JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 8, No. 3, June 2006, p. 1033 - 1036

Long wave instabilities in binary mixture thin liquid films

I. D. BORCIA[a,b*], R. BORCIA[a], M. BESTEHORN[a]
[a]*Lehrstuhl Statistische Physik / Nichtlineare Dynamik, Brandenburgische Technische Universität, Erich-Weiner-Strasse 1, 03046-Cottbus, Germany*
[b]*"Alexandru Ioan Cuza" University, Faculty of Physics, Department of Solid State & Theoretical Physics, Blvd. Carol I, 11, 700506, Iaşi, Romania*

An extension of lubrication approximation from pure liquid systems to binary mixtures is presented. The method permits to reduce the dimension of the equation system by one. To apply the model for binary mixtures an extra term related to the Soret effect is added to the thin film equation. This assures the coupling between the film height and the concentration field. An equation for the concentration is considered in order to describe the mass conservation. 3D fully non-linear simulations were done. According with the linear stability diagram one obtains monotonic or oscillatory instabilities, depending on the control parameters. We demonstrate the appearance of typical structures such as static or soliton like traveling drops.

(Received March 15, 2006; accepted May 18, 2006)

Keywords: Pattern formation, Self-organization, Binary mixtures, Soret effect

1. Introduction

Thin liquid mixture films, most notable polymeric, lying on a solid flat substrate have widespread technological applications like coating and wetting processes. Consequently the interest in the study of instabilities occurring in such mixtures is increasing from both experimental [1-3], and numerical [4-6] point of view. For thick liquid layers an approach using Navier-Stokes, heat and mass conservation equations coupled by adequate boundary conditions is suitable [4]. The upper film surface is considered nondeformable and usually 2D simulations are performed in this case, 3D simulations being time-consuming. In order to allow free upper film interface and to simplify the mathematical model appears the idea to consider for these thin films an approximation similar to the lubrication approximation [7] which includes the Soret effect. In this case, one eliminates the velocity field from the thin film equation, and one reduces the dimension of the problem by one. Thus 3D simulations become reasonable because in fact it rests to solve numerically a 2D problem.

In the present paper we consider the case of a thin liquid mixture layer heated from below (Fig. 1). Our aim is to use the lubrication approximation for describing this system. Simulations using such a model were performed also for systems of two stacked ultrathin layers of different immiscible liquids on a solid substrate [8]. The results show interesting zigzag and varicose modes, but no oscillatory behavior was found. We will show that in our case the Soret effect is able to trigger such an oscillatory behavior.

For many liquid mixtures the surface tension depends on temperature and on concentration [9,10]. If a small inhomogeneity appears somewhere along the liquid surface, a temperature difference between the high and low regions appears. This leads to a mass flux due to the surface tension gradient. Due to the Soret effect a concentration gradient is observed which will induce in turn a mass flux. The interesting case is when the flux due to the temperature gradient destabilizes the system. This case was studied for pure liquids and it was found that only monotonic instabilities can develop. Depending on the system parameters, the flux due to the concentration gradient can have a destabilizing or a stabilizing effect. The first case corresponds to a positive value of the Marangoni separation ratio [4] and don't differ drastically from the case of pure fluids. The same behavior is found for small negative value of the separation ratio. For higher negative values the strength of the second flux is high enough to generate an oscillatory behavior of the system.

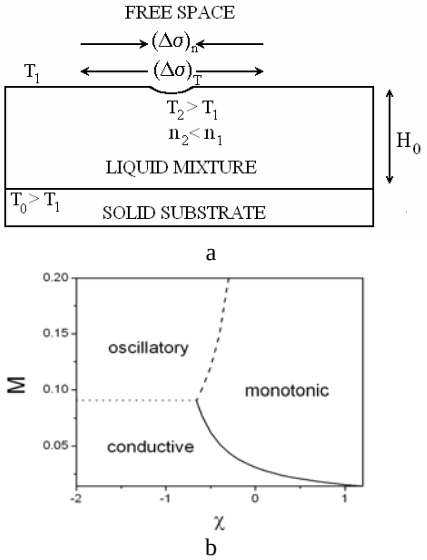

Fig. 1. (a) System sketch: on a solid substrate lies a binary mixture thin liquid film with free upper surface. A temperature gradient $T_0>T_1$ is maintained between the lower and upper surfaces; (b) stability diagram for the considered system computed from the linear problem.

2. Model

We derive the film equation system taking as starting point the thin film equation from the lubrication approximation [7]:

$$\partial_t H = -\frac{1}{\mu}\nabla_2 \cdot \left[\frac{1}{2}H^2\nabla_2\sigma - \frac{1}{3}H^3\nabla_2\left(\Theta|_{Z=H} - \sigma\nabla_2{}^2 H - \Pi\right)\right] \quad (1)$$

Where $\nabla_2 = \partial_x\vec{i} + \partial_y\vec{j}$ denotes the horizontal gradient, H is the liquid height, μ - the fluid dynamic viscosity, σ - the surface tension, Θ - the potential of the conservative body force and Π the external force. In our case we consider that the conservative force is the gravitational one and the external force is caused by the long-range intermolecular interactions between the flat solid substrate and the liquid surface. In this case one can take $\Theta|_{z=h} = \rho g H$ and $\Pi = A'/\left(6\pi H^3\right)$ [7]. A' is the Hamaker constant and is negative for a positive disjoining pressure (interfaces repel each other). One also supposes that the surface tension is a linear function on the temperature and concentration:

$$d\sigma = -\sigma_T dT - \sigma_N dN \quad (2)$$

where usually $\sigma_T > 0$ and the sign of σ_N depends on the components of the mixture. The term $\nabla_2\sigma$ from equation (1) will be replaced by $-\sigma_T\nabla_2 T - \sigma_N\nabla_2 N$. This assures in the thin film equation the coupling between the film height and the concentration field. One introduces the concentration as a new variable and adds a supplementary equation, the mass conservation for the dissolved component. We use the linear relation [11]:

$$\partial_t N = D(\Delta N + \beta_N/\beta_T\,\Delta T) \quad (3)$$

where $\beta_T = T_1 - T_0$ and $\beta_N = N_1 - N_0$ are the temperature and concentration gradients in the non-disturbed state and D is the diffusion coefficient. One considers the following no dimensional variables:

$$t' = \frac{\sigma}{3\mu H_0}t,\ (x,y,h) = \frac{1}{H_0}(X,Y,H),\ n = \frac{N-N_0}{\beta_N} \quad (4)$$

and one assumes that the temperature is a linear function on the vertical coordinate $T = T_0 + \beta_T h$. The concentration gradient induced by the Soret effect is an almost linear function on z, so that we can drop the second derivative in vertical direction. Therefore one replaces Δ by Δ_2 in the relation (3).

The equation system after dropping primes for time and indices "2" at Δ and ∇ becomes:

$$\begin{cases} \partial_t h = -\nabla\left[Mh^2\left(\nabla h + \chi\nabla n\right) + h^3\nabla\left(\Delta h - Gh - \frac{A}{h^3}\right)\right] \\ \partial_t n = L\left(\Delta n - \Delta h\right) \end{cases} \quad (5)$$

After rescaling appear in the above equation system the following nondimensional parameters: $M = -\frac{3}{2}\frac{\sigma_T}{\sigma}\beta$ - rescaled Marangoni number, $\chi = \left(\sigma_N\beta_N\right)/\left(\sigma_T\beta_T\right)$ - Maragoni separation ratio, $G = \rho g H_0^2/\sigma$ - rescaled Galileo number, $A = A'/\left(6\pi H_0^2\sigma\right)$ - rescaled Hamaker constant, and $L = 3\mu D/H_0\sigma$ - rescaled Lewis number.

3. Numerical results

We begin our study by performing a linear stability analysis of the problem taking all the perturbations of the form $\exp(\lambda t)\exp(-i\vec{k}\cdot\vec{r})$. Fig. 1b shows the results for $L = 0.03$, $A = -0.02$, $G = 0.0$. One can see, there are three different behaviors in the early development phase: for small values of the modified Marangoni number an instability can not develop and a conductive state of the system is acquired (Fig. 3a), for positive or small negative values of χ a monotonic instability is developed (Fig. 3b). In the case of higher Marangoni numbers and big enough negative separation ratios oscillatory instabilities develop (Fig. 3c).

For the fully nonlinear simulations we have considered a 3D system with periodical boundary conditions. In fact we compute h(x,y) and n(x,y) only at the liquid surface, so that we will have to solve numerically a 2D problem. We consider a mesh of 128 × 128 points in the xOy plane and we use a semi-implicit method: the linear part is computed using fast Fourier transforms and, in order to increase the speed of the algorithm, the non-linear part is computed using appropriate expressions for the derivatives in the finite differences method. All three behaviors from the linear stability analysis are found also in the non-linear simulations. We have performed simulations corresponding to the following points from Fig. 1: $M = 0.08$, $\chi = -1$ - conductive case, $M = 0.3$, $\chi = -0.01$ - monotonic case, $M = 0.3$, $\chi = -1$ - oscillatory case. Fig. 2 shows the real part of the growth rate $\mathrm{Re}(\lambda)$ as function of the wave length, computed for the linear problem. We have to mention that in the regions where the two solutions are superposed one has also an imaginary part of the growth rate. If there exists a positive real part, an instability can develop for the corresponding wavelength in the early state. If to this positive region corresponds non zero values of the imaginary part then the instability is oscillatory. Otherwise one has a monotonical instability.

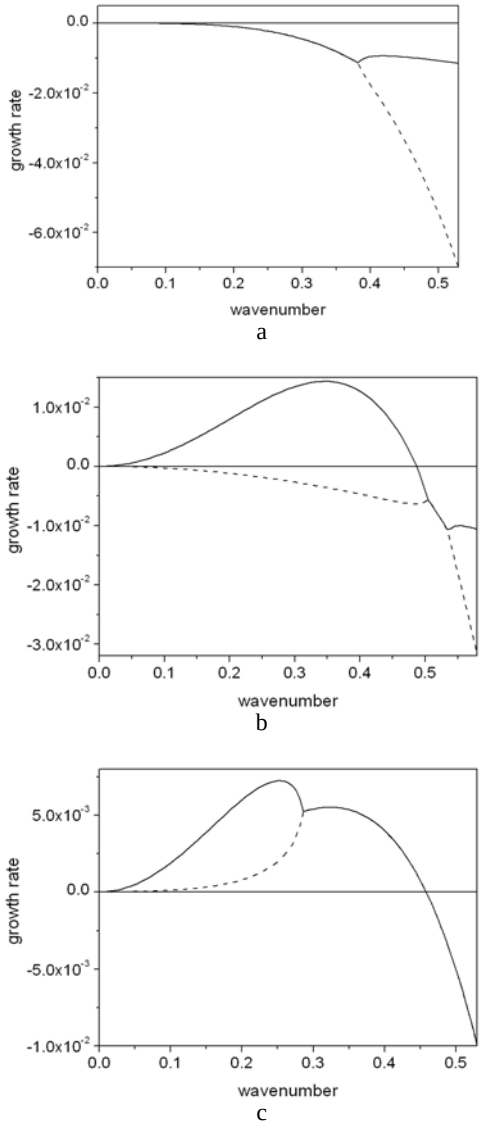

Fig. 2. Growth rate versus wave number computed in the linear approximation with different parameters values for three cases: (a) conductive state; (b) monotonic instability; (c) oscillatory instability.

The nonlinear simulations show in these cases the same behavior as predicted by the linear investigation. For all the parameter values corresponding to points on the conductive region from Fig. 1 all initial small perturbations are very fast suppressed. For the case corresponding to Fig. 2b one obtains at the beginning an amplification of the perturbations with the wavelength corresponding to the maximum of the growth rate. Afterwards a coarsening process appears and in the final state of the system turns up a single static drop.

More interesting is the case of oscillatory instability. Snapshots from 3D simulations are presented in Fig. 3. The gray value of the regions is proportional with the local film weight. The concentration is shown by mean of contour lines. The initial state is a uniformly distributed layer. In the early stage a number of drops are rapidly formed (see Fig. 3a). The drops start to move in randomly directions. The drop movement is driven by the concentration field: the direction is from higher to lower concentration regions. In the mean time a coarsening is established and the number of drops is decreasing (Fig. 3b) until one single soliton like traveling drop rests in the system (Fig. 3c).

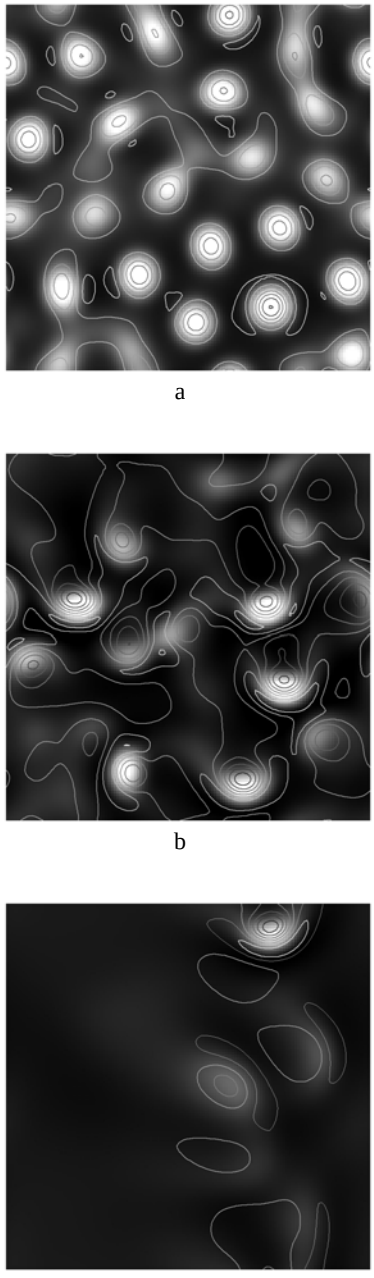

Fig. 3. Time series for the case of oscillatory behavior (same parameters as in Fig. 2c). White correspond to elevated regions of the free surface. Contour lines represent the concentration field. The drops are moving towards low values of the concentration (white lines). For exemplification, the final drop near the up-right corner in figure (c) is moving downwards.

4. Conclusions

In summary, we developed a model based on the lubrication approximation for very thin liquid binary mixtures with Soret effect. Linear analysis prediction for the system was confirmed by the 3D fully nonlinear simulations for a wide range of parameters values. We have found that the Soret effect can dramatically change the system behavior and can lead to oscillatory behavior for specifically parameters values. In the late stage of the temporal evolution static drops or traveling soliton like drops are obtained.

Acknowledgement

I. D. B. acknowledges funding from the project "Modellierung, Optimierung, Simulation komplexer Systeme", Brandenburg, Germany. R.B. thanks for partial financial support from the ESA-MAP AO-99-110 CIMEX project (Convection and Interfacial Mass Exchange).

References

[1] R. W. Walden, P. Kolonder, A. Passner, C. M. Surko, Phys. Rev. Lett. **55**(5), 496 (1985).
[2] K. Lerman, D. S. Cannell, G. Ahlers, Phys. Rev. E **59**(3), 2975 (1999).
[3] R. Kita, R. Wiegand, J. Luettmer-Strathmann, J. Chem. Phys. **121**(8), 3874 (2004).
[4] M. Bestehorn, P. Colinet, Phys. D **145**, 84(2000).
[5] A. Bergeon, D. Henry, H. BenHadid, L. S. Tuckerman, J. Fluid Mech. **375**, 143 (1998).
[6] I. Mercader, A. Alonso, O. Batiste, Eur. Phys. J. E **15**, 311 (2004).
[7] A. Oron, S. Davis, S. Bankoff, Rev. Mod. Phys. **69**, 931 (1997).
[8] A. Pototsky, M. Bestehorn, Phys. Rev. E **70**, 025201-1 (2004).
[9] M. Z. Saghir, C. G. Jiang, S. O. Derawi, E. H. Stenby, M. Kawaji, Eur. Phys. J. **15**, 241 (2004).
[10] B. A. Dilmohamud, J. Seeneevassen, S. D. D. V. Rughooputh, P. Ramasami., Eur. J. Phys. **26**, 1079 (2005).
[11] J. K. Platten, J. C. Legros, Convection in Liquids (Springer-Verlag, 1984).

*Corresponding author: iborcia@physik.ti-cottbus.de