Nonlinear stability of buoyant-thermocapillary flows in two-layer systems with a temperature gradient along the interface

A. A. Nepomnyashchy, I. B. Simanovskii

Department of Mathematics, Technion - Israel Institute of Technology, 32000, Haifa, Israel

Abstract

Results of nonlinear simulations of convective flows in two-layer systems on different scales under the action of a temperature gradient along the interface, are presented. Both purely thermocapillary flows and buoyant-thermocapillary flows are considered. Also, the nonlinear development of the instability in ultra-thin films caused by intermolecular forces, is investigated.

1 Introduction

The stability of convective flows in systems with an interface has been the subject of an extensive investigation. Several classes of instabilities have been found by means of the linear stability theory for purely thermocapillary flows [1]-[4] and for buoyant-thermocapillary flows [5]-[8]. For the most typical kind of instability, hydrothermal instability, the appearance of oblique waves moving upstream has been predicted by theory and justified in experiments [9]-[11]. However, two-dimensional waves moving downstream have also been observed in experiments [12]. The change of the direction of waves propagation can be caused by the influence of buoyancy [5].

Most of the investigations have been fulfilled for a sole liquid layer with a free surface, i.e., in the framework of the one-layer approach. In the present paper, we consider thermocapillary flows in two-layer systems. The paper is organized as follows. In Sec. 1, we consider the stability of purely thermocapillary flows. In Sec. 2, the influence of buoyancy is studied. Sec. 3 is devoted to the investigation of nonlinear stability of ultra-thin two-layer films flowing under the action of thermocapillary stresses. Sec. 4 contains some concluding remarks.

2 Thermocapillary flows in two-layer systems

In the following section, we will consider thermocapillary flows in a two-fluid system. We denote the variables of the bottom layer by subscript 1, and the variables of the top layer by subscript 2. Let the space between two parallel rigid plates $z = -a_1$ and $z = a_2$ be filled by two immiscible viscous fluids. The temperature on these plates is fixed in the following way: $T(x, y, -a_1) = Ax + \Theta$, $T(x, y, a_2) = Ax$. It is assumed that the interfacial tension coefficient σ decreases linearly with temperature: $\sigma = \sigma_0 - \alpha T$. The buoyancy force is neglected. In the present section, the interface is assumed to be a plane: $z = 0$.

The complete system of nonlinear equations can be written in the following dimensionless form:

$$\frac{\partial \mathbf{v}_1}{\partial t} + (\mathbf{v}_1 \cdot \nabla)\mathbf{v}_1 = -\nabla p_1 + \Delta \mathbf{v}_1, \tag{1}$$

$$\frac{\partial T_1}{\partial t} + \mathbf{v}_1 \cdot \nabla T_1 = \frac{1}{P}\Delta T_1,\ \nabla \cdot \mathbf{v}_1 = 0;$$

$$\frac{\partial \mathbf{v}_2}{\partial t} + (\mathbf{v}_2 \cdot \nabla)\mathbf{v}_2 = -\rho\nabla p_2 + \frac{1}{\nu}\Delta \mathbf{v}_2, \tag{2}$$

$$\frac{\partial T_2}{\partial t} + \mathbf{v}_2 \cdot \nabla T_2 = \frac{1}{\chi P}\Delta T_2,\ \nabla \cdot \mathbf{v}_2 = 0,$$

where $\nu = \nu_1/\nu_2$, $\rho = \rho_1/\rho_2$, $\chi = \chi_1/\chi_2$ are ratios of kinematic viscosities, densities and thermal diffusivities of fluids, $P = \nu_1/\chi_1$ is the Prandtl number of the bottom fluid.

On the rigid horizontal plates, the following boundary conditions are used:

$$z = -1: \ \mathbf{v}_1 = 0,\ T_1 = x, \tag{3}$$

$$z = a: \ \mathbf{v}_2 = 0,\ T_2 = x - b, \tag{4}$$

where parameter $b = \Theta/Aa_1$ describes the relation between the characteristic vertical and horizontal temperature differences. At the interface, the normal components of the velocity vanish:

$$z = 0: \ v_{z1} = v_{z2} = 0; \tag{5}$$

and the continuity conditions for the tangential components of the velocity

$$z = 0: \ v_{x1} = v_{x2},\ v_{y1} = v_{y2}, \tag{6}$$

for the tangential stresses

$$z = 0: \ \eta\frac{\partial v_{x1}}{\partial z} = \frac{\partial v_{x2}}{\partial z} - \frac{M\eta}{P}\frac{\partial T_1}{\partial x},\ \eta\frac{\partial v_{y1}}{\partial z} = \frac{\partial v_{y2}}{\partial z} - \frac{M\eta}{P}\frac{\partial T_1}{\partial y}, \tag{7}$$

for the temperature

$$T_1 = T_2, \tag{8}$$

and for the heat fluxes

$$\kappa\frac{\partial T_1}{\partial z} = \frac{\partial T_2}{\partial z} \tag{9}$$

are fulfilled. Here, $\eta = \eta_1/\eta_2$, $\kappa = \kappa_1/\kappa_2$ are ratios of dynamic viscosities and heat conductivities of fluids, correspondingly, $M = \alpha A a_1^2/\eta_1\chi_1$ is the Marangoni number.

In the limit of an infinite layer, it is necessary to impose some additional conditions determining the pressure gradients in the system. If the flow occurs in a channel that connects two vessels kept under the same pressure, the mean longitudinal pressure gradient in the system is zero. The corresponding thermocapillary flow is usually called "linear flow" [4]. In the case of a closed cavity, the mean longitudinal flux of fluid is zero, so that the "return flow" occurs which is characterized by a nonzero longitudinal pressure gradient. In the latter case,

$$\int_{-1}^{0} dz\, U_1^{(0)}(z) = 0, \int_{0}^{a} dz\, U_2^{(0)}(z) = 0. \tag{10}$$

The calculation of stationary parallel flow profiles of both types has been carried out in [13], [14]. The linear stability of a two-layer return flow has been investigated in [14]. For relatively small values of b and large values of M, the excitation of inclined hydrothermal waves has been predicted. These waves move in the opposite direction to that of the flow at the interface. For relatively large values of b and small values of M, the theory predicts the appearance of stationary convective rolls due to Pearson's instability. The axes of rolls are ordered by the thermocapillary flow along the direction of the imposed horizontal temperature gradient ("spiral flow"). For intermediate values of M, the convective rolls are ordered *across* the direction of the horizontal temperature gradient, and they are drifted by the thermocapillary flow. Unlike the hydrothermal waves, the drifted rolls move in the same direction as the flow at the interface.

The nonlinear simulations have justified these predictions [14].

3 Buoyancy-thermocapillary convection in two-layer systems

In the present section, we include the action of buoyancy. We consider only the case of a horizontal temperature gradient imposed in the direction opposite to that of the axis x.

Recently, Madruga et al. [15], [16] studied the linear stability of two superposed horizontal liquid layers bounded by two solid planes and subjected to a horizontal temperature gradient. The analysis has revealed a variety of instability modes. Specifically, for the system 5cS silicone oil - HT70, the analysis predicts a change in the direction of the wave propagation with the growth of the ratio of the layers thicknesses.

In the present section, we describe results of nonlinear simulations of the wavy convective regimes for the above-mentioned system of liquids [17].

For periodic lateral boundary conditions, there is a good coincidence between the predictions of the linear theory and the numerical results. Specifically, the direction of the wave propagation is changed with the growth of M (see Fig. 1).

The diagram of regimes is shown in Fig.2 (note that the Marangoni number and the Grashof number are defined through the parameters of the top liquid).

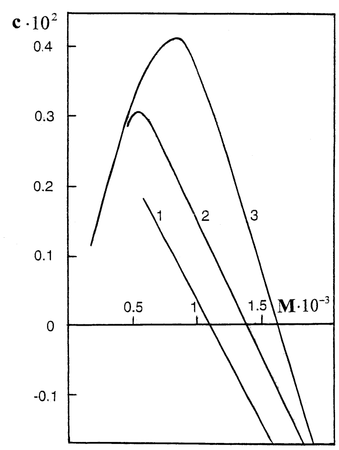

Figure 1: Dependence of the wave velocity c on the Marangoni number M for $G = 0$ (line 1), 1.5 (line 2) and 4 (line 3).

In the case of a purely buoyant flow ($G \neq 0$, $M = 0$), where $G = R/P$ is the Grashof number, convective cells of both signs which occupy a large part of the area, are developed in the top layer. In the case of a purely thermocapillary flow ($M \neq 0$, $G = 0$), the waves are developed simultaneously in both layers. All the vortices are positive in the top layer and negative in the bottom layer. Under the combined action of the buoyancy and the thermocapillary effect, the wavy motion takes place mainly in the top layer. One observes an essential asymmetry between the positive vortices, which occupy a large area in the top layer, and a rather compact negative vortices localized near the upper rigid plane.

The theoretical predictions obtained for infinite layers, cannot be directly applied to flows in closed cavities, because of several reasons. First, in the case of periodic boundary conditions one observes waves generated by a *convective* instability of parallel flow, while for the observation of waves in a closed cavity a *global* instability is needed [7]. Also, it should be taken into account, that in the presence of rigid lateral walls the basic flow is not parallel anymore. The lateral walls act as a stationary finite-amplitude perturbation that can produce a *steady* multicellular flow in the part of the cavity or in the whole cavity [7], [9], [18].

Nonlinear simulations have been carried out for the same system of fluids in a finite region with $L = 16$. For sufficiently small values of M and G, a steady flow is observed, which contains one cell in each fluid layer. With the increase of M and G, an additional maximum of the stream function field appears in each layer near the hot end. A *subcritical* oscillatory instability of the steady flow generates an unsteady multicellular structure.

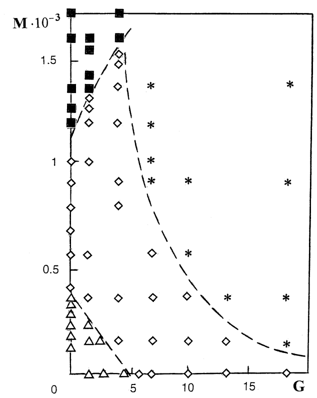

Figure 2: The diagram of flow regimes for $a = 1$. △ Parallel flow, ◊ traveling wave moving to the left, □ traveling wave moving to the right, $*$ pulsating wave moving to the left.

4 Two-layer ultra-thin films

In the case of very thin (but still macroscopic) films, when the film thickness is less than about 100 nm, a new physical phenomenon has to be incorporated into the model. It is necessary to take into account the *long-range intermolecular forces* acting between molecules of the liquid and substrate [19], [20]. It is essential that these forces act on distances *large* relative to interatomic distances. Hence, despite their microscopic (quantum) origin, they can be incorporated into a macroscopic theory. In the framework of the continuum approach, the intermolecular forces manifest themselves as 'surface forces' or *'disjoining pressure'* $\Pi(h) = df(h)/dh$ (see, e.g., [21]), which can be considered as a certain external normal stress imposed on the free surface. The sign of the disjoining pressure can be either positive or negative.

If the film is formed from an apolar fluid, the only relevant kind of long-range intermolecular interaction is van-der-Waals interaction $U(r) \sim 1/r^6$. In that case, the disjoining pressure can be taken as

$$\Pi(h) = A/6\pi h^3, \tag{11}$$

where A is the dimensionless *Hamaker constant* [19], [22].

A theoretical description of two-layer ultra-thin films has been developed in [23]-[26]. The system involves two deformable interfaces $z = H_1(\mathbf{r}, t)$ (liquid-liquid) and $z = H_2(\mathbf{r}, t)$ (liquid-gas). One has to take into account *two disjoining pressures* $\Pi_1(H_1, H_2)$ and $\Pi_2(H_1, H_2)$, applied at each interface:

$$\text{at } z = H_1,\ P_1 - P_2 = -\sigma_1 \nabla^2 H_1 + \Pi_1(H_1, H_2);$$

$$\text{at } z = H_2,\ P_2 - P_0 = -\sigma_2 \nabla^2 H_2 + \Pi_2(H_1, H_2),$$

where σ_1 and σ_2 stand for interfacial tensions at the corresponding interfaces, and P_0 is the atmospheric pressure.

The total potential energy of the van der Waals interactions $\Psi = \Psi_1 + \Psi_2 + \Psi_{12}$ includes the energy Ψ_1 of the substrate interaction with layer 2 across layer 1, the energy Ψ_2 of the gas phase interaction with layer 1 across layer 2, and the energy Ψ_{12} of the interaction between the gas phase and the substrate across two layers [19], [26]

$$\Psi(H_1, H_2) = -\frac{A_{sg} - A_{s1} - A_{g2}}{12\pi H_1^2} - \frac{A_{s1}}{12\pi H_2^2} - \frac{A_{g2}}{12\pi(H_1 - H_2)^2}, \tag{12}$$

where A_{sg}, A_{s1} and A_{g2} are Hamaker constants characterizing the interactions between the solid substrate and the gas across the two layers, between the solid substrate and liquid 1 across liquid 2, and between the gas phase and liquid 2 across liquid 1, correspondingly. The disjoining pressures are computed as

$$\Pi_j(H_1, H_2) = \frac{\partial \Psi}{\partial H_j},\ j = 1, 2. \tag{13}$$

Finally, the dimensionless equations governing the evolution of layers thicknesses can be written as [26]

$$H_{1T} + \nabla \cdot \mathbf{Q}_1 = 0,\ H_{2T} + \nabla \cdot \mathbf{Q}_2 = 0, \tag{14}$$

where the fluxes $\mathbf{Q}_1$ and $\mathbf{Q}_2$ include the contribution of the flow generated by pressure gradients and that of the thermocapillary flow:

$$\mathbf{Q}_1 = f_{11}\nabla P_1 + f_{12}\nabla P_2,\ \mathbf{Q}_2 = f_{21}\nabla P_1 + f_{22}\nabla P_2. \tag{15}$$

The mobility functions are:

$$f_{11} = -\frac{1}{3\eta_1}H_1^3;\ f_{12} = -\frac{1}{2\eta_1}H_1^2(H_2 - H_1);\ f_{21} = \frac{1}{6\eta_1}H_1^3 - \frac{1}{2\eta_1}H_1^2 H_2;$$

$$f_{22} = (H_2 - H_1)\left[H_1^2\left(\frac{1}{2\eta_1} - \frac{1}{3\eta_2}\right) + H_1 H_2\left(-\frac{1}{\eta_1} + \frac{2}{3\eta_2}\right) - \frac{1}{3\eta_2}H_2^2\right].$$

The pressures P_1 and P_2 include the disjoining pressures:

$$P_1 = -\sigma_1\nabla^2 H_1 - \sigma_2\nabla^2 H_2 + W_1(H_1, H_2), \tag{16}$$

$$P_2 = -\sigma_2\nabla^2 H_2 + W_2(H_1, H_2), \tag{17}$$

where

$$W_1(H_1, H_2) = \frac{A_{sg} - A_{s2} - A_{g1}}{6\pi H_2^3} + \frac{A_{s2}}{6\pi H_1^3}, \tag{18}$$

$$W_2(H_1, H_2) = \frac{A_{sg} - A_{s2} - A_{g1}}{6\pi H_2^3} + \frac{A_{g1}}{6\pi(H_2 - H_1)^3}. \tag{19}$$

The linear stability theory of a two-layer film reveals bending modes (in-phase deformations of interfaces), and squeezing modes (out-of-phase deformations of interfaces). Unexpectedly, gravity effects can be relevant for the dynamics of a two-liquid system (Fisher and Golovin, 2005). Depending on parameters, the time evolution may lead either to a blow-up film rupture ("swiss-cheese" dewetting pattern), or to formation of coarsening drops.

Merkt *et al.* [27] investigated a two-layer system sandwitched between two rigid plates, with one liquid-liquid interface. It turns out that the dynamics of such systems cannot be described in a local way by the interface profile $h(\mathbf{r})$, but needs for its full description also the stream function $f\mathbf{r})$ of the horizontal mean flow.

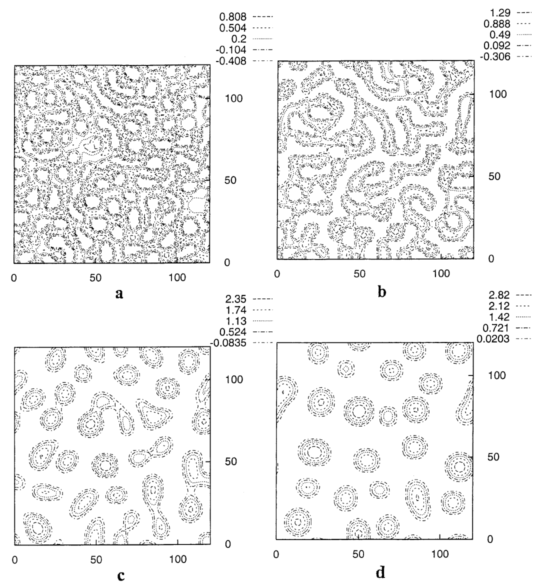

Figure 3: Isolines of h_1 at different moments of time: (a) $t = 240$; (b) $t = 330$; (c) $t = 500$; (d) $t = 800$; $M = 0$. The mean values $\langle h_1 \rangle = 1.2$; $\langle h_2 \rangle = 1$.

Below we consider the decomposition of a *laterally heated* two-layer film caused by the intermolecular forces. Longwave equations which incorporate both the thermocapillary flows and the influence of the van der Waals interactions have been derived. These equations contain additional term in the expressions for fluxes,

$$\mathbf{Q}_1 = F_{11}\nabla P_1 + F_{12}\nabla P_2 + Q_1^T \mathbf{e}_x, \; \mathbf{Q}_2 = F_{21}\nabla P_1 + F_{22}\nabla P_2 + Q_2^T \mathbf{e}_x, \quad (20)$$

where $\mathbf{e}_x$ is the unit vector of the axis x,

$$Q_1^T = -\frac{(\alpha_1 + \alpha_2)A}{2\eta_1} H_1^2; \quad (21)$$

$$Q_2^T = -\frac{\alpha_2 A}{2\eta_2}(H_2 - H_1)^2 - \frac{\alpha_1 + \alpha_2}{2\eta_1} A H_1 (2H_2 - H_1); \quad (22)$$

A is a constant temperature gradient imposed in the direction of the axis x at the substrate. Expressions (21) and (22) provide a generalization of a similar expression known for a one-layer film [28].

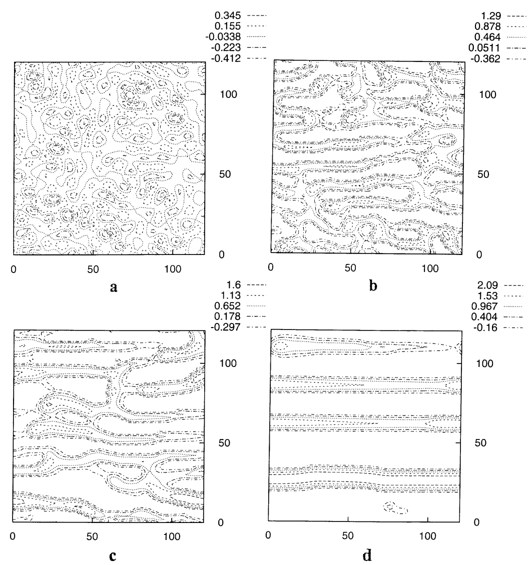

Figure 4: Isolines of h_1 at different moments of time: (a) $t = 200$; (b) $t = 330$; (c) $t = 440$; (d) $t = 2000$; $M = 0.35$. The mean values $\langle h_1 \rangle = 1.2$; $\langle h_2 \rangle = 1$.

In the absence of a lateral heating, droplets are developed on the background of a thin film. A slow coarsening of the droplets system takes place due to the droplets coalescence and Ostwald ripening (see Fig. 3). In the presence of a lateral heating, the droplets of different sizes are drifted with different velocities. This circumstance leads to a fast anisotropic coalescence of droplets and formation of “rivulets” oriented along the direction of the thermocapillary force (see Fig. 4). The last phenomenon resembles processes observed in simulations of the driven Cahn-Hilliard equation [29].

5 CONCLUSIONS

We have performed nonlinear simulations of wavy convective regimes that developed under the action of a temperature gradient along the interface. The diagram of regimes for buoyant-thermocapillary convection has beem obtained. Different scenarios of the decomposition of ultra-thin two-layer films into droplets have been revealed, and the droplets dynamics has been studied.

References

[1] Smith M K and Davis S H 1983 Instabilities of dynamic thermocapillary liquid layers Part 1 Convective instabilities *J. Fluid Mech* **132** 119-44

[2] Smith M K and Davis S H 1983 Instabilities of dynamic thermocapillary liquid layers Part 2 Surface-wave instabilities *J. Fluid Mech.* **132** 145-62

[3] Smith M K 1986 Instability mechanisms in dynamic thermocapillary liquid layers *Phys. Fluids* **29** 3182-86

[4] Davis S H 1987 Thermocapillary instabilities *Annu. Rev. Fluid Mech.* **19** 403-35

[5] Parmentier P M, Regnier V C and Lebon G 1993 Buoyant-thermocapillary instabilities in medium-Prandtl-number fluid layers subject to a horizontal temperature gradient *Int. J. Heat Mass Transfer* **36** 2417-27

[6] Mercier J F and Normand C 1996 Buoyant-thermocapillary instabilities of differentially heated liquid layers *Phys. Fluids* **8** 1433-45

[7] Priede J and Gerbeth G 1997 Convective, absolute, and global instabilities of thermocapillary-buoyancy convection in extended layers *Phys. Rev. E* **56**, 4187-99

[8] Perez-Garcia C, Madruga S, Echebarria B, Lebon G and Burguete J 2004 Hydrothermal waves and corotating rolls in laterally heated convection in simple liquids *J. Non-Equilib. Thermodyn.* **29** 377-88

[9] Riley R J and Neitzel G P 1998 Instability of thermocapillary-buoyancy convection in shallow layers Part 1 Characterization of steady and oscillatory instabilities *J. Fluid Mech.* **359** 143-64

[10] Pelacho M A, Garcimartin A and Burguete J 2000 Local Marangoni number at the onset of hydrothermal waves *Phys. Rev. E* **62** 477-83

[11] Burguete J, Mukolobwiez N, Daviaud F, Garnier N and Chiffaudel A 2001 Buoyant-thermocapillary instabilities in extended liquid layers subjected to a horizontal temperature gradient *Phys. Fluids* **13**, 2773-87

[12] Ezersky A B, Garcimartin A, Mancini H L and Perez-Garcia C 1993 Spatiotemporal structure of hydrothermal waves in Marangoni convection *Phys. Rev. E* **48** 4414-22

[13] Doi T and Koster J N 1993 Thermocapillary convection in two immiscible liquid layers with free surface *Phys. Fluids* **5**, 1914-27

[14] Nepomnyashchy A A, Simanovskii I B and Braverman L M 2001 Stability of thermocapillary flows with inclined temperature gradient *J. Fluid Mech.* **442**, 141-55

[15] Madruga S, Perez-Garcia C and Lebon G 2003 Convective instabilities in two superposed horizontal liquid layers heated laterally *Phys. Rev. E* **68** 041607

[16] Madruga S, Perez-Garcia C and Lebon G 2004 Instabilities in two-liquid layers subject to a horizontal temperature gradient *Theor. Comput. Fluid Dyn.* **18** 277-84

[17] Nepomnyashchy A A and Simanovskii I B 2006 Convective flow in a two-layer system with a temperature gradient along the interface *Phys. Fluids* **13** 032105

[18] Shevtsova V M, Nepomnyashchy A A and Legros J C 2003 Thermocapillary-buoyancy convection in a shallow cavity heated from the side *Phys. Rev. E* **67** 066308

[19] Israelachvili J 1992 *Intermolecular and Surface Forces* (London: Academic Press)

[20] Derjaguin B V, Muller V M and Churaev N V 1987 *Surface Forces* (New York: Consultants Bureau)

[21] Williams M B and Davis S H 1982 Nonlinear theory of film rupture *J. Colloid Interface Sci* **90** 220-8

[22] de Gennes P-G, Brochard-Wyart F and Quéré D 2002 *Capillarity and Wetting Phenomena* (New York: Springer)

[23] Pototsky A, Bestehorn M, Merkt D and Thiele U 2004 Alternative pathways of dewetting for a thin liquid two-layer film *Phys. Rev. E* **70** 025201(R)

[24] Pototsky A, Bestehorn M, Merkt D and Thiele U 2005 Morphology changes in the evolution of two-layer films *J. Chem. Phys.* **122** 224711

[25] Bandyopadhyay D, Gulabani R and Sharma A 2005 Instability and dynamics of thin liquid bilayers *Ind. Eng. Chem. Res.* **44** 1259-72.

[26] Fisher L S and Golovin A A 2005 Nonlinear stability analysis of a two-layer thin liquid film: Dewetting and autophobic behavior *J. Coll. Interface Sci.* **291** 515-28

[27] Merkt D, Pototsky A, Bestehorn M and Thiele U 2005 Long-wave theory of bounded two-layer films with a free liquid-liquid interface: Short- and long-time evolution *Phys. Fluids* **17** 064104

[28] Oron A, Davis S H and Bankoff S G 1997 Long-scale evolution of thin liquid films *Rev. Mod. Phys.* **69** 931-80

[29] Yeung C, Rogers T, Hernandez-Machado A and Jasnow D 1992 Phase separation dynamics in driven diffusive systems *J. Stat. Phys.* **66** 1071-88