

**EVOLUTION EQUATION FOR NONLINEAR
LONG-WAVELENGTH MONOTONIC MARANGONI
INSTABILITY IN A BINARY LIQUID LAYER WITH
NONLINEAR Soret EFFECT***

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ABSTRACT. The Soret effect in binary systems is called nonlinear when the thermo-diffusive flux is proportional to the temperature gradient with a coefficient being linear function of the concentration of one of the solute components. This effect is significant in highly dilute solutions. The long-wavelength Marangoni instability in a thin layer of binary liquid, in the presence of the nonlinear Soret effect, is considered. The nonlinear dynamic behaviour of the liquid system is studied in the case of *monotonic* instability. The solution of the dimensionless equations of mass and momentum balances, heat transfer and mass diffusion is searched near the linear instability threshold, in the form of series in a small parameter that measures the supercriticality. An equation for spatiotemporal evolution of the liquid system is derived based on the first two approximations.

KEY WORDS: Binary liquid, nonlinear Soret effect, long-wavelength Marangoni instability, nonlinear stability analysis, evolution equation.

1. Introduction

When gas or liquid mixtures are subjected to temperature gradient, solute gradients can be established spontaneously in an initially uniform concentration field via thermodiffusion. The phenomenon (named also Soret effect)

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is associated with various transport processes encountered in technology and nature. Intensive theoretical and experimental investigations of binary-fluid convection have been motivated by its importance to separation processes, terrestrial material processing, crystal growth, groundwater pollutant migration, etc. (see, for instance, [1], [2]). The references of some works related to the present study of the Soret effect on the Marangoni instability are listed in [3] and [4]. In the case of nonlinear Soret effect, the linear stability analysis for a binary liquid layer is performed in [5], [6] and [7]. The onset of linear monotonic and oscillatory instabilities in the binary liquid with respect to long-wavelength disturbances is studied in our papers [3] and [4].

According to the theory of non-equilibrium thermodynamics [8], the heat flux in binary mixtures is given by Fourier's law, whereas the total mass flux is a sum of the fluxes driven by concentration and temperature gradients,

$$(1) \quad \mathbf{J}_Q^* = -k \nabla T^*, \quad \mathbf{J}_C^* = -\rho D_C \nabla c^* - \rho D_T c^* (1 - c^*) \nabla T^*,$$

where T^* is the mixture temperature, k is the thermal conductivity of the mixture, ρ is the mixture mass density, c^* is the concentration of one of the mixture components, and D_C , D_T are the isothermal mass diffusion and thermodiffusion coefficients of the solute, respectively. In some binary liquid mixtures, especially in dilute ones, the heat flux and the total mass flux can be presented by [5]:

$$(2) \quad \mathbf{J}_C^* = -\rho D_C [\nabla c^* + S_T (\beta_0 + \beta_1 c^*) \nabla T^*],$$

where the ratio $S_T = D_T/D_C$ is called the Soret coefficient, and β_0 and β_1 are constants. For highly dilute solutions, $\beta_0 = 0$ and $\beta_1 = 1$. If $\beta_1 = 0$ and $\beta_0 = c_r^*(1 - c_r^*)$, where c_r^* is some reference value, one recovers the case of linear Soret effect. In the present study, we assume $\beta_0 > 0$ and β_1 to be either positive or negative. Eqn. (2) for the mass flux corresponds to the case of *nonlinear* Soret effect.

The aim of the present paper is to develop a *nonlinear* analysis of the monotonic Marangoni instability with respect to long-wavelength disturbances and to derive a spatiotemporal evolution equation for the convective pattern.

2. Formulation of the problem and governing equations

The formulation of the problem is fully given in [3] and [4] and here we will recall it shortly. We consider a thin two-dimensional horizontal layer of an incompressible viscous binary liquid, bounded from below by a rigid

plate and opened to the ambient gas phase. It is subjected to a constant vertical temperature gradient $-\beta$, with positive β corresponding to heating from below. On the liquid free surface, assumed nondeformable, Newton's cooling law takes place. Buoyancy is neglected in comparison with the Marangoni effect. At the free interface, surface tension σ depends linearly on temperature and concentration:

$$(3) \quad \sigma = \sigma_0 - \alpha_T(T^* - T_0^*) + \alpha_C(c^* - c_0^*), \quad \alpha_T = \left(-\frac{\partial \sigma}{\partial T^*} \right)_0, \quad \alpha_C = \left(\frac{\partial \sigma}{\partial c^*} \right)_0,$$

where subscript (0) denotes the reference state of the system, and the coefficient $\alpha_T > 0$ whereas α_C is either positive or negative. Both boundaries of the layer are considered poorly heat conducting and impermeable for the solute.

The governing equations of the dynamic behaviour of the layer are the equations of mass and momentum balances, heat transfer and solute diffusion written in dimensionless form:

$$(4) \quad \begin{aligned} \nabla \cdot \mathbf{v} &= 0, \\ \frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} &= -\nabla p + \text{Pr} \nabla^2 \mathbf{v}, \\ \frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T &= \nabla^2 T, \\ \frac{1}{Le} \left(\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c \right) &= \nabla^2 c + \chi \nabla^2 T + S_1 (\nabla c \cdot \nabla T + c \nabla^2 T), \end{aligned}$$

with boundary conditions:

$$(5) \quad \mathbf{v} = 0, \quad \frac{\partial T}{\partial z} = -1, \quad \frac{\partial c}{\partial z} = \chi + S_1 c_w \equiv \psi \quad \text{at} \quad z = 0;$$

$$(6) \quad \begin{aligned} w = 0, \quad \frac{\partial T}{\partial z} + Bi T &= 0, \quad \frac{\partial c}{\partial z} - Bi (\chi T + S_1 c) = 0, \\ \frac{\partial \mathbf{u}}{\partial z} + M \nabla_S (T - c) &= 0 \quad \text{at} \quad z = 1. \end{aligned}$$

Last equation in (6) represents the tangential stress balance. Here, $\mathbf{v}(\mathbf{u}, w)$ is the fluid velocity, p is the pressure, $M = \frac{\alpha_T \beta d^2}{\mu \kappa}$ is the Marangoni number, $\text{Pr} = \frac{\nu}{\kappa}$ is the Prandtl number, $Le = \frac{D_C}{\kappa}$ is the Lewis number, $Bi =$

$\frac{hd}{k}$ is the Biot number, $\chi = \frac{\beta_0 S_T \alpha_C}{\alpha_T}$ is the linear Soret number, $S_1 = S_T \beta \beta_1 d$ is the so-called nonlinear Soret number, $\psi = \chi + S_1 c_w$ is the Marangoni separation factor, $c_w = \frac{\alpha_C c_w^*}{\alpha_T \beta d}$ is dimensionless concentration on the layer bottom, κ is the thermal diffusivity, μ is the dynamic viscosity, $\nu = \mu/\rho$ is the kinematic viscosity, d is the layer depth and ∇_S^2 is two-dimensional Laplace operator. The Lewis number for liquid binary mixtures is small being of the order of 10^{-2} . The typical value of the linear Soret number varies on the range of $-1 \leq \chi \leq 1$ [3], [9]. The Biot number and the absolute values of the nonlinear Soret number are small, but the order of their values will be shown below. The Prandtl number is assumed large which is typical for many binary liquids. The Marangoni number is positive for heating from below ($\beta > 0$).

In the equilibrium state, the liquid is at rest and the steady-state temperature $T_{st}(z)$ and concentration $c_{st}(z)$ are given by:

$$(7) \quad \begin{aligned} T_{st}(z) &= -z + \frac{1 + Bi}{Bi}, \\ c_{st}(z) &= \left(c_w + \frac{\chi}{S_1} \right) \exp(S_1 z) - \frac{\chi}{S_1} = c_w(1 + S_1 z) + \chi z + \dots \end{aligned}$$

Linearization of Eqs (4)–(6) around the base state yields the proper equations and boundary equations used for studying the instability of the layer [3].

3. Nonlinear long-wavelength instability

Applying the approach used by Oron and Nepomnyashchy [9], we study the emergence of the nonlinear long-wavelength instability in a binary liquid layer with poorly conducting boundaries. This means that the wave number a and the Biot number are asymptotically small. Considering the absolute value of the nonlinear Soret number, $|S_1|$, also small, we introduce the following scaling of the parameters [9], [3]:

$$(8) \quad Bi = \varepsilon^4 B, \quad S_1 = \varepsilon^2 S, \quad a = \varepsilon K, \quad M = M_0 + M_2 \varepsilon^2 + \dots,$$

where ε is a small parameter measuring the supercriticality and M_0 is the critical Marangoni number for the linear instability. The analysis shows that the instability threshold and the condition for the instability to be monotonic are [9], [3]:

$$(9) \quad M_0 = \frac{48}{1 + \chi(1 + Le^{-1})}, \quad \chi > \chi_2 \equiv -\frac{1}{1 + Le^{-1} + Le^{-2}}.$$

The critical Marangoni number is positive if $\chi > \chi_1 \equiv -\frac{1}{1 + Le^{-1}}$. As $\chi_1 < \chi_2 < 0$ for any (positive) Lewis number, in what follows the linear Soret number varies in the interval $\chi_2 < \chi \leq 1$.

To study the nonlinear instability of the layer, the bifurcation analysis is applied using the technique of asymptotic expansions. We introduce the rescaled spatial and temporal variables [9]:

$$(10) \quad X = \varepsilon x, \quad Y = \varepsilon y, \quad Z = z, \quad \tau = \varepsilon^4 t,$$

and appropriate expansions for the velocity, temperature, concentration and pressure fields:

$$(11) \quad \begin{aligned} \mathbf{u} &= \varepsilon \mathbf{U}(X, Y, Z, \tau) = \varepsilon (\mathbf{U}_0 + \varepsilon^2 \mathbf{U}_2 + \varepsilon^4 \mathbf{U}_4 + \dots), \\ w &= \varepsilon^2 W(X, Y, Z, \tau) = \varepsilon^2 (W_0 + W_2 \varepsilon^2 + W_4 \varepsilon^4 + \dots), \\ T &= \Theta(X, Y, Z, \tau) = \Theta_0 + \Theta_2 \varepsilon^2 + \Theta_4 \varepsilon^4 + \dots, \\ c &= \Sigma(X, Y, Z, \tau) = \Sigma_0 + \Sigma_2 \varepsilon^2 + \Sigma_4 \varepsilon^4 + \dots, \\ p &= \Pi(X, Y, Z, \tau) = \Pi_0 + \Pi_2 \varepsilon^2 + \Pi_4 \varepsilon^4 + \dots, \\ \Theta_0 &= F(X, Y, \tau), \quad \Sigma_0 = G(X, Y, \tau). \end{aligned}$$

The quantities (8), variables (10) and series (11) are substituted into Eqns (4)–(7) and a corresponding set of linear, but partial differential equations is derived. Integrating the resulting temperature and concentration equations across the layer, one obtains integral relations expressing the heat and solute conservation equations, which are considered as solvability conditions in our analysis. The set of equations is solved analytically up to the second order in ε , but due to the lack of space, the solutions are not presented here. Using the solvability conditions, we derived two equations for the functions $F(X, Y, \tau)$ and $G(X, Y, \tau)$ which, in the case of large times, can be reduced to one equation for F , namely:

$$(12) \quad \begin{aligned} \alpha F_\tau + B(1 + \chi)F + \tilde{\gamma}_1 \nabla_S^2 F + \gamma_2 \nabla_S^4 F + \gamma_3 \nabla_S \cdot (\nabla_S F \nabla_S^2 F) \\ + \gamma_4 \nabla_S^2 (|\nabla_S F|^2) - \gamma_5 \nabla_S \cdot (\nabla_S F |\nabla_S F|^2) + \gamma_6 \nabla_S^2 (F^2) = 0. \end{aligned}$$

The coefficients in (12) are as follows:

$$\alpha = 1 + \chi(1 + Le^{-1} + Le^{-2}), \quad m_0 = [1 + \chi(1 + Le^{-1})]^{-1},$$

$$\begin{aligned}
\gamma_1 &= \frac{1}{48} M_2 [1 + \chi (1 + Le^{-1})]^2 = \frac{M_2}{48m_0^2}, \\
\tilde{\gamma}_1 &= \gamma_1 + S \left\{ \chi \left(\frac{2}{5} + Le^{-1} \right) + (1 + Le^{-1}) [c_w + \chi (1 + Le^{-1}) \langle\langle F \rangle\rangle] \right\}, \\
\gamma_2 &= \frac{1}{15} [1 + \chi (1 + Le^{-1})] = \frac{1}{15m_0}, \\
\gamma_3 &= \frac{1}{5} \frac{1}{\text{Pr} m_0} + \frac{1}{10} [1 + \chi (1 + Le^{-1} + Le^{-2})], \\
(13) \quad \gamma_4 &= \frac{1}{10} \frac{1}{\text{Pr} m_0} + \frac{3}{5} [1 + \chi (1 + Le^{-1} + Le^{-2})], \\
\gamma_5 &= \frac{48}{35} [1 + \chi (1 + Le^{-1}) (1 + Le^{-2})], \\
\gamma_6 &= -\frac{S}{2} \chi (1 + Le^{-1})^2, \\
\nabla_S &\equiv \left(\frac{\partial}{\partial X}, \frac{\partial}{\partial Y} \right), \quad \nabla_S^2 = \frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2}, \\
\nabla_S^4 &= (\nabla_S^2)^2, \quad \langle\langle F \rangle\rangle \equiv \int_0^1 F d\eta.
\end{aligned}$$

Using the following transformations [9]:

$$\begin{aligned}
(14) \quad \tau &= c_1 \tau_1, \quad F = c_2 F, \quad (X, Y) = c_3 (\xi, \eta), \quad \nabla_S = \frac{1}{c_3} \nabla_1, \\
c_1 &= \frac{4\alpha\gamma_2}{\tilde{\gamma}_1^2}, \quad c_2 = \left(\frac{\gamma_2}{\gamma_5} \right)^{\frac{1}{2}}, \quad c_3 = \left(\frac{2\gamma_2}{\tilde{\gamma}_1} \right)^{\frac{1}{2}},
\end{aligned}$$

Eqn. (12) is rewritten in the final form:

$$\begin{aligned}
(15) \quad F_{\tau_1} + bF + 2\nabla_1^2 F + \nabla_1^4 F - \nabla_1 \cdot \left(\nabla_1 F |\nabla_1 F|^2 \right) \\
+ s_1 \nabla_1 \cdot (\nabla_1 F \nabla_1^2 F) + s_2 \nabla_1^2 (|\nabla_1 F|^2) + s_3 \nabla_1^2 (F^2) = 0,
\end{aligned}$$

where

$$(16) \quad b = \frac{4B(1+\chi)\gamma_2}{\tilde{\gamma}_1^2}, \quad s_1 = \frac{\gamma_3}{\sqrt{\gamma_2\gamma_5}}, \quad s_2 = \frac{\gamma_4}{\sqrt{\gamma_2\gamma_5}}, \quad s_3 = \frac{\gamma_6}{\sqrt{\gamma_2\gamma_5}}.$$

Eqn. (15) is the spatiotemporal evolution equation for the liquid system under consideration. The modified nonlinear Soret number S appears in the coefficients b and s_3 through the quantities $\tilde{\gamma}_1$ and γ_6 , respectively. For $S = 0$, i.e. for $\tilde{\gamma}_1 = \gamma_1$ and $\gamma_6 = 0$, the last term in (15) vanishes and one recovers the evolution equation for the case of linear Soret effect reported in [9]. The presence of the last term in Eqn. (15) may change substantially the conditions for the existence of roll, square and hexagonal patterns, compared to those for the linear Soret effect.

4. Conclusion

In the case of nonlinear Soret effect, an evolution equation is derived for the nonlinear dynamic behaviour of a horizontal binary liquid layer, confined between poorly conducting boundaries, when the fluid is heated from below. The layer depth is assumed sufficiently small to ignore the influence of buoyancy comparing to the Marangoni effect and to neglect the deformation of the liquid interface. The long-wavelength monotonic instability is studied.

In comparison with the analogous equation for the case of linear Soret effect, the new one has an additional term which may play an important role in determining the conditions for the emergence of different patterns. The study of the evolution equation is under progress and the results will be published in near future.

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