ($\rho_{p1}=0.25 \text{ g/cm}^3$ and $\rho_{p2}=4.0 \text{ g/cm}^3$), we found the lighter particles in the vortex centre due to centrifugal foces and the heavier particles assembled on the torus of streamlines grazing the free surface, mainly due to Stokes drag and centrifugal forces (Fig.1 shows a central vertical light sheet. The particles in the surface flow are not visible because of optical problems with the liquid cylinder). At $(g=g_0)$ the particles will escape from these orbits during long times due to the action of gravity; the heavier particles will sink and assemble on the lower solid boundary of the LB (the bottom), whereas the lighter particles will rise and tended to assemble at the upper boundary (the ceiling). At zero gravity (g=0), all of the lighter particles will be trapped in the vortex centre, where they cannot interact with the HTW and thus cannot form PAS at g=0. At normal gravity these lighter particles have another possibility to perform step (2) for PAS-formation: it was observed that a good fraction of particles with $\rho = 0.78$ gcm⁻³ escaped from the return flow and from the vortex centre (they rose due to buoyancy), assembled under the "ceiling" and drifted there in creeping flow in the flow boundary layer towards the edge of the LB (Fig. 2 shows a sketch of these obsevatons in a vertical central cut). There they join the surface flow, come directly under the action of the HTW and PAS was reformed continously. The final state of these lighter particles at $g=g_0$ is a PAS-state which is continuosly eroded and continously restored due to the action of gravity. Note that the final state of the heavier particles at $g=g_0$ is the bottom of the LB whereas some of the lighter particles will always be found in suspension and in PAS. Note once more that PAS cannot be formed under 0-g with these lighter particles but PAS is formed effectively under $g=g_0$ due to buoyancy assistence.

The effectiveness of buoyancy to assist PAS can be seen comparing the formation time τ_{onset} for the onset of PAS of different particles after homogenizing the particle distribution in the liquid by stirring it (Fig.3). Particles from three different materials (different densities), but with almost the same particle diameter around 13 µm have been used in melts of different densities and τ_{onset} has been measured. The density relations are expressed as $\alpha = \rho_{particle}/\rho_{liquid}$. On the side of particles which are more dense than the liquid ($\alpha > 1$) one observes the trend of decreasing formation time with decreasing density difference. Neutrally buoyant particles form PAS fastest. One the side of less dense particles, the buoyancy-assisted way of these rather light particles into the flow along the free surface must be very effective because τ_{onset} of the particles with $\alpha=0.4$ is comparable to particles with $\alpha=1.2$.











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IMA benchmark results in liquid bridges

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The scientific results for small Prandtl number ($Pr = v/\chi$) fluids have direct applications to crystal growth as their typical values for liquid metals is $Pr \sim O(10^{-2})$. The majority of experiments and real growth processes are carried out in ground conditions, where the shape of melt zone is deformed. Numerical solutions of thermocapillary and buoyant convection in liquid bridges for small Prandtl number fluids ($Pr \approx 10^{-2}$) with curved free interfaces were benchmarked [1] and presented in the Second International Marangoni Association, IMA-2 Congress, Brussels, 2004. The study consisted of two parts: (1) investigation of axisymmetric steady states in curved liquid bridges in a wide range of contact angles and different gravity conditions; (2) calculation of critical Reynolds numbers for the first stationary bifurcation with straight and concave interfaces. The numerical simulations were performed for two aspect ratios $\Gamma=H/R=1$ and $\Gamma=1.2$. As there are no experimental data for this first bifurcation the benchmark results may be considered as a standard for different numerical codes.



Nine research groups participated in this effort and one of the most active participants was Prof. A. Zebib. The benchmark results for the onset of instability in 3D calculations revealed a large scattering and some participants withdrew their data. Prof. A. Zebib was curious for comparison his results with other once, though for this particular issue they were in the tail of final distribution. He was open for any discussion on the subject and he was one of the first, who calculated large Prandtl number liquids with deformed interface [2-3]. It would be difficult to overestimate his permanent help in the preparation of the manuscript [1].

An *experimental* benchmark for flows in liquid bridge with high Prandtl number fluids was carried out during Fourth International Marangoni Association, IMA-4 Congress, Noda, Japan, 2008. The goal of this study was to build an experimental database of benchmark test cases for the validation of numerical models. Experiments in liquid bridges are very sensitive to the experimental environment, geometry of particular set-up and physical properties of a test liquid. Furthermore, perfectly conducting rigid walls and, especially, boundary conditions at the free surface defined in numerical works are not always realized in experiments. The comparison of numerical results with a single experimental results promise to be reliable and useful for the validation of advanced numerical models. Silicone oil 5 cSt (Pr=68) was used as test fluid in the experimental benchmark. An experimental benchmark has been performed for liquid bridges with different aspect ratios and interface deformation.

Five research groups took part in the experimental benchmark which used three principally different types of experimental set-ups [4]. The stability diagram for deformed LB and $Pr \sim O(10^2)$ consists of two major branches. If the results of different groups for the same volume and aspect ratio belong to the same branch, they are in a good agreement. Scattering of the results falling to different branches can be large. Note that 'large' scattering of results does not mean a difference by an order of magnitude. In most distinctive cases, the measurements maximally differ by a factor 3.

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Buoyant-thermocapillary convection instability in rotating annular pools by linear stability analysis

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In a Czochralski melt pool, buoyancy force, thermocapillary force and pool rotation play important roles in the stability of melt convection. In our previous work [1], we conducted a linear stability analysis (LSA) and nonlinear numerical simulations to evaluate the effects of pool rotation on the stability of the basic axisymmetric thermocapillary flow in a rotating shallow annular pool of silicone oil under OG. However, there are few reports [2, 3] on the stability of buoyant-thermocapillary convection in shallow annular pools of silicon melt rotating system. In the present work, a series of LSA are conducted to investigate the effect of pool rotation and gravity level on the stability of thermocapillary convection of silicon melt. Figure 1 shows the model system of a differentially heated annular pool of silicon melt with an inner radius $R_i = 20$ mm, an outer radius $R_o = 40$ mm and depths d = 1 to 10 mm. The critical conditions (the critical Marangoni number (Ma_c) , critical azimuthal wave number (m_c) and critical phase velocity (ω_c) for the incipience of 3-D oscillatory flow are determined by means of LSA for both 0G and 1G conditions over a range of rotating rate (Ω) from 0 to 10 rpm. The results indicate Ma_c increases with increase in the pool rotation rate. LSA also provides us with the pattern of critical perturbation temperature. Figure 2 indicates the surface temperature patterns in rotating pools under 1G condition. These correspond to the hydrothermal wave (HTW) type instability. The HTW propagates in counter-clockwise direction in case of Fig.2(a) (Ω =0). However, as the pool rotation rate increases, pattern's phase velocity of propagation in the azimuthal direction is reduced and the spiral arms become rather straight (Fig.2(b) at Ω =2.5 rpm). Further increase in Ω changes the propagation direction and makes the pattern more complex (Fig.2(c) at Ω =6 rpm).



Fig.2 Temperature fluctuations on the surface of pool with depth d=1mm under 1G condition (a) in non-rotating pool (Ω =0), m_c =39, (b) at Ω =2.5 rpm, m_c =34, and (c) at Ω =6 rpm, m_c =34.

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Longwave Oscillatory Mode in Marangoni Convection

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We consider Marangoni convection in a layer of liquid heated from below. The thermal conductivity of the substrate is assumed small in comparison to the thermal conductivity of the liquid; the heat transfer from the upper deformable free surface is governed by the Newton's law of cooling. The monotonic mode of instability for this setup was studied within the linear stability framework [1] and in the weakly nonlinear approximation [2].

In contrast to all previous studies, we assume that the capillary number Ca is large and the Biot number B is small, but their product is finite. Also, we use non-traditional scaling, setting the wavenumber k the order of $B^{1/2}$. (The usual choice is $k \sim B^{1/4}$ [2].) Under these assumptions, we derive a set of nonlinear amplitude equations. Analysis of these equations shows existence of the oscillatory mode of instability. Existence of the oscillatory mode and its properties are well known in the situation when the liquid layer is heated from above [3]. However, to the best of our knowledge, the existence of this mode is not known when heating is from below.

The novel oscillatory mode emerges when the product B Ca is neither large nor small. Perhaps this (and/or the scalings) explains why this mode has not been found yet. The majority of the previous studies dealt with either case B = 0 (i.e., the heat flux at the liquid-substrate interface is fixed), or with the case $Ca \gg 1$ and $G \gg 1$ (i.e., the free surface is nondeformable).

We perform weakly nonlinear analysis within the set of amplitude equations. For the monotonic mode hexagonal patterns are selected and transcritical bifurcation takes place. For the oscillatory mode all patterns bifurcate supercritically in a certain domain of parameters. In this case there is a competition between Traveling Rolls and Traveling Rectangles.

We also carried out direct numerical simulation of the amplitude equations for the one-dimensional case (two-dimensional convective flows) and studied secondary instabilities of the branching solutions.

Details of this work can be found in Ref. [4]. The work is partially supported by joint grants of the Israel Ministry of Sciences (Grant 3-5799) and the Russian Foundation for Basic Research (Grant 09-01-92472).

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Different Types of Nonlinear Convective Oscillations in a Multilayer System Under the Joint Action of Buoyancy and Thermocapillary Effect

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Convective phenomena in fluid systems with an interface have been a subject of an extensive investigation (for a review, see [1].

There are two basic physical phenomena that produce convective instability in systems with an interface: buoyancy and thermocapillary effect. When heating is from below, the buoyancy instability generates the Rayleigh - Bénard convection [2], while the thermocapillary effect is the origin of the Marangoni - Bénard convection [3]. Buoyancy (a volume effect) is more important for relatively thick layers, while the thermocapillarity (an interfacial effect) plays the dominant role in the case of thin layers or under microgravity conditions. The case where buoyancy and thermocapillary effect act simultaneously, is the most typical.

During the past few decades, a new scientific direction of investigation, convection in multilayer systems, was developed [4].

The interfacial convection in *multilayer systems* is a widespread phenomenon that is of great importance in numerous branches of technology. Among the modern techniques requiring an investigation of convection in systems with many interfaces are liquid encapsulation crystal growth technique ([5], [6]) used in space labs missions, droplet-droplet coalescence processes, where Marangoni convection in the interdroplet film can considerably affect the coalescence time during extraction ([7]), and others.

In the present work, the nonlinear regimes of convection in a multilayer system where the Marangoni convection is produced by the upper interface and hence developed in the top layer and in the middle layer, while the buoyancy convection is produced in the bottom layer, are investigated. Two types of boundary conditions, the periodic boundary conditions on the lateral boundaries and rigid heat-insulated lateral walls, are considered.

The boundary value problem was solved by the finite-difference method. Nonlinear equations were approximated on a uniform mesh using a second-order approximation for the spatial coordinates. The Poisson equations were solved by the iterative Liebman successive over-relaxation method on each time step. The details of the numerical method can be found in the book by Simanovskii and Nepomnyashchy [1].

In the case of periodic boundary conditions, it is shown that the joint action of buoyancy and thermocapillary effect, leads to the development of a specific type of nonlinear traveling wave. The thermocapillary convection in the top and middle layers coexists with the buoyancy convection in the bottom layer. The maximum values of the stream function in all the layers are constant in time. The oscillatory flow keeps its periodicity even on a large distance from the linear stability boundary. The weakening of the thermocapillary effect leads to the development of the pulsating traveling wave in the system. For this flow, the maximum values of the stream function in all the layers are not constant in time and oscillate in a periodic way.

For rigid heat-insulated lateral walls various types of symmetric and asymmetric standing waves are obtained. Transitions between the motions with different spatial structures are investigated. It is found that for both, periodic boundary conditions and rigid heat-insulated lateral walls, the oscillatory motion is observed in a finite interval of the Grashof number values, between the stability regions of a quiescent

state and stationary convection.

The cavities with different lengths, are considered.

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Transition to chaos of thermocapillary convection

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Transition of fluid convection to chaos in dissipative dynamical systems is a subject of great interest for both its theoretical and practical aspects in the fluid mechanics. Extensive studies have shown that there are several routes of the buoyant natural convection to chaos depending on parameters of the dissipative dynamical systems such as the Rayleigh number, the Prandtl number and geometry aspect.

Another important type of natural convection is thermocapillary convection driven by the surface-tension gradient prominent in fluid systems with interface in the microgravity condition or in small-scaled terrestrial configurations (The relative importance of the gravity effect to the capillary effect is scaled by the static Bond number, $B_0 = \rho g l^2 / \sigma$, and the dynamic Bond number, $B_d = \sigma g l^2 / |\sigma_T| \Delta T$, the geometrical scale of the system in the terrestrial experiments, therefore, was significantly reduced to make the capillary effect dominant). The thermocapillary convection has become one of the fundamental subjects in the microgravity fluid physics and space fluid/heat management. However, most studies now available were focused on the onset of oscillatory thermocapillary convection, the initial regime of the route to chaos. A complete route to chaos in such a new sort of dissipative system is still an attractive open question, especially in the experimental study.

In present study, the route to chaos of the thermocapillary convection has been investigated experimentally and numerically. Several routes to chaos, e.g. period oscillatory convection to quasi-period oscillatory convection with 2 to 3 major frequencies, a series of successive period doubling bifurcations and their combination, of the thermocapillary flow is reported through the temperature measurements and the corresponding real time analysis of frequency spectra accomplished by Fast-Fourier-Transformation (FFT) or numerically.

Numerical study of convection induced by evaporation in cylindrical geometry

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The drying of binary fluid films including one volatile component appears in many industrial processes like distillation, painting, inking or packaging.

When drying a polymer/solvent solution, the temperature at the free surface decreases due to the vaporization latent heat and then the temperature field evolves from the surface to the bulk. The solvent evaporation also induces an increase of the polymer concentration. In the same time, the film thickness decreases and the solution viscosity increases with the polymer concentration. The fluid forces applied into volume (buoyancy) and on the free surface (surface tension) due to temperature and / or concentration gradients may generate a flow into the bulk.

The development and evolution of convective patterns, induced by the solvent evaporation in a Polyisobutylene/toluene solution, have been experimentally studied for a large range of initial thicknesses and viscosities [1]. The Lewis number (ratio of the thermal diffusivity to mass diffusivity) is very large (about 10^3), so that scales for the thermal and mass diffusion times are very different and the process is dominated successively by temperature and concentration gradients. Moreover it was shown that the lifetime of the convective patterns that were experimentally observed at the beginning of the drying is small compared to the whole drying time and is related to the duration of the transient thermal regime [1]. In this work only the thermal regime is considered. The unknowns are the velocity and temperature fields. In view of the experimental data, a simplified model has been developed for this thermal regime valid at the beginning of the drying only. The configuration is cylindrical (see Fig. 2). The thermal regime being short, the amount of evaporated solvent is small so that the thickness of the layer is assumed constant; the free surface is flat. The evaporation is only taken into account in the energy equation through the thermal equilibrium at the free surface between convective and diffusive fluxes and vaporization latent heat. The fluid is Newtonian and the physical properties are kept constant except the mass density in the buoyant term and the surface tension which depend linearly on the temperature. The flow and thermal behaviors in the solution are governed by four non-dimensional parameters (Ra, Ma, Bi, Pr) depending on two control parameters used in the experiments : the initial thickness and viscosity [1].

The Navier-Stokes, under Boussinesq assumption, and energy equations, with their associated boundary conditions, are solved using a 3D pseudo-spectral code. Computations are initialized with a zero velocity field and a perturbation on the temperature field.

In a first part, we evaluate the diffusive-convective thresholds and compare the 3D results with experimental [1] and 2D numerical simulations [2].

The basic solution of the problem is time-dependent so that it is not possible to define a definitive threshold value as in classical linear stability analysis because critical values are sensitive to initial conditions. This is inherent to the transient nature of the problem, and a criterium must be defined to characterize the onset of convection. In this study, we have chosen a condition based upon the Peclet number (Pe) which measures the ratio between the average fluid flow velocity and the thermal diffusion velocity. If Pe > 1 is verified, convection is considered as significant. To get a meaningful analysis it is important to analyze the sensitivity of the obtained thresholds to the initial condition. Using a statistical analysis, we showed that the impact of initial random distribution on the threshold values is very small. The impact of changes in perturbation magnitude is more important but much more smaller than the range of critical values obtained when varying the dimensionless parameters of the problem.

Thresholds maps as function of thickness and viscosity will be given, displaying the frontier between the domains respectively dominated by Bénard-Maragoni and Rayleigh-Bénard convection. These results are in good agreement with experimental and 2D results.

In a second part, we analyze the evolution of convective patterns for different initial perturbations. We first choose a local and structured perturbation at the free surface. Figure 1 shows, for a small aspect ratio, the temperature field on the free surface at a dimensionless time (t = 1) for four different initial disturbances (axisymmetric, asymmetric and symmetric (×2)). In all cases, there is a memory of the initial perturbation on the transient flow.

Then we choose an initial random perturbation on the full temperature field. As for deterministic disturbances each initial random perturbation leads to different patterns at a given time (see Fig. 2 (left)). The variable chosen to describe the evolution of the flow is the number of cells. A cell is defined by a local maximum of temperature. In order to estimate cell areas, a Voronoi diagram is used. Figure 2 (right) shows iso values of the temperature field at the free surface and its Voronoi diagram built on the cell centers. Overall, the Voronoi diagram seems to reproduce correctly the structure of the temperature field. With this description, we can evaluate each cell area and estimate the dispersion around the averaged size as a function of time.

Characterization of the convective patterns with Voronoi representation will be presented and compared to experimental results and temperature fields will be given in sevral configurations.



FIG. 1: (((Left :))) Temperature field at t = 1 (red/pink : hot, blue : cold); From left to right: perturbation in $(r = 0); (r = 0), (r = R/2, \theta = \pi/2); (r = 0), (r = R/2, \theta = \pm \pi/2); (r = 0), (r = R/2, \theta = 0, \pi, \pm \pi/2).$ (Ra = 150, Ma = 1950, Bi = 0.2, Pr = 36, A = R/e = 2.5)



FIG. 2: Left : Temperature field (red/pink : hot, blue : cold); Right : Iso-values of the temperature field a the free surface and Voronoi diagram built on cells centers. (Ra = 150, Ma = 1950, Bi = 0.2, Pr = 36, A = R/e = 10)

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Solutal Rayleigh-Bénard-Marangoni convection

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Solutal driven flow is studied for an horizontal binary solution submitted to solvent evaporation at the upper free surface. Solvent evaporation induces an increase of the solute concentration close to the free surface and solutal gradients may induce a convective motion driven by buoyancy and/or surface tension. This problem is studied numerically, using several assumptions deduced from previous experiments of polymer solution drying. The mathematical model of solutal Rayleigh-Bénard-Marangoni convection is based on a one-layer model. The cavity is two-dimensional. We restrict the analysis to the regime characterized by a constant solvent evaporative flux, i.e. before the strong decrease of evaporation flux observed for a typical polymer solution around $\varphi_{s-int} \sim 0.4$, where φ_{s-int} is the solvent volume fraction at the interface. In this concentration domain we can also assume that the mass diffusion coefficient is constant. Boussinesq approximation is used. The fluid is assumed newtonian and incompressible but with a viscosity that may depend strongly on the polymer concentration. The thickness is time dependent but the free surface is assumed flat.

The basic state is purely diffusive. Starting from a uniform volume fraction, the concentration field evolves due to solvent evaporation and the thickness decreases, so that there is no stationary basic state in this evaporation problem. Classical stability analysis is not suitable for this transient problem and a criterium based on the solutal Peclet number was chosen to characterize the presence of convection. Convection will be considered significant if, when the system is submitted to an initial perturbation on the concentration field, there is a time t where the perturbation is significantly amplified (Pe > 1). For a given initial perturbation, the critical Marangoni (or Rayleigh) number is the smallest one which satisfies the above criterium. First we analyze the sensitivity of the thresholds to the initial perturbations and show that the "blurring" effect inherent to any transient problem is small. We then investigate the stability of the system as a function of the solutal Rayleigh and Marangoni numbers, the evaporative flux, the Schmidt number and the initial concentration. Comparisons are made between solutions with a constant or concentration dependent viscosity.

Effect of Forced Flow of Ambient Gas upon Thermocapillary-driven Convection in Half-Zone Liquid Bridge

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The authors carry out an experimental study on the thermocapillary-driven flow in the half-zone liquid bridge of 2-cSt silicone oil exposed to a forced convection in the external shield coaxially wrapping the bridge. Effect of the ambient gas flow on the critical condition of the onset of the transition of the flow in the bridge from the two-dimensional steady flow to the three-dimensional oscillatory one after the onset are examined. Vortex formation around the liquid bridge due to the forced convection is described based on the observation from the side. The authors also indicate a relation between the critical condition in terms of the critical Marangoni number and the oscillation of the temperature inside the liquid bridge as a function of the velocity of the forced ambient-air flow.

Experimental apparatus is indicated in Fig. 1. The liquid bridge of 5 mm in diameter is completely wrapped by placing the external shield made of Pyrex® glass of 1.5 mm in thickness. The inner diameter of the external shield $D_{ES} = 2R_{ES}$ is of 25 mm. The external shield is hold by the shield holders made of aluminum. Each shield holder has four channels for inlet/outlet of the ambient gas flow. The direction of the external flow, that is, upward or downward, is able to be varied. A diffuser is placed right after the inlet and before the external shield to realize a uniform flow field inside the shield. The flow field in the external shield is described with an averaged velocity U_{avg} and non-dimensional Reynolds number $Re = U_{avg}L/v_{air}$, where Q is the controlled flow rate in the external shield, L the hydraulic diameter of the granular channel and v_{air} is the kinematic viscosity of the air. Test fluid for ambient gas is a dried air at 25 °C. The Reynolds number for the ambient gas flow is ranged as -100 < Re <100 in the present system. The ambient gas flow is visualized by adding an incense smoke illuminated with a sheet of a laser light (see Fig. 2).

Three-dimensional particle tracking velocimetry (3D-PTV) is also applied to detect the particle behavior inside the liquid bridge; the authors are especially focused on the particle behaviors near the free surface of the bridge to examine the effect of the mechanical stress by the forced flow of the ambient gas around the liquid bridge.

The authors will introduce the effect of the ambient gas flow on the flow field in the liquid bridge of various aspect ratio and volume ratio.



Fig. 1 Experimental apparatus. Case of upward forced convection in the external shield is described. In the case of downward convection, the diffuser is located upper part of the top rod, and the top channels become 'Inlets' and the bottoms 'Outlets.'



Fig. 2 Snapshot of ambient gas flow around the liquid bridge of 0.8 in volume ratio v/v_0 observed from the side through the external shield. Forced flow rate of the ambient gas is null (Reynolds number of ambient gas $Re_{AG} = 0$).

Thermocapillary-driven Flow in a Half-Zone Liquid Bridge Accompanying with its Dynamic Oscillation on 'Kibo' aboard the International Space Station (ISS)

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On-orbit long-duration fluid physics experiments on 'Kibo,' the Japanese Experimental Module, aboard the International Space Station (ISS) have been successfully carried out since August in 2008 [1]. Thermocapillary-driven flow in a half-zone liquid bridge of 30 mm in diameter and its critical condition of flow transition are examined. That series of experiments indicates an invaluable knowledge on the effects of the gravity and the size of the liquid bridge itself.

The authors pay their special attention to the flow field inside the liquid bridge of up to 4.0 in aspect ratio $\Gamma = H/R$, where H is the liquid bridge height (a distance between the rod surfaces) and R the liquid bridge radius. In the present study, effect of dynamic oscillation of the liquid bridge due to the g-jitter on the ISS on the flow field inside the bridge is also focused. Figure 1 indicates the side and top views of the liquid bridge of 5-cSt silicone oil under the temperature difference between the rods $\Delta T \sim 3.9$ K (non-dimensional Marangoni number Ma $\equiv \sigma_T \Delta T \cdot H/(\rho v \kappa) \sim 4.0 \times 10^4$, where σ_T (\equiv $\left|\partial\sigma/\partial T\right|$) is the temperature coefficient of the surface tension, ρ the density, v the kinematic viscosity, and κ the thermal diffusivity); (a) at a two-dimensional steady state right after imposing ΔT , and (b) at a fully developed three-dimensional oscillatory state. These images are obtained by integrating the successive frames (taken at 30 fps) for 80 s, thus the path line of the particles suspended in the liquid bridge are observed. One can clearly see the oscillating behavior of the flow in the bridge after the onset of the oscillation. Noted that these images were taken under a situation of almost no dynamic oscillation of the liquid bridge. During the experiment, the liquid bridge is exposed to the g-jitter, and sometimes exhibits a dynamic oscillation as predicted by Sanz & Diez for the isothermal case [2]. The authors will introduce the effect of the g-jitter and resultant dynamic oscillation of the liquid bridge on the flow field in the liquid bridge.



(a)

Fig. 1 Side and top views of liquid bridge of 5-cSt silicone oil under $\Delta T \sim 3.9$ K (Ma $\sim 4.0 \times 10^4$) (a) at a two-dimensional steady state right after imposing ΔT , and (b) at a fully developed three-dimensional oscillatory state. These images are obtained by integrating the successive frames (taken at 30 fps) for 80 s. The hot rod is located on the right-hand side of the side view, and the cold rod on the left.

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Precursor Film Formation ahead Macroscopic Contact Line of Droplet Spreading on Smooth Substrate

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Characteristics of wettability of a liquid on a solid substrate are generally discussed by considering static and dynamic contact angles. This is of course useful if the system concerned is large enough; that is, the apparent boundary of the droplet almost corresponds with the boundary line of solid-liquid-gas phases, which is called as 'macroscopic contact line.' It has been revealed after Hardy [1], on the other hand, that there exists a thin liquid film ahead the macroscopic contact line in wettable liquid-solid system even in the systems of $O(\mu m) - O(mm)$. This thin film is generally called as 'precursor film.' It is of great importance to understand the mechanism of the precursor film formation especially for ultra-fine control of such as transport processes and chemical reactions on a test substrate. This is the motivation of the present study.

It has been indicated that the precursor film consists of two major regions; 'adiabatic' and 'diffusive' precursor films after Joanny & de Gennes [2]. The existing length of the adiabatic precursor film, which is dominated by the fluid dynamics, on a smooth and isothermal substrate was predicted by Hervet & de Gennes [3], and was measured by a beautiful experiment by Kavehpour et al. [4] with a phase-shift interference microscopy. Little knowledge has been accumulated, however, on a very early stage of that formation process and on the diffusive precursor film that is governed by a molecular diffusion. In the present study, the formation process of the precursor film including the diffusive region at a very early stage of the droplet spreading on a smooth substrate is evaluated by applying a Brewster-angle microscopy (see Figs. 1 & 2) as well as a conventional laser interferometer. The authors extend their attention to the advancing process of the precursor film on inclined substrate.

In the present study, a droplet of 2-cSt silicone oil of $O(\mu)$ spreading on the silicon wafer substrate covered with SiO₂ is the system concerned. Through the experiments, the authors detect the precursor film including the diffusive region, whose total length is of about 1-2 mm ahead the macroscopic contact line. Typical example of spatio-temporal distribution of the spreading droplet on a horizontal substrate detected by the Brewster angle microscopy is shown in Fig. 3. This distribution is reconstructed by integrating the time series of the brightness data detected at a certain measuring point on a line perpendicular to the macroscopic contact line (shown in Fig. 2) in space. Time at t = 0 corresponds to the instance when the authors place a droplet on the test substrate, and position at x = 0 corresponds to the position where the center of the mass of the drop is placed. The boundary of the red



Fig. 1 Experimental apparatus (Brewsterangle microscopy). Droplet is placed on the smooth substrate. Whole system can be inclined without varying the incident angle of the laser light.



Fig. 2 Examples of (a) detected image with BAM, and (b) time series of a brightness of BAM image at a certain measuring point.

region in this figure corresponds to the macroscopic contact line of the spreading droplet. One can clearly see a vague region spreading ahead the macroscopic contact line as a function of time. This indicates the spreading behavior of the precursor film right after the droplet is placed on the substrate. Such detected lengths by our research are much longer than the prediction by Hervet & de Gennes [3] for the 'adiabatic' precursor film.

The authors indicate that tiny but distinct effect of the gravity on the precursor film formation. Figure 4 indicates temporal variations of the of precursor film length ahead the macroscopic of droplet on horizontal or inclined substrate. Angle indicates the attitude of the substrate. 'Front' and 'Rear' indicate the edges located lower and higher on the substrate, respectively. In the case of horizontal substrate, the precursor film length grows almost exponentially. The exponent is slightly smaller than 1/2, which corresponds to the exponent in the diffusive growth. The authors must indicate that the growth rate of the precursor film length is dependent on the attitude of the substrate. If the droplet spreads on the inclined substrate, the growth rate at the front of the droplet becomes slightly higher. That means, the growth of the diffusive region of the precursor film is significantly affected by the macroscopic spreading of the bulk droplet. Further research is surely needed to comprehend the mechanism of the formation and growth process of the precursor film including the diffusion-dominant region.

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Fig. 3 Example of spatio-temporal diagram of precursor film formation ahead the macroscopic contact line (M-CL) of a droplet.



Fig. 4 Temporal variations of the of precursor film length ahead M-CL of droplet on horizontal or inclined substrate. Angle indicates the attitude of the substrate. 'Front' and 'Rear' indicate the edges located lower and higher on the substrate, respectively.

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Evaporative Instability in Binary Mixtures

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Evaporative convection plays a critical role in engineering applications such as film coating, the space processing of glass, paint drying and most of those applications involve mixtures of fluids.

To understand why evaporation leads to an instability consider the first figure where fluid flow is omitted. When the interface is perturbed, a trough comes closer to the heat source causing a higher evaporation rate there than at a crest. Therefore a trough will grow deeper into the liquid destabilizing the interface. This behavior is like a cell moving in the presence of a nutrient gradient. In other words the trough becomes more pronounced as the gradient in temperature increases.



Figure 1 The thermal gradient argument for the generation of the instability

With fluid dynamics in the system, note that the system is closed so the liquid which evaporates at the trough has to condense at the crest. In other words, we can think of a crest as a local condensation point and the trough as the local evaporation point (Figure 2). As a result of evaporation there will be an upwards flow at troughs and a downward flow at the crest. This cycle results in rolls of circulating fluid, or convective "cells," that form cellular flow patterns.



Figure 2 Fluid flow in the presence of evaporation

The flow is much more dominant in gas phase because of the low density of the gas. The flow from troughs carries hot fluid to cold crests as a consequence stabilizes the interface.

In this work the linear stability analysis is used to investigate the effect of evaporation and concentration on the natural convection in binary mixture. The liquid and vapor mixtures are

assumed to be ideal mixtures and in equilibrium. In the numerical calculations the dynamics of both phases are taken into account and the interface is allowed to deflect. The inclusion of vapor phase dynamics is pivotal as the vapor plays a dominant role in the stabilization of the surface.

Experimental investigation of Marangoni effect in hexanolwater system^{*}

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Marangoni effect, induced by interfacial tension gradient generated in the process of heat or mass transfer, exists broadly in industrial processes. Marangoni convection can change mass/heat transfer efficiency evidently. Agble et al.^[1] found that ionic surfactant can intensify the Marangoni convection in binary systems. On the other hand, most of researches indicated that surfactants would dampen interfacial convection in general. To our knowledge, few experiments about surfactants changing the solute Marangoni convection are reported. It is necessary to carry out more experiments to investigate the Marangoni effect and the influence of surfactants.

In this work, mass transfer data, dynamical interfacial tension and interfacial phenomena of a single stagnant drop were obtained for the hexanol (drop)-water (continuous phase) system both with and without surfactant in the continuous phase. The surfactant chosen is ionic surfactant sodium dodecyl sulphate (SDS) and non-ionic surfactant Triton X100. During the saturation process, weak interfacial instabilities can be observed for this system by a Schiliren optical system. As the concentration of SDS was higher than a certain value, the interfacial instability during the mass transfer process of water saturated with hexanol was intensified. For mutual saturated binary system, we have captured interfacial instabilities for both cases with ionic surfactant and non-ionic surfactant at a high enough concentration.

For the ternary system with acetic acid, Marangoni effect can be induced by the solute transport of acetic acid from the hexanol drop to water. The higher the initial solute concentration is, the more intensive Marangoni instabilities are, and in the process of mass transfer Marangoni convection died away with the elapsed time. For the system with surfactant, at the beginning of mass transfer period, Triton X100 and SDS dampened the solute Marangoni convection. However, as shown in Fig.1 the presence of SDS introduced an intensified mode of interfacial instability accompanied with drop oscillation at certain moment in the process of mass transfer, and higher SDS content in the continuous phase is needed for the case with higher solute concentration to induce this phenomenon. In the case with Triton X100, it can trigger another different mode at the late period of mass transfer only for the surfactant content higher than CMC, which seems like smoke rising up along the interface and no drop oscillation was observed. Analyzing the dynamical interfacial tension of the system with surfactant, we found that the interfacial instabilities occurred at certain time and seems in good agreement with the situation that interfacial tension sharply decreasing at some moment, which shows surfactant adsorption dynamics played an important role on the interfacial phenomena's variation. The addition of Triton X100 reduced mass transfer rate significantly (Fig.2). However, SDS reduced mass transfer rate only at high solute

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concentration, while high content of SDS can enhance mass transfer rate in the case of low solute concentration (Fig.3). The effect of surfactant on interfacial phenomena that we found in the ternary system and the interesting effect of SDS on mass transfer may give us a hint to use surfactant to manipulate interfacial phenomena for enhancing the mass transfer process.



(a) Without surfactant, 8.8s (roll cells)



(b) SDS 0.1g/L, 37.8s (ripples with oscillation)



(c) Triton X100 0.5g/L, 60s (smoke rising)

Fig. 1. Interfacial Instabilities of a hexanol Drop in Water with interphase mass transfer (Initial concentration of acetic acid in the hexanol drop is 3 g/L)



Fig. 2. Influence of Triton X100 on mass transfer efficiency ($C_{\text{acetic acid}}=3.7 \text{ g/L}$)



Fig. 3. Influence of SDS on mass transfer efficiency. ($C_{acetic acid}=3 \text{ g/L}$)

Keywords: Marangoni Effect, Mass Transfer, Surfactant, Stagnant drop [1] Agble, D. and Mendes-Tatsis, M.A., *Int. J. Heat Mass Transfer*, 43(6), 1025-1034, 2000.

Thermocapillary-Driven Flow in Free Liquid Film Formed in A Rectangular Hole with Temperature Gradient

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Thermocapillary-driven flow in a free rectangular liquid film under a temperature gradient parallel to the free surfaces is examined by experimental approaches. Target geometry is illustrated in Fig. 1. A liquid film was formed in the rectangular hole of center of an aluminum plate. In the president geometry, two kinds of aspect ratios are defined; that is, $\Gamma_x = L_z/L_x$, $\Gamma_y = L_x/d$. One end wall is maintained at a temperature of T_h , and the other at T_c ($T_h > T_c$). Thus the film is exposed to a temperature difference $\Delta T = T_h - T_c$. Intensity of the thermocapillary effect imposed to the film is described by non-dimensional Marangoni number Ma $\equiv \sigma_T(\Delta T/L_x)d^2/\rho\nu\kappa$, where σ_T is the absolute value of the temperature coefficient of the surface tension ($\equiv |\partial\sigma/\partial T|$), ρ the density, ν the kinematic viscosity, κ the thermal diffusivity. The Marangoni number can be described as the product of the thermocapillary Reynolds number Re_{σ} and Prandtl number Pr. The temperature difference was measured at 2 mm apart from each end wall by using T/Cs. Silicone oil of 2 and 5 cSt are employed as examined fluids, and golden coated acrylic spheres of 15 μ m in diameter are chosen as tracer particles. The authors examine free liquid films with different thickness d in the range of 0.2 mm to 1.0 mm.

Under a small temperature difference, a two-dimensional basic flow appears; the basic flow is double-layered, that is, the fluid flows from a hot wall to a cool wall on free surfaces and returns in the middle region of the film. The flow exhibits a transition to an 'internal' oscillatory flow, that is, a three-dimensional flow with characteristic particle behaviors inside the film by increasing the temperature difference, although the flow over the free surface never exhibits an oscillatory manner. This flow pattern consists of steady-state vortices in spirals are observed near the hot wall. This scenario of the flow transition in the liquid film is indicated in Fig. 2

The authors find out that the flow exhibits a significantly different bifurcation in the case of rather large Γ_y (see Fig. 3). The flow never exhibits a double-layered basic flow even with a small temperature difference, but exhibits a single-layered cellar flow consisting of counter-vortices. The authors will introduce such unique bifurcations in the thin free liquid film and their occurring condition. The authors will also discuss the hydrothermal-wave instability emerged in the thin film.



Fig. 1 Target geometry; a free liquid film is sustained in a rectangular hole of d in thickness. One end wall is heated and the other is cooled to realize a designated ΔT along the free surfaces of the film.



Cold-end wall (a) 2 mm (b)

Fig. 2 Typical example of flow transition observed from above in the case of rather small Γ_y ; ($\Gamma_x = 2.0$, $\Gamma_y = 3.3$ and d = 0.6 mm). (a) $\Delta T = 4.7$ K (Ma = 2.2×10^2), (b) $\Delta T = 31.5$ K (Ma = 1.5×10^3). The images are obtained by exposed for 1.0 s.

Fig. 3 Typical example of flow transition observed from above in the case of rather large Γ_y ; ($\Gamma_x = 2.0$, $\Gamma_y = 10$ and d = 0.2 mm). (a) $\Delta T = 6.8$ K (Ma = 26), (b) $\Delta T = 17.8$ K (Ma = 68). The images are obtained by exposed for 1.0 s.

Linear Stability and Nonlinear Evolution of Marangoni Convection Structures due to Localised Axisymmetrical Inhomogeneity of Surface Tension

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The results will be presented of analysis of steady states, their stability and nonlinear development of disturbances for Marangoni convection arising in thin liquid layer with inhomogeneous source of surface tension, caused by axisymmetrical localized heating of liquid near the surface, accompanied in general case by its temporal modulation.

Only Marangoni convection is treated, assuming that liquid layer is thin enough, so dynamic Bond number is small and the effect of thermogravitational convection could be neglected. Further assumptions are that the heat inhomogeneity is weak enough, and is superimposed on vertical temperature gradient, corresponding to homogeneous heating from below. The vertical to horizontal scales ratio is considered as small parameter, which allows to deduce the long –wave approximation of Marangoni convection near the threshold for thermo-capillary convection excitation at homogeneous heating [1]. As the result initial three-dimensional hydrodynamical problem reduces to the set of two-dimensional nonlinear amplitude equations for the weak disturbances of temperature, vorticity and surface deformation. This approach generalizes one known for the thermocapillary convection in a liquid layer at uniform heating [2] and allows to capture the basic features of the flows and surface deformation, observed in the laboratory experiments with localized heat source induced by laser or incoherent radiation [1].

After the expansion of the second order, the mathematical model takes the form of the system of nonlinear differential equations for two-dimensional amplitudes of functions $\Phi(x, y)$, $\Psi(x, y)$, and H(x, y), characterizing the deviation of the temperature field from the distribution ensuring a vertical gradient in equilibrium, the vorticity, and the free surface deformation, respectively:

$$\partial_{t}\Phi + \nabla\Phi\cdot\nabla\times(\vec{e}_{z}\Psi) + \frac{1}{15}\nabla^{4}\Phi - \nabla^{2}H - \frac{48}{35}\nabla\cdot\left(\left|\nabla\Phi\right|^{2}\nabla\Phi\right) + 2\nabla\cdot(H\nabla\Phi) + \\ + \left(\frac{1}{10} + \frac{1}{5P}\right)\nabla\cdot\left(\nabla^{2}\Phi\nabla\Phi\right) + \left(\frac{3}{5} + \frac{1}{10P}\right)\nabla^{2}\left|\nabla\Phi\right|^{2} - \nabla\cdot(q\nabla\Phi) + \frac{1}{2}\nabla^{2}q + \zeta qH = 0.$$

$$\vec{e}_{z}\nabla^{2}\Psi = \frac{312}{35P}\nabla(\nabla^{2}\Phi)\times\nabla\Phi - 24\nabla H\times\nabla\Phi; \qquad (1)$$

$$\delta\nabla^{2}H - \nabla^{4}H = -c \nabla^{2}\Phi.$$

The model is valid near the critical value of Marangoni number of thermo-capillary convection excitation $Ma_{cr} = 48$, which corresponds to the threshold to the long-wave instability for the limiting case of a uniformly heated layer with a thermally insulated bottom. The dimensionless parameters of model are Prandtl number P, parameters c, δ , connected with capillary number Ca and Galileo number G ($c = 72/Ca; \delta = G/Ca$) and ζ , characterizing vertical inhomogeneity of heat flux. Values $c = 0, \delta = 0$ corresponds to the case of non-deformable upper surface. $\zeta = 0$ means that $q_z(z) = const$, where inhomogeneous heat flux is assigned by the formula $Q(x, y, z, t) = q(\xi) \cdot q_z(z) \cdot q_t(t)$, where the first two cofactors describe respectively the spatial inhomogeneity of heat flux in the horizontal plane and on the vertical direction, and describes the modulation of heat flux with the amplitude ε and frequency ω . In the present study the axysimmetrical stepwise form of inhomogeneous heat flux $q(\xi) = q(r)$ is considered, parametrized by α, β being characteristic values inside and outside hot spot respectively.
One-dimensional axisymmetrical solutions were examined and their dependence on the parameters of the problem in the limit case of the non-deformable free surface and with account of its deformations was investigated. The presence of the multiple solutions at the fixed parameters is shown, differing from each other in terms of the number of local extrema (Fig. 1).

At nonzero amplitude of heat flux modulation, the basic localized solutions of the system of amplitude equations, which preserve the spatial symmetry of inhomogeneous heat flux and change in time with the same period, are determined, their dependence on amplitude and frequency of modulation, of parameters of the heat flux inhomogeneity and of physical parameters of system is studied numerically on the basis the Galerkin method with the transformation of space coordinate $\xi = \tanh(x)$, that makes it possible to reduce infinite function domain to a finite one.

The analysis of linear stability of the periodic ground state was carried out with the use of Floquet's method. The solutions of non-linear 1D problems was treated by the Galerkin method with basic rational-linear functions, which take finite value at the central point and approach to zero at infinity. The confirmation of convergence of numerical solution was obtained with an increase in the number of basic functions.



Fig.1 Dependence of amplitudes of temperature and surface deformation from the heat transfer in the center of the hot spot (Parameters: $\beta = 0.1$, Pr=13, $\zeta = 0$, c=0.1, $\delta = 0.1$, R=1).

The analysis of stability of the steady oscillatory ground states to the two-dimensional disturbances and the study of their nonlinear development is executed on the basis of solution of complete 2D problem (1-3) by the pseudo-spectral method. Numerical results show that steady axisymmetric periodic solutions could realize in the specific interval of the variation in the parameters, outside which disturbances with the same (synchronous) or with the doubled period (with the half frequency, half-integral) with different azimuthal wavenumbers can develop. Comparison of results with available experimental data and with numerics for limit cases of unmodulated heat source [1] and homogeneous modulated heating [3] was performed.

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Shapes of receding contact lines: from sliding drops to immersion lithography

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Instabilities of receding contact lines often occur after the formation of a corner with a very sharp tip. For small capillary numbers, the dewetting contact line can stabilize itself by changing its shape. With increasing capillary number it becomes more cornered ('v'-shaped, Fig. 1a) while the dynamic contact angle decreases according to the Cox-Voinov law. Reaching the critical capillary number this shape changing mechanism fails, resulting in instabilities, droplet deposition, rivulets or breakup of the contact line. This mechanism has been observed in different partially wetting systems, from zigzagging patterns in the withdrawal of a plate from a liquid bath [1] to drops sliding down an inclined plane [2].

Similar dewetting corners are encountered in the technology of Immersion Lithography [3], where water is put between the lens and the silicon wafer to increase the optical resolution (Fig. 1b). In this paper we aim to compare corners appearing in immersion lithography to those at the tail of sliding drops. The immersion lithography data is obtained from high speed recordings of a liquid meniscus on a turntable setup. Water is continuously supplied and extracted through two concentric needles, holding the drop at fixed position. The turntable is used to move the wafer, while the shape of the meniscus is imaged from the side and from below. In a similar way we extract shapes of silicon oil drops sliding down an incline. We measure the dynamic receding contact angle, sharpness (curvature) of the tip and the opening angle of the cornered tail as a function of contact line speed. It is shown that these quantities display very similar behaviors in the two systems. In addition, the results agree well with predictions by a lubrication model for cornered contact lines [4], hinting at a universal structure of dewetting corners.



Fig. 1: a) Shape changes of silicon oil drops sliding down an inclined plane for increasing velocity. Little droplets are deposited above a critical speed. b) Similar structures occur in immersion lithography.

- [1]
- [2]
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Simulating Multiphase Flows in Porous Media

with High Order CE/SE Method

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In this paper an updated second-order accuracy CE/SE (space-time conservation element and solution element) method is proposed. The SEs (Solution Elements) and CEs (Conservation Elements) [1] are demonstrated in Fig.1. Combining the Level-set method [2], we adopted the second-order accuracy CE/SE to calculate unsteady multi-phase incompressible flows in porous medium. The numerical simulations of three benchmark problems have been completed.



(a) SEs (Solution Elements)(b) CEs (Conservation Elements)Fig. 1 Mesh construction of the updated CE/SE method

1. Transformation and motion of droplets by gravity in porous media

The results of transformation and motion of droplets by gravity in porous media put into Fig. 2. From Fig.2 we can find that the second-order CE/SE method performs well agreement with the results by FVM (Finite Volume Method) [3]. But the interfaces calculated by CE/SE are more accurate than that of FVM. It demonstrated that there is no discontinuity occurrence in the final deformed droplet calculated by CE/SE.



Fig. 2 Deformation and motion of droplet (left: present CE/SE; Right: FVM)

2. Dual lid-driven cavity flows in porous medium

The lid-driven cavity flow [5] has become a benchmark problem for the evaluation of various numerical methods. Here we set up a new model for dual-layer lid-driven cavity flows in porous medium. A light fluid is laid on the heavy fluid. The flow is driven by a lid with the velocity of 1.0 on the up boundary. We calculated the flows in the dual cavity for the porosity of 0.3 and Darcy number of 100. Fig.5, Fig.6 and Fig.7 show the stream lines and the

interface for Darcy number of 10^6 , 0.25 and 0.21, respectively. The results show that the interface becomes as a shear layer. As the Darcy number varies from 0.01 to 10^6 , the interface changes from horizontal lines to complex curly lines. The number of eddies and vortex increase and their strength are enhance, as expected. From all results mentioned above the application of update high-order CE/SE method is successfully extended. It is a effective and reliable approach to calculate multiphase porous flow with interface, which can be treated accurately. CE/SE method has following advantages: it is easy to implement and programming, higher accuracy and efficiency.



Fig. 5 Revolution of the interface and stream lines at time of (a) 50, (b) 100, (c) 150, (d) 200, respectively, for Darcy number 10^6 and viscosity ratio 50



Fig. 6 Revolution of the interface and stream lines at time of (a) 50, (b) 100, (c) 150, (d) 200, respectively, for Darcy number 0.25 and viscosity ratio 50



Fig. 7 Revolution of the interface and stream lines at time of (a) 50, (b) 100, (c) 150, (d) 200, respectively, for Darcy number 0.01 and viscosity ratio 50

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A Novel Method of Simulating Marangoni Phenomena

-Perturbational Finite Volume Method-

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In this paper a perturbation Finite Volume Method (PFV) has been introduced to simulate Marangoni phenomena. The elemental thought of PFV method is: the perturbation concept in Mechanics is expended into numerical perturbation in computational mathematics. The PFV method adopts the first-order upwind difference scheme or second-order central difference scheme for the convective-diffusion integral equation as staring point. The mass flux through each face of the control volume (CV) and the source term are modified and expanded into a power-series of the grid spacing. Then, the Coefficients of the power-series are determined with the use of space splitting technique and the relations between the convection and diffusion flux. After some operations, the PFV Scheme for the convective-diffusion integral equation with source term can be written as:





Where φ represents any transported variable. ρ is density, **u** is velocity vector, Γ is diffusion coefficient and *q* is source term. **n** is unit normal vector of the CV's surface, S and Ω are surface and volume of the CV. A typical CV's face labeled "j-face" is shown in Fig.1.

P is CV's center and jP is adjacent CV's center. The second-order central difference scheme of the integral equation (1) can be written <u>as:</u>

Fig.1 A typical CV's face and adjacent nodes

 $\sum_{j=1}^{J} \left\{ \left[\mu \frac{\vec{d}_{j} \cdot \vec{S}_{j}}{\left| \vec{d}_{j} \right|^{2}} - (1 - \delta) m_{j} \right] \phi_{jp} - \left[\mu \frac{\vec{d}_{j} \cdot \vec{S}_{j}}{\left| \vec{d}_{j} \right|^{2}} + \delta m_{j} \right] \phi_{p} \right\} + q_{p} \Omega_{p} = 0 \quad (2)$

Where m_j is the mass flux through the j-face, j=1,2,...,J. For 2-D problem, J=3 and 4 indicate the triangle and Cartesian grids respectively. φ_p is the value of the φ at the node P, φ_{jp} is the value of the φ at node jP. \mathbf{d}_j is the distance vector linking two central nodes P and jP with the direction from P to JP. $\delta |\mathbf{d}_j|$ is the distance from j-face to node jP. \mathbf{S}_j is the area vector of the j-face and its direction agrees with the outer of the face.

By using the spaces splitting technique the PFV scheme can be obtained as follows:

$$\sum_{j=1}^{j} \left\{ \left[\frac{\mu}{1 + \sum_{n=1}^{j} A_n d^n} \frac{\overrightarrow{d_j} \cdot \overrightarrow{S_j}}{\left| \overrightarrow{d_j} \right|^2} - (1 - \delta) m_j \right] \phi_{jp} - \left[\frac{\mu}{1 + \sum_{n=1}^{j} A_n d^n} \frac{\overrightarrow{d_j} \cdot \overrightarrow{S_j}}{\left| \overrightarrow{d_j} \right|^2} + \delta m_j \right] \phi_p \right\} + q_p \Omega_p = 0 \quad (3)$$

In order to certificate the accuracy and efficiency of the PFV scheme, a dam break flow is simulated by using PFV scheme coupled with the particle Level Set method. The numerical results put into Fig.2 and Fig.3. All show that the PFV scheme can simulate successfully the dam break flow of the two-phase fluids with interface.



Then we adopt PFV scheme with Particle-Level Set method to simulate numerically Marangoni phenomena of liquid-liquid extraction process induced from. The results show in Fig.4a, Fig.4b and Fig.6. The Marangoni phenomenon have been occurred in Fig.4a and no Marangoni phenomenon is occurred in Fig.4b.

The numerical results mentioned above show that the PFV scheme can simulate the Marangoni Phenomena successfully. It proves that the PFV method is an efficient and reliable way to simulate numerically Marangoni phenomena induced from the surface tension of two-phase liquid-liquid extraction process. It can be widely applied to numerical simulation of Chemical and oil engineering and other two-phase flow engineering.





Fig.4a Marangoni phenomenon Fig.4b no Marangoni phenomenon



Fig.5 The developing process of Marangoni phenomenon.

Linear spatiotemporal instability analysis of ice growth under a falling water film

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Interesting wave patterns often form in nature, such as wavy growing icicles that are covered with a thin layer of falling water film, ripples on stalactites in caves, and wave patterns on limestone deposits near hotsprings. Detailed observation of growing icicles shows the ripples is growing upstream. These ripples are associated with phase change, flow of liquid, deformation of the free surface, evaporation and heat exchange at the free surface. In this study, we focus on the problem coming from the ripples on icicles.

Linear spatiotemporal stability analysis is conducted about ice growth on an inclined plane under a falling water film. The evaporation on the free surface is neglected and an isothermal boundary condition is applied on the free surface. The full system of linear stability equations is solved by using the Chebyshev collocation method.

Three different instability modes, named shear mode, surface mode and ice mode are recognized. We present a detailed analysis of the ice mode behavior under the influence of the surface tension, Stefen number, initial ice height and inline angle of the plane, on the neutral curves and critical parameters of the ice mode. Furthermore, boundary curves between absolute and convective instabilities (AI/CI) are calculated and analyzed.



Fig 1. Schematic of the water film falling on ice plane. Fig 2. AI/CI boundary curve with solid line and critical Reynolds number curves with dashed line for the downstream branch and dash-dotted line for the upstream branch as a function of the inclined angle.

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Micrometric particle collection by a moving triple contact line

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The Lotus flower which is revered as a symbol of purity in Asian religions, rises from muddy waters and stays clean and untouched by dirt, organisms, and pollutants. This property comes from the Lotus leaf "self-cleaning" surface which is hydrophobic and rough (see, e.g., [1, 2]). Self-cleaning efficiency of a superhydrophobic surface is directly associated with the amount of particles left on the pathway of a drop when the latter is moved on a substrate on which particles are deposited. In order to improve the self-cleaning efficiency one needs, of course, to understand the particle-liquid collision/collection mechanism at the contact line when the drop rolls over the particles.

One simple way of studying particle collection and their interaction with the triple contact line is to work with capillary tubes on which walls particles are deposited (Fig. 1). A capillary tube partially filled with a liquid presents a meniscus at the air-liquid interface. This configuration has the advantage of having axi-symmetric advancing or recessing contact angles if the liquid is withdrawn or injected into the tube with a syringe. Furthermore, the velocity of the moving interface can be easily controlled by placing the syringe on a syringe pump (Fig. 1).

Here, we report on an experimental work where $20\mu m$ particles are collected by a moving waterair interface in a capillary tube of $288\mu m$ (see setup in Fig. 1). The results show that particles usually cluster at the interface and move with it. However, they can affect the contact line motion if their number becomes large. In that case, the interface shows a stick-slip behavior in its motion. For very large quantity of particles at the interface, clusters of particles are left behind the interface. This can occur at regular basis and form a pattern if there is an evenly distribution of particles in the capillary tube prior to their collection. Fig. 2 shows a typical picture of pattern that forms when the interface moves over evenly deposited particles on a capillary tube wall. A simple theoretical model based on the Coulomb friction of the particles on the walls is given for the stick-slip behavior of interface.



FIG. 1: Skecth of the experimental setup for studying particle collection by a moving contact line.



FIG. 2: Typical pattern formed by clusters of particles left behind the interface.

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